Nuclear Science in Europe
Impact, Applications, Interactions
JUNE 2002
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NuPECC REPORT*

JUNE 2002

NUCLEAR SCIENCE IN EUROPE:
IMPACT
APPLICATIONS
INTERACTIONS

Edited by:
Daniel Guerreau, Juha Äystö, Dominique Guillemaud-Mueller, and Gabriele-Elisabeth Körner

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Preamble

The rich phenomena of nature have always fascinated humankind. We have long been driven by an inherent curiosity to gain a deeper understanding of its underlying laws and mechanisms. This is the basis of science which has become an essential element of culture. At present it not only satisfies our thirst for knowledge but also leads to many benefits in our daily lives. This is particularly true for nuclear physics, which studies the properties and interactions of the basic building blocks of matter. Although most nuclear phenomena are far beyond our daily experience, there is a great variety of related techniques and applications which have considerable impact on society. However, in many cases even scientists are unaware of the benefits associated with the widespread use of nuclear techniques.

A comprehensive survey of the impact of nuclear physics was presented in *Impact and Applications of Nuclear Physics in Europe*, a report issued by the Nuclear Physics European Collaboration Committee (NuPECC) in 1994. It showed the wide spectrum of applications of nuclear techniques and the numerous interactions with different fields of science. Since the report’s publication the impact of nuclear science has progressed and new ideas have emerged, leading to significant developments of technological interest. Therefore, it is timely to repeat a similar exercise. Using the 1994 survey as a comprehensive basis, the present report focuses on three topics of general interest which have seen significant dynamic progress in recent years, delineating their developments and achievements.

Part 1 deals with nuclear physics contributions to the energy problem, one of the primary problems facing humankind at present. Here, important initiatives have been started to reduce nuclear waste, considered a major problem by society, which appears to be manageable by modern nuclear techniques. Significant progress is reported also in inertial-confinement fusion driven by heavy-ion or laser beams. Although these developments are still at a fundamental level, interesting applications may result from this research. Whichever strategy society adopts to solve the energy problem, research on transmutation and fusion will be of great importance for future developments.

Part 2 focuses on the interfaces between nuclear physics, biology and medicine. Intensified collaborations with biologists and doctors have led to the development of methods, instruments and facilities which, besides leading to a better understanding of biological processes, have had a large impact on our health-care systems. The contribution of nuclear physics is still growing and its great potential is visible in the given review.

Part 3 focuses on the strong interactions with related fields, namely atomic and condensed-matter physics. It exemplifies the cross-fertilization of excellent interdisciplinary collaboration and demonstrates the importance of a unified view of science where borders between fields are only weakly defined.

It is the main task of NuPECC as an expert committee of the European Science Foundation (ESF) to co-ordinate and promote nuclear physics in Europe. NuPECC provides updated information, presents highlights and suggests new directions and opportunities. More ambitious is the objective to propose a well-defined strategy for the harmonious and effective development of nuclear physics research in Europe. Such a process of broad reflection within the European scientific community has been started in 2002 with the aim of setting up a long range plan for nuclear physics in Europe.

In this spirit, the present report on the impact, applications and interactions of nuclear physics in Europe is offered to colleagues of the scientific community, science-policy makers and the general public. It gives an up-to-date view of the current developments in nuclear techniques.
which have a major impact on related fields and society. It is important to stress that, apart from the three topics extensively treated in the report, important progress has been achieved in numerous applications in other fields. Specifically, the strong interactions with environmental sciences, astrophysics, art and archaeology should be mentioned in this context.

The expertise required for broad surveys, such as the present report, obviously exceeds the expertise of NuPECC. Therefore, three working groups were established with specialists in the fields under review. Their reports were presented and discussed in a workshop, organized with the support of the European Union, held in Dourdan, France on 22 and 23 November 2001.

The present document contains the full contributions of all three groups. NuPECC wishes to express its gratitude to the members of the working groups and especially to the chairpersons of these groups: C. Cohen, G. Kraft and J.P. Schapira.

The reader who wishes to get an overview of the main content of the report may refer to the summaries at the beginning of each chapter. Some ‘Observations by NuPECC’ follow this preamble; they suggest a series of specific actions aiming at progress in the most promising nuclear physics techniques and at strengthening nuclear science in all its diversity, from fundamental research to the dedicated and most efficient development of applications.
Observations by NuPECC

The three topics presented in this report illustrate the continuously increasing contribution of nuclear physics to the demands of society and to technological developments. Furthermore, nuclear physics has interfaces with numerous fields. Maintaining a high level of involvement in basic sciences is a necessary condition for an efficient development of multidisciplinary activities. The impact — both direct and indirect — of basic research, is very often underestimated by scientists and the public alike. This is particularly true for nuclear physics, which makes an essential contribution to the welfare of our society. The energy involved in any nuclear phenomenon exceeds our everyday experience and — triggered by military use and accidents — has generated strong negative emotions towards the field of nuclear physics. Therefore, much effort must be made to improve our understanding of nuclear science and increase public awareness of its benefits for society.

The report clearly indicates the potential of nuclear physics for interdisciplinary research. This requires that open-minded scientists pay attention to the demands of society and communicate beyond the borders of their traditional fields. Support not only from its own community but also from the scientific community at large and from the outside world is an important prerequisite for the healthy development of any field of science. As an outcome of this report the following observations have been made by NuPECC.

- Many problems of modern society can be solved with the help of nuclear science. The nuclear physics community should address the challenges arising from the demands of society related to the field and should assume its responsibility in the search for solutions. At present the provision of energy and the management of high-level nuclear waste are of major concern for humankind. Whatever the future of nuclear energy is, nuclear physicists must play a role in finding innovative solutions, help decision-makers to identify valid options and contribute to rational public discussions of this issue. From the present standpoint, strong support for applied research associated with the development of new concepts such as hybrid reactors for the transmutation of nuclear wastes (e.g. accelerator-driven systems, ADS) appears necessary. A preliminary design study for such an ADS is an important step and should be considered in the forthcoming European framework programmes.

- The contribution of nuclear physics to medical science, biology and radiobiology is now well recognized and is an excellent example of specific know-how applied to another field. Indeed nuclear techniques applied to life sciences play a major role. They have made an essential contribution to recent progress via the use of radioactive tracers, accelerator mass spectroscopy (AMS), the employment of nuclear imaging techniques of various types (SPECT, PET, NMR, MicroPET), the study of radiation effects on biological material, and the development of dedicated accelerators and high-quality beams for proton and hadron therapy. In this context, there is an urgent need to forge further interdisciplinary collaborations and the activities of combined teams must be encouraged. Specifically, nuclear physicists should help medical doctors in the promotion and development of techniques associated with proton and ion-beam therapy.

- Even when dealing with basic problems, the interaction with other communities could be of mutual benefit as different fields very often share concepts, models and techniques. This report presents a very nice illustration of the cross-fertilization that can exist between nuclear science and atomic and condensed-matter physics. Good examples are the use and development of ion sources and atomic
traps. These communities are, next to nuclear physicists, the most important users of the large infrastructures oriented towards nuclear physics. Collaborations must be encouraged and further developed.

- Spin-offs of nuclear technology, namely accelerators and detectors, are of major importance and are quite often the link with other fields for basic research. These technological developments should be strongly supported and the discussion on future tools for nuclear science should systematically include the multidisciplinary aspects of the projects. The importance of technological developments has been recognized through many RTD contracts financed within the fifth European framework programme. However, a general assessment and better planning should be envisaged at the European level for an improved coordination of such developments.

- The widespread applications of nuclear techniques and their strong impact on society require education of the general public in the concept of elementary nuclear phenomena. This is needed to promote effective and balanced discussions of nuclear science issues that are of importance for society at large. Enhancing nuclear physics knowledge will help the understanding of science in general. Therefore, the basics of nuclear physics have to be taught in all secondary schools and the curricula at all universities should include nuclear physics courses. This, together with training of graduate students in nuclear science fields, will ensure that future facilities have experienced staff, and that expertise will be available to tackle problems and provide innovative solutions should the need arise in the future. At present these conditions are far from being achieved everywhere in all NuPECC countries. An increased visibility of applied nuclear science at universities could significantly help. In this context, the formation of small nuclear science research teams in many more universities is of prime importance. NuPECC alone has no direct influence to improve the present situation but, together with other institutions, it is ready to initiate discussions with the appropriate authorities.
PART 1: ENERGY

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1. Introduction

J.P. Schapira

Energy production throughout the world is today facing a number of issues related to the increasing energy needs foreseen for this century, especially in developing countries. Two important issues, i.e. resources preservation and environmental impact (especially related to climate change), can largely be addressed by a significant use of nuclear energy. Nuclear science and technology have a variety of applications in the field of nuclear (as well as conventional) energy and are therefore the basis for a large and safe nuclear contribution to energy production.

Discovered in the early days of nuclear physics, neutron-induced fission of certain heavy nuclei and fusion between two light elements are the basic nuclear processes responsible for the energy production in fission and fusion reactors (the latter being not presently at an industrial stage). Beside these, there are other applications of nuclear science and technology in non-nuclear energy fields. One example is the intensive use of nuclear detection and accelerator techniques in the field of petroleum prospecting. Another is environment and waste monitoring as a consequence of energy related activities.

As in the 1994 NuPECC report on the Applications of Nuclear Science in the field of Energy, here we concentrate on two essential subjects. One is the development of accelerator-driven systems (ADS), see Fig. 1.1, which seem to be best suited for the transmutation of some long-lived and highly radiotoxic radionuclides present inside spent nuclear fuels unloaded from a reactor. The second is related to the energy production by heavy-ion inertial confinement fusion (ICF), an alternative to the strongly supported magnetic confinement fusion (MCF) and a variant of laser-induced ICF.

Figure 1.1: Schematic view of an accelerator-driven system (ADS).

Why focus on these two subjects?

In the early days of nuclear energy development, neutronic and reactor physics were only one branch of nuclear science, just like nuclear and particle physics or radiochemistry. E. Fermi, E. Wigner and F. Joliot-Curie are distinguished examples of physicists who were able at that time to contribute profoundly to most of these branches. Since then, these branches have become more and more independent of each other because of their increased technical complexity and specialization. As a consequence, different and separate scientific and technical communities have emerged with their own culture and scientific approaches. Moreover, the nuclear energy community was, and still is, in charge of developing peaceful as well as military applications of nuclear energy and became therefore more and more linked to technical, industrial and strategic issues. Meanwhile, the academic world, i.e. universities, national and international research organizations (e.g. CERN at a European level), has been dealing almost exclusively with fundamental nuclear science and related techniques such as accelerators.
In the 1980s, heavy-ion driven ICF was probably the first proposed use of accelerators as a tool in the context of a power plant. It appeared as an alternative to laser-induced ICF as well as MCF and relies to a much larger extent on the expertise of nuclear and particle physicists in the field of high-energy accelerators. Later on, at the beginning of the 1990s when the nuclear-waste issue came to the fore, nuclear physicists like Ch. Bowman in Los Alamos and C. Rubbia at CERN revived some proposals for breeding artificial fissionable materials such as plutonium or U-233, developed in various American and Canadian laboratories since 1952. This revival was based on the new idea to make use of high-energy accelerators as drivers for waste transmutation, often including considerations of using thorium, a fuel which has the advantage of a very low actinide discharge and low radiotoxicity. These activities have led to the proposal of using a sub-critical reactor to burn large quantities of actinides currently produced in commercial light water reactors (LWR). Indeed this appears to be a promising and flexible option for the nuclear energy sector, satisfying many demands of society. In addition it has the potential to exploit the thorium cycle, which increases significantly the availability of fuel resources and reduces the hazard of proliferation.

A sub-critical reactor needs to be driven by an intense external neutron source. These neutrons are produced inside a so-called neutron spallation target bombarded by a proton beam from a high-power accelerator (typically from one to a few tens of megawatts). Because the nuclear and high-energy physics communities are very familiar with these two components (spallation target, accelerator), as well as with the related Monte Carlo computing techniques, they can make a major and specific contribution to the concept and even to an advanced design of such an ADS. Moreover, they are acquainted with nuclear data measurements needed for the design of novel reactors dedicated to actinide transmutation or based on the thorium fuel cycle. To work out specific proposals for waste transmutation requires close collaboration between nuclear physics, reactor physics and material sciences. This is particularly true for the development of ADS.

Worldwide, most of the studies related to transmutation deal almost exclusively with ADS and with the development of advanced fuels and fuel processing methods related to ADS (e.g. inert matrix, pyrochemistry). Recently, increased effort in the nuclear energy sector has been observed in the USA with the Advanced Accelerator Applications (AAA) supported by the Department of Energy (68 M\$ in the 2001 fiscal year), in Japan (the OMEGA project, and especially the joint KEK–JAERI accelerator project), and in the European Community as will be discussed in more detail below.

There is increasing support from the European Union for transmutation and ADS through the Nuclear Fission Action of the EURATOM programmes. Although still modest, the 28 M\$ allocated to partitioning and transmutation out of a total of 142 M\$ for nuclear fission in the present fifth framework programme (FP5) indicates, with respect to FP4 and FP3 (5.8 M\$ and 4.8 M\$, respectively), an increasing interest of the community in these issues. Within FP5, three basic research and eight technologically oriented programmes funded under the title of partitioning and transmutation deal exclusively with ADS and involve up to 28 partners (see Table 1.1). It is worthwhile to point out the close collaboration between research institutions and industrial partners such as Framatome or Ansaldo, making use of the valuable synergies in some of these programmes.

In 1998 an important initiative was taken by the research ministers of France, Italy and Spain to investigate the potential of ADS for transmutation. A Minister’s Advisory Group (MAG) and a Technical Working Group (TWG) were set up, the latter under the chairmanship of Carlo Rubbia. In October 1998, an interim report, endorsed by the MAG in March 1999, concluded that there was the need for an ADS demonstration programme and the corresponding basic and technological research. Soon after, the MAG was enlarged to other European countries (Austria, Belgium, Denmark, Finland, Germany, Portugal, UK and Sweden) on the basis of this TWG interim report. The TWG was
also enlarged to new EU members and in its last report, issued in April 2001, it concludes that there is the need for a European demonstration facility (XADS: experimental ADS) and defines a roadmap to achieve such a goal (see Table 1.2). With the programme PS-XADS of FP5 (see Table 1.1), the options are chosen by 28 partners from three ADS proposals (France, Italy and Belgium), all using fast neutrons and mixed U–Pu oxide fuels but differing in accelerator type, cooling fluid and target technology.

As far as the development of energy release based on controlled fusion is concerned, the impact of nuclear physics is greatest for ICF, while the main developments required for MCF are related to other fields, e.g. plasma physics and material sciences. At present the support of ICF within FP5 is rather modest (about 1% of the 700 M€ allocated to fusion) and most of the funding is provided at the national level via basic research. The state of the art on heavy-ion induced ICF is well described by D. Hoffmann and A. Tauschwitz in Chapter 3. The authors also attempt to evaluate the environmental impact of this option.

In Chapter 2, H. Condé reviews the basic reasons for waste transmutation, the specific contribution of ADS to this technique and how these dedicated reactors could be deployed within a park of light water reactors. Emphasis is put on the double-strata strategy where the major actinides — uranium and plutonium — are used in a first stratum to produce energy, whereas minor actinides — neptunium, americium and eventually curium — could be recycled in a second stratum made of ADSs.

In Chapter 4, the various issues of high-power accelerator technology in the ADS context are reviewed by M. Napolitano. Low-energy injection is at present being studied in France and Italy.

In Chapter 5, M. Salvatores describes the basic physical principles and neutron transport formalism in the case of a sub-critical reactor and refers to the important MUSE experimental programme being carried out at Cadarache on the mock-up reactor Masurca. This programme is the result of a very fruitful collaboration between nuclear and reactor physicists.

Basic nuclear physics research concerning the neutron spallation target is reviewed by P. Armbruster and J. Benlliure in Chapter 6. This concerns neutron and spallation residue production characteristics as well as nuclear data measurements above 20 MeV in order to achieve a better theoretical description of spallation and neutron transport. It shows the implication of many European nuclear physics laboratories in France, Germany, Belgium, Sweden and the Netherlands. Primarily designed for exotic nuclei studies with relativistic heavy-ion beams, the FRS facility at GSI has been used for the measurement of spallation residues by inverse kinematic reactions; this is a very good example of a nuclear physics technique applied to the ADS, especially with respect to material issues.
Table 1.1: Transmutation part (24 M€ ) of the partitioning and transmutation (P&T) programme (28 M€) of the European Union’s fifth framework programme

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Research area</th>
<th>Coordinator</th>
<th>Number of partners</th>
<th>Duration (months)</th>
<th>EU funding (M€ )</th>
</tr>
</thead>
<tbody>
<tr>
<td>MUSE</td>
<td>Experiments for sub-critical neutronics validation</td>
<td>CEA (France)</td>
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<td>36</td>
<td>2.0</td>
</tr>
<tr>
<td>HINDAS</td>
<td>High and intermediate energy nuclear data for ADS</td>
<td>UCL (Belgium)</td>
<td>16</td>
<td>36</td>
<td>2.1</td>
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Table 1.2: Roadmap for a 100 MW (th) experimental ADS

(Technical working group report, April 2001)

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<td>Years (in 20–)</td>
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<td>Construction</td>
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<td>Power testing</td>
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<td>Operation</td>
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<table>
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<th>Estimated costs (M€ )</th>
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<td>R&amp;D</td>
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<tr>
<td>Engineering design</td>
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<tr>
<td>Construction</td>
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<tr>
<td>Fuel</td>
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<tr>
<td>Total</td>
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2. Introduction to ADS For Waste Incineration and Energy Production

H. Condé

2.1 Background

Accelerator-based transmutation as a concept has been discussed since the 1940s, mostly in relation to breeding fissile material from fertile thorium and uranium [1]. The high-power accelerator needed for the transmutation techniques was not within technical reach until fairly recently. Accelerator development for high-energy physics over the last few decades has prompted proposals to develop accelerator-driven systems (ADS) for nuclear waste incineration and energy production.

One of the most pronounced problems with the use of nuclear power today is how to safely take care of the burned nuclear fuel, commonly called the reactor waste, which is highly radioactive and has a very long radiological half-life. Different means to ensure that the waste will not interplay with biological life on earth have been proposed through the years. At present, the main option in most countries having nuclear power production plants in operation is deep geological disposal with or without reprocessing. An alternative option to first incinerate and transmute the high-level radioactive waste to reduce the quantity, the radiotoxicity and the half-life before disposing of the remaining waste in a geological repository is now under consideration.

More than 99% of the amount of transuranium actinides can be separated from the spent fuels with realistic assumptions in relation to present and foreseen partitioning techniques.

A special problem is to take care of the strategic materials — highly enriched plutonium ($^{239}$Pu) and uranium ($^{235}$U) — from dismantled nuclear weapons. Different means to minimize the risk of proliferation and at the same time make use of the energy content of the materials by incineration in reactors or accelerator-driven sub-critical systems have been proposed [2]. The problem is most acute in Russia but also exists in the USA.

To overcome the safety and environmental issues with the present fission reactors, different concepts of accelerator-driven sub-critical systems have been proposed for safe and clean nuclear energy production using the thorium–uranium or uranium–plutonium fuel cycles. The environmental cleanness is given by the use of the thorium–uranium fuel cycle, which produces fewer heavy transuranic elements than the uranium–plutonium cycle, or by using the uranium–plutonium cycle with a simultaneous incineration of long-lived elements. Furthermore, the ADS techniques involve breeding of $^{233}$U or $^{239}$Pu from the most abundant isotopes in natural thorium ($^{232}$Th, 100%) and uranium ($^{238}$U, 99.3%), respectively. Thus, by using the total fission energy content of these abundant natural elements the resources are sufficient to meet the world’s energy needs for thousands of years. Also the proliferation risk of strategic materials can be diminished with the use of the thorium–uranium cycle by adding a few percent of $^{238}$U to the initial feed, which will result in an infinite critical mass of the uranium.

2.2 Main technology components of an ADS

An ADS (see Fig. 2.1) consists of a high-power proton accelerator, cyclotron or linac [3], delivering about 10 to 20 MW ($E_p = 1$ GeV, $I_p = 10 – 20$ mA) beam power to a heavy-metal target, e.g. lead, tungsten or uranium ADS.
systems. The beam power, of the order of 100 MW, has also been studied for special purposes, e.g. accelerator-driven tritium production (APT) in the USA. A proton of about 1 GeV energy generates about 30 neutrons by knocking out nucleons or clusters of nucleons in ‘spallation’ reactions with the heavy target nuclei. The spallation neutron intensity will be \(10^{14} - 10^{15}\) n/s at the actual power level of the proton beam.

The spallation neutron energy spectrum is dominated by evaporation neutrons (about 90%) with energies of a few MeV from the de-excitation of the reaction residues. It has a tail of high-energy neutrons up to the full energy of the proton beam from pre-compound reactions with the target nuclei.

The spallation target is surrounded by a sub-critical thermal or fast sub-critical assembly \((k < 0.98)\) [4] which contains the nuclear waste to be incinerated and/or nuclear fuel for energy production. The coolant is normally a liquid heavy metal or helium gas for a fast sub-critical reactor and a molten salt for a thermal reactor. The fuel is generally in solid form for the fast system and dissolved in the salt for the thermal.

Furthermore, processes are needed to separate the species to be transmuted in the spent reactor fuel as well as to clean the spent ADS fuel from fission products. The remaining transuranium elements (TRUs) and fission products from the separation processes are buried in a repository. The energy produced is fed to the electricity grid, except 10–20% which is used for running the ADS.

2.3 Waste incineration

Different concepts to reduce the amount of transuranic actinides and long-lived fission products in nuclear waste have been investigated. Transmutation by proton or heavier charged-particle bombardment has been studied to eliminate certain radioactive fission products with very low neutron capture cross-sections but is at present of minor practical interest. Also, studies have been made on fusion–fission hybrid systems for nuclear waste incineration utilizing the intense 14 MeV neutron field at the first wall of a magnetic confined fusion plasma Tokamak reactor. However, such systems are not further discussed in the present report as they are unlikely to be developed within the foreseeable future.

In the following, the discussion is limited to incineration by neutron irradiation in a critical or sub-critical assembly. The basic principle of these concepts is to induce nuclear reactions that cause the actinides to fission and the fission products to transmute by neutron capture to more short-lived and stable species.
2.3.1 The physics of nuclear incineration

A number of physical constraints have to be taken into account when one is looking for an optimal transmutation system. The incineration efficiency is basically determined by the neutron economy and the neutron energy spectrum of the critical or sub-critical reactor.

The actinide nuclei with odd neutron numbers, like the uranium and plutonium isotopes $^{233,235}$U and $^{239}$Pu, have large thermal fission cross-sections in contrast to those with even neutron numbers which only fission by fast neutrons. Some of the latter transmute to thermally fissionable nuclei by neutron capture, like $^{232}$Th and $^{238}$U which transmute to $^{233}$U and $^{239}$Pu, respectively, and are for that reason named fertile nuclei. Most of the fission products have large thermal capture cross-sections and also large epithermal resonance capture cross-sections in comparison with the cross-sections for fast neutrons. This means that a thermal or a fast neutron transmutation system can only transmute a certain fraction of the nuclear waste in an optimal way.

A reactor having a fast neutron spectrum is needed to incinerate the non-thermally fissionable transuranic isotopes. Among them are isotopes of the 'minor actinide' elements (neptunium, americium and curium), the incineration of which is a primary goal. A fast critical reactor will present safety problems when burning fuel with a large content of minor actinides. This is for two main reasons: The minor actinides have a lower delayed neutron fraction, resulting in a shorter neutron doubling time of the reactor; furthermore the Doppler feedback is low for these actinides. These safety problems of the fast critical reactor as a minor-actinide burner are overcome by the sub-criticality of the reactor in an ADS.

The accelerator driven sub-critical system with molten salt as coolant has a thermal or epithermal neutron spectrum. For this type of system, the reactor waste is dissolved in salt consisting of light- (beryllium and lithium) or medium-weight (sodium and zirconium) fluorides. The thermal neutron energy spectrum of the molten salt concepts means that one obtains a very effective incineration of thermally fissionable actinide isotopes like $^{239}$Pu and transmutation of fission products. On the other hand, the incineration of, for example, the minor actinides will be less efficient and small amounts of very heavy actinide isotopes will build up by neutron capture in non-thermally fissionable actinide isotopes. To counteract the reactivity loss by production of reactor poisons, an intense spallation source (10$^{15}$–$^{16}$ n/s) is required and/or an online separation system. Alternatively, more reactivity (plutonium or highly enriched uranium) could be added at the beginning of the cycle. Experience of the molten salt techniques exists from the running of an experimental critical facility for about four years in the 1960s at the Oak Ridge National Laboratory [5].

An excess of neutrons has to be available for transmutation purposes in each step of the chain reaction for a critical reactor and in each neutron generation step for ADS. The ADS concepts have in general a larger excess of neutrons per fission for transmutation than the critical reactors, in particular the thermal critical reactor. The neutron excess per fission ($G$) can be expressed as follows [6]:

$$G = - \sum e_J D_J - (CM + L)$$

for a critical thermal or fast reactor

and

$$G = S_{ext} - \sum e_J D_J - (CM + L)$$

for a sub-critical ADS with an external source.

The $(CM + L)$ term is the number of neutrons per fission lost in parasitic captures, absorption in construction materials and leakage. $D_J$ is the number of neutrons per fission needed to transmute isotope $J$ down to fission products (can be positive or negative) and $e_J$ is the fraction of isotope $J$ in the fuel of the system.

The $(CM + L)$ term is for all realistic power reactors about 0.3 neutrons per fission. A positive $G$ means that transmutation can be achieved, while a negative $G$ indicates a lack of neutrons.
Physics analysis shows that the minor actinides and long-lived fission products can be transmuted in standard critical reactors. However, for light water reactors one has to increase the enrichment to high values (about 10%) to transmute the minor actinides while if fast critical reactors are used one has to conceive a nuclear power park with a very large fraction of this type of reactor.

2.3.2 Light water reactor waste inventory

The burned reactor fuel contains two major element fractions: the actinides — mainly uranium but also in smaller amounts heavier elements, the transuranium elements (TRUs) like plutonium and the minor actinides (MA) neptunium, americium and curium — and fission products which are medium-weight elements from fission processes in the uranium fuel (Table 2.1). The TRUs are formed by successive neutron captures, starting with neutron capture in $^{239}$U and $^{235}$U. The actinide elements are highly radiotoxic and quite a few of their isotopes have half-lives of 10,000 years or more. Most of the fission products have very short half-lives but a few of them have long half-lives up to millions of years. Some of the long-lived products also have high solubility in water and there is a risk of migration of these elements if groundwater enters the repository.

The time dependence of the specific radiotoxic inventory of spent light water reactor fuel is shown in Fig. 2.2. Whereas the fission products decay to the equilibrium radiotoxic inventory level of the natural corresponding uranium needed in less than 1000 years, it takes about 200,000 years for the transuranium elements to decay to the same radiotoxic level.

2.3.3 Partitioning

Two different types of process can be applied to the separation of long-lived radionuclides: hydrochemical (‘wet’) and pyrochemical (‘dry’) processes [7].

The PUREX process is the most important hydrochemical reprocessing technique to separate uranium and plutonium from spent fuel and is based on the dissolution of the fuel in nitric acid. For the extraction of minor actinides the process should be modified or extended, for which extensive research is currently being conducted.

An alternative to hydrochemical processes are pyrochemical processes in which refining is carried out in molten salt. In nuclear technology, they are often based on electorefining or on distribution between non-miscible molten salt-metal phases. The major advantages of pyrochemical over hydrochemical techniques for reprocessing advanced fuels are a higher compactness of equipment and the possibility to form an integrated system between irradiation and reprocessing facility, thus reducing considerably transport of nuclear materials. For advanced oxide fuel (mixed transuranium, inert matrix or composite) and metal fuels in particular, but also for nitride- or thorium-based fuels, pyrochemistry is to be preferred. In addition, the radiation stability of the salt in the pyrochemical

<table>
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<th>Isotope</th>
<th>Half-life (yr)</th>
<th>Mass kg/yr</th>
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<tbody>
<tr>
<td>$^{237}$Np</td>
<td>2,100,000</td>
<td>14.5</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>80</td>
<td>4.5</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>24,000</td>
<td>166.0</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>6,000</td>
<td>76.7</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>14</td>
<td>25.4</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>380,000</td>
<td>15.5</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>430</td>
<td>16.6</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>7,400</td>
<td>3.0</td>
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<tr>
<td>$^{244}$Cm</td>
<td>18</td>
<td>0.6</td>
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Table 2.1: Annual production of plutonium, minor actinides and fission products from a 3000 MW(th) pressurized light water reactor with fuel burned to 33,000 MWD/ton (after 10 years’ decay)

<table>
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<tr>
<th>Element</th>
<th>Isotope</th>
<th>Mass kg/yr</th>
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<tr>
<td>$^{78}$Se</td>
<td>65,000</td>
<td>0.2</td>
</tr>
<tr>
<td>$^{90}$Sr</td>
<td>29</td>
<td>13.4</td>
</tr>
<tr>
<td>$^{93}$Zr</td>
<td>1,500,000</td>
<td>23.2</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>210,000</td>
<td>24.7</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>6,500,000</td>
<td>7.3</td>
</tr>
<tr>
<td>$^{126}$Sn</td>
<td>100,000</td>
<td>1.0</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>17,000,000</td>
<td>5.8</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>3,000,000</td>
<td>9.4</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>30</td>
<td>31.8</td>
</tr>
<tr>
<td>$^{153}$Sm</td>
<td>90</td>
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</table>
process compared to the organic solvent in the hydrochemical process offers an important advantage when dealing with highly active spent minor-actinide fuel. Shorter cooling times reduce storage costs.

The separation techniques for the minor actinides and the long-lived fission products are at a laboratory research level for both the wet and dry techniques. The goal is to obtain a separation efficiency of > 99.9% for the actinides and > 95% for the fission products to minimize the amount of waste from the reprocessing [8].

2.3.4 Incineration strategies and concepts

Two different incineration routes can be followed: the ‘double’ or the ‘single’ strata.

In the double-strata scenario the plutonium is separated from minor actinides and long-lived fission products. Uranium and plutonium are first extracted from the spent fuel and the remaining minor actinides and long-lived fission products sent to an independent partitioning and transmutation facility (Fig. 2.3a). The plutonium is mixed with uranium, both as oxides, to form the so-called MOX fuel which is burned in thermal and/or fast reactors. Several different sub-critical fast reactors for the double-strata ADS are being studied with either liquid lead or eutectic liquid lead–bismuth, liquid sodium or helium gas as coolant and solid metallic, oxide, nitride or carbide fuel. Furthermore, most of the sub-critical fast reactors are designed to contain a special thermal/epithermal zone for the transmutation of the long-lived fission products $^{99}$Tc and $^{129}$I.

All these concepts rely to some extent on experiences with power or prototype fast reactors like the Russian submarine reactors for eutectic liquid lead–bismuth, and the Phenix/Superphenix and JOYO/MONJU programmes for liquid sodium in France and Japan, respectively. There is also the project of the French (Framatome) and US (General Atomics) HTGR project for a critical gas-cooled reactor.

In particular, gas-cooled reactors with fuel pellets have also been investigated by General Atomics [9] and a Spanish–Israeli team [10] where the pellets can be continuously loaded and unloaded.

In the single-strata scenario the plutonium, minor actinides and long-lived fission products are transmuted together (Fig. 2.3b). A number of studies focusing on the single-strata option are in progress. They include studies on ADS with the same types of fast sub-critical reactor as for the double-strata concept, but also thermal or epithermal accelerator-driven sub-critical systems with molten salt as fuel and coolant.
fission products. Both sub-critical fast systems with liquid lead–bismuth or He-gas coolant and thermal systems with molten salt are being studied for multipurpose use.

### 2.5 Research on ADS

The research on ADS for waste incineration/transmutation and energy production has considerably expanded during the last few years. Current ADS research worldwide focuses on the incineration and transmutation of nuclear waste, while the production of nuclear energy by ADS is on a longer time-scale.

The funds available for partitioning and transmutation research have increased by about a factor of ten going from the fourth to the fifth Framework Programme of the European Union. The support now covers 14 research projects involving studies on partitioning, fuel, spallation, sub-critical reactors, nuclear data, etc.

The research ministries of France, Italy and Spain have agreed to collaborate on research of ADS. Several other European countries have joined this effort at a working-group level. This Technical Working Group (TWG), chaired by C. Rubbia, presented in April 2001 a ‘roadmap’ report on the research and development leading to a European ADS demonstration facility (XADS) in 2015 and a prototype (XADT) in around 2030 with close synchronization with the sixth and seventh Framework Programmes of the EU [16]. Two different concepts have been proposed for XADS, namely a liquid-metal cooled ADS by Ansaldo and a gas-cooled ADS by Framatome, both with MOX fuel.

A new project (AAA or 3A) has been initiated in the USA to merge and harmonize the research related to high-power accelerators. AAA stands for ‘Advanced Accelerator Applications’ and includes the former ‘Accelerator Production of Tritium (APT)’ and ‘Accelerator Transmutation of Waste (ATW)’ projects. It has three components:

- transformation of the APT project into an Accelerator Demonstration Facility project;
• building of an Accelerator Demonstration Facility based on APT design;

• testing and demonstrating the technologies relevant for transmutation following a roadmap study initiated by the US Department of Energy [17].

Several studies are also being carried out in Russia under the auspices of Minatom. Among them is one project of sub-critical ADSs for energy production using the thorium/uranium cycle with weapons-grade plutonium as a primer which eliminates the proliferation risk and at the same time makes use of the energy content of the plutonium [18].

The OMEGA project in Japan [19] with the aim to study different means to optimize the use of the reactor waste has been ongoing since 1988. The aim of the first phase of the project, which was to evaluate different concepts and conduct R&D on key technologies, has been achieved. The second phase is under way, which includes engineering experiments on key technologies and demonstrations. Some key issues for the research are ‘reduction of minor actinides and long-lived fission products and the time required’, ‘generation of secondary waste’, ‘increase in radiation dose’, and ‘economy’.

The Korea Atomic Energy Research Institute is developing a HYbrid Power Extraction Reactor (HYPER) for the transmutation of nuclear waste and energy production [20]. The system is being designed to utilize fast neutrons (lead–bismuth eutectics as target and coolant) for the transmutation of TRUs and has four thermal target regions for the transmutation of fission products.

The problem of finding an acceptable place for a geological repository in the Czech Republic has speeded the research on accelerator-driven transmutation. A national research programme (LA-0) has been initiated which focuses on the physics, chemistry and material problems in a sub-critical reactor with molten salt [21].

While design studies of the accelerator part of the system are being made [3], more basic studies are in progress on the spallation target, the sub-critical reactor and the coupling between these two system components [4]. At the same time, partitioning methods are being developed for certain long-lived elements in the waste, as required for an efficient transmutation.

Furthermore, fundamental research related to ADS applications is in progress in many fields like nuclear data (both experiments and reaction model code developments) [22], material research (in particular corrosion and radiation resistance), fuel development, liquid-metal and molten-salt thermal hydraulics, etc. Coordination and information exchange for mostly basic research projects are arranged at an international level by the International Atomic Energy Agency (IAEA) and the OECD/Nuclear Energy Agency.
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3. Heavy-ion Inertial Confinement Fusion

D. Hoffmann, A. Tauschwitz

3.1 Introduction

Basic research in nuclear physics has paved the way to harvest the energy stored in the binding energy of atomic nuclei. Fusion is the energy source of stars like our sun and constitutes the major source of energy in the visible universe. All chemical elements up to the maximum of the nuclear binding energy originate from hydrogen through fusion processes. While stellar objects, because of their enormous mass, make use of gravitational forces to confine the fusion fuel, this route cannot be followed for energy production on earth. Thus, there are essentially two ways to confine fusion reactions on a laboratory scale. These are magnetic and inertial confinement schemes. It was pointed out in an earlier report [1], that both schemes are approaching the relevant regime for energy production with similar progress. The thermonuclear fusion reaction which is under consideration here combines the heavy isotopes of hydrogen, such as deuterium and tritium, at extreme temperatures and pressures to form helium.

In inertial confinement fusion (ICF) the fuel is usually an equimolar mixture of deuterium and tritium at cryogenic temperature in the solid state. It is contained in a hollow spherical capsule made from glass or plastic. The dimension of the fuel capsule is on the order of a few millimetres. An intense pulse of radiation heats the surface of the capsule. The evaporating and ablating matter creates a pressure pulse, which is directed inward towards the capsule centre and thus serves to compress, in a spherically convergent process, the inner part of the capsule containing the fuel. During this process the density of the fuel is increased by a factor of 1000 or higher and a central ignition spot with high temperature is finally created, while the bulk of the still converging fuel mass remains at relatively low temperature. A burn front is created originating from the central hot spot which then propagates through the still imploding fuel. Owing to the mass inertia, the fuel mixture remains in the compressed state long enough for fusion reactions to take place at a rate which is high enough to produce more energy from fusion reactions than the amount of energy that was needed to heat and compress the fuel. In magnetic fusion the Lawson criterion,

\[ n \cdot \tau \geq 10^{14} \text{ s/cm}^3, \]

sets a lower limit on the product of the fuel particle density \( n \) and the confinement time \( \tau \). When this criterion is fulfilled, at the proper temperature \( T \) the same amount of energy is released from fusion processes as was necessary to heat the fuel sample. A similar condition applies for inertial fusion, but here it is convenient to replace the particle density \( n \) by the ratio \( \rho/m_i \) of mass density \( \rho \) and ion mass \( m_i \), whereas the confinement time is given approximately by the time \( \tau = R/c_s \) it takes for a rarefaction wave to travel the distance of the capsule radius \( R \) at the speed given by the speed of sound \( c_s \) of the compressed fuel material. Thus the condition of \( n \cdot \tau \geq 10^{14} \text{ s/cm}^3 \) is replaced by \( \rho \cdot R \geq 3 \text{ g/cm}^2 \).

The process is called a direct drive scenario, if the outer surface of the capsule is heated directly by either an intense ion or laser beam. Here it is crucial to preserve spherical convergence throughout the whole process, which is technically very challenging. In an indirect drive scenario, as shown in Fig. 3.1, the beam energy is first converted into soft X-rays through the interaction processes of the ion beam with converter material inside the hohlraum. The pellet is then located at the centre of this hohlraum and absorbs its Plackian radiation field. Even
though some efficiency is lost in the conversion process, the irradiation symmetry is improved to a level which is necessary for the process.

A power plant based on the principle of inertial fusion would typically require a continuous series of pellets to be injected into a reactor chamber at a rate of 1–10 Hz, to heat the coolant by the fusion products. The inertial fusion driver must therefore fulfill the conditions of high repetition frequency and efficiency. Intense heavy-ion beams are considered to be a promising driver candidate. This opinion is based on the high technological standard and reliability which has been achieved with accelerators in basic nuclear and high-energy physics research.

### 3.2 Accelerator drivers for ICF

The success of nuclear and particle physics in past decades is intimately connected to the impressive development of accelerator physics and accelerator technology, which strove for ever-increasing particle energies. A new trend is, however, now observable. A number of applications, which are discussed in this report call for very high beam intensities. An Inertial Fusion Energy (IFE) accelerator driver may well be viewed as the ultimate challenge for accelerator physics, since the ICF scenario requires a total energy of some MJ to be delivered into a beam spot of some millimetres within a time range of approximately 10 ns. In spite of these difficult conditions to be met by particle accelerators, heavy-ion drivers remain the most promising candidate for IFE production. They offer the advantage of high repetition rate, which is already demonstrated in a number of high-energy machines. Moreover the efficiency to convert radio-frequency (rf) energy into kinetic energy of heavy ions is inherently very high (up to 25%). This is a prerequisite for the high overall efficiency of an IFE power plant.

Laser drivers will play a unique role in the investigation of basic technological and physics issues associated with the process of inertial confinement. Therefore it is necessary for the scientific community in Europe to participate in the vast growing field of laser plasma phenomena with a sufficiently large number of high-power lasers. The two laser projects in progress — the National Ignition Facility (NIF), USA [2] and the Laser MegaJoule (LMJ), France [3] — are devoted to laboratory experiments on the behaviour of matter under very high temperature and pressure conditions and will eventually achieve ignition of fusion pellets on a laboratory scale under reproducible conditions. The repetition rate of laser slots will be compatible with the demands of such experiments; it will, however, be far from the repetition rates of some hertz required by IFE production. Even though there are ideas to increase the repetition rate and also the efficiency of laser systems to convert electrical energy into laser light, particle accelerators — heavy-ion accelerators in particular — are superior in this respect.

The concept of light-ion beam drivers has been investigated very thoroughly during the last decade. The concept is based upon intense beams of light ions generated from multi-terawatt, multi-megavolt pulsed power generators. The most advanced development is concerned with two-stage ion acceleration and charge neutralized transport [4]. It was
demonstrated that the required power level can be generated at relatively low cost. In experiments at the Sandia National Laboratories in Albuquerque a record deposition power of $10^{14}$ W/g was achieved, which is necessary to heat converters and hohlraums up to a temperature regime of close to 100 eV. These beams are, however, difficult to focus and to transport because of the space-charge and micro-divergence problems at relatively low beam energies. The pulsed power generator at Sandia was then converted to drive a Z-pinch with remarkable success. The prospects of pulsed power devices like Z-pinchers were reviewed recently [5].

During the past decades reactor studies have been performed to identify and address key issues concerning IFE driven by heavy-ion accelerators. These reactor studies were based on the concept of heavy-ion beams and a first reactor wall of the reactor chamber consisting of SiC tubes containing flowing liquid PbLi to protect the structure from hard X-rays, neutrons and pellet debris [6]. The reactor concept investigated in these studies was based on the technological state of development at that time. When it became obvious that laser facilities like NIF and LMJ were planned with the scientific goal to achieve ignition, the prospects of an accelerator facility to demonstrate ignition and gain were reviewed recently. The Heavy-Ion Driven Inertial Fusion (HIDIF) study [7] addressed the most challenging aspects of a fusion driver for ignition, namely the very short beam pulse length, small focal spots, and beam transport under severe space-charge conditions [8]. A schematic outline of such a driver scenario is shown in Fig. 3.2. The scheme is modular in the sense that doubling the driver energy requires a doubling of the number of storage rings and bunches.

The basic requirement for a heavy-ion driver accelerator is to provide short beam pulses with the highest possible intensity and phase-space density on a target at an extremely low rate of beam loss during acceleration and beam manipulation. As shown in Fig. 3.2 this will be achieved by injecting a high-intensity beam from a linac into a set of storage rings. Multi-turn injection and non-Liouville injection schemes to reduce phase-space conservation requirements have not yet been investigated in full detail in experiments, but they constitute key issues with respect to the feasibility of IFE.

While in Europe the concept of rf linacs associated with storage and buncher rings is pursued because of the successful history in nuclear physics with this kind of machine, the USA with a solid experience in high-current electron induction accelerators favours induction linacs [9]. The effort there is concentrated at the Heavy Ion Fusion Virtual National Laboratory (HIFVNL), and the focus of the current research is the design of an Integrated Research Experiment (IRE). The planning calls for a multiple-beam induction accelerator that would address, on a small scale, many of the critical technological and physics issues of an inertial-fusion driver. Induction accelerators are interesting for several reasons. They can in principle handle higher currents than the rf accelerators which are used for high-energy physics, and they allow a beam bunch to be compressed in time during the acceleration process, thus eliminating the need for storage rings. Also, induction accelerators allow key problems to be studied at low energy in small-scale experiments. The envisaged IRE is intended as an integrated experiment to test simultaneously all aspects of a driver-scale accelerator: the beam injection, transport through electrostatic and magnetic quadrupole lattices, final focusing, and transport through a reactor chamber. The results of this experiment will be a solid basis for the community to choose from several options.

### 3.3 Transport and focusing

Plasma-based transport and focusing systems have become an interesting alternative to traditional focusing devices [10], since they combine the property of high focusing strength with current- and space-charge neutralized beam transport.

The scheme shown in Fig. 3.3 [11] makes use of conventional beam transport and focusing schemes based on magnetic quadrupoles up to the final quadrupole lens array. An adiabatic
plasma lens then combines several beams. In this region space-charge effects of the intense beam become important, but the plasma of the plasma lens reduces or even eliminates this effect on a time-scale given by the plasma frequency $\omega_p$. The final transport to the pellet inside the reactor chamber is achieved inside a plasma channel.

Figure 3.3: Schematic view of plasma channel based reactor and final transport system [11].

Plasma-lens focusing has been investigated over a number of years in experiments with intense heavy-ion beams. Such systems have achieved quite remarkable results [12] with respect to focusing power and focal-spot shape. Moreover they have proved to be a reliable tool in the environment of accelerator laboratories.

The transport of the heavy-ion beam over the distance of several metres through the target chamber of a heavy-ion-beam driven fusion reactor requires a different concept. For this purpose plasma channels of considerable length are required and the particles will undergo a large number of betatron oscillations inside the transport channel. Moreover the discharge channel has to be established without any guiding structures. One way to achieve this is to initiate a discharge channel with a laser beam. Recent experiments at Berkeley and GSI Darmstadt [13] show that such a laser-initiated plasma transport channel can be generated with sufficient stability. Figure 3.4 shows the experimental set-up to study beam transport in a laser-initiated discharge plasma channel. The laser channel is generated in an NH$_3$ atmosphere. A pepper-pot mask splits the incoming beam into a large number of beamlets. Analysis of the beam at the exit of the channel shows that the quality of the beam may be maintained in plasma channel transport.

3.4 ICF target development

A necessary condition to achieve ignition and high gain is the irradiation symmetry of the pellet. Especially for a low number of beams the
symmetry requirement is difficult to achieve in a direct drive scheme. Therefore different approaches of indirectly driven targets have been investigated. Figure 3.5 shows the introduction of a number of radiation shields inside the hohlraum, which reduced the irradiation asymmetries below the required level of approximately 1% [14].

Since a two-sided illumination of the target seems to be the most suitable irradiation symmetry for intense heavy-ion beams, it is necessary to convert the ion beam energy into radiation and distribute it into almost $4\pi$ steradians surrounding the implosion capsule. In the example shown in Fig. 3.5 radiation shields are used with sufficient success. A different system was suggested by Tabak and Miller [15, 16], where the converter target is distributed in solid angle to achieve the desired radiation symmetry. The case-to-capsule ratio is of course an important issue affecting the energy gain of the fusion pellet. Two-dimensional integrated calculations show, however, that for an optimized heavy-ion target a gain of 130 can still be expected in a close-coupled version of the distributed radiator target. Optimized high-gain targets have been designed and investigated by a number of research groups [17, 18] contributing to the European Study Group on Heavy Ion Driven Inertial Fusion (HIDIF) [7].

The effort to optimize case-to-capsule ratio results has the effect that the resulting hohlraum geometry tends to become smaller. Therefore the demands on the final beam spot are also increasing and the demands on final focusing, space-charge neutralization in the reactor chamber, beam transport and emittance growth in the accelerator increase likewise.

### 3.5 Environmental aspects of thermonuclear fusion

The main fusion reactions that are relevant to energy production by thermonuclear fusion are listed in Table 3.1.

From the parameters listed in Table 3.1, it is obvious that the D + T reaction is the easiest to achieve, since it has a large cross-section at a
temperature of about 5 keV. For other reactions to occur at an appreciable rate, one would require much more stringent physical conditions. It is therefore highly probable that the first generation of fusion reactors will use a DT fusion cycle. The environmental and safety aspects of such a reactor system are of great importance. There are two major concerns in this respect. The first problem is the large number of highly energetic neutrons created in DT reactions. These neutrons cause activation of the structural material, which can pose a long-term radioactive waste disposal problem. This difficulty can be overcome to a large extent by using low activation materials [19] like vanadium alloys; however this still requires development of new materials. The second problem is tritium handling and the tritium inventory required to run the power plant. This is because tritium is a radioactive material, which is difficult to contain, and any accident leading to release of tritium would cause radioactive hazards. The safety of the power plant can be increased considerably by minimizing the tritium inventory, which is primarily determined by the daily consumption of the fuel. Designing tritium lean targets as discussed below can substantially reduce daily consumption of tritium [20]. The number of reactions taking place in DT plasma per unit volume per unit time is given by

\[ R_{DT} = \langle \sigma \cdot v \rangle_{DT} \cdot f_D f_T N_i^2 \]

where \( \langle \sigma \cdot v \rangle_{DT} \) is the DT reaction rate which is a function of ion temperature \( T_i \), and \( f_D \), \( f_T \) and \( N_i \) denote the fraction of deuterium ions, tritium ions and the total number density, respectively. In the case of an equimolar mixture of deuterium and tritium atoms one has \( f_D = 0.5 \) and \( f_T = 0.5 \), and the product \( f_D \cdot f_T \) that appears in the rate equation is 0.25.

In Table 3.2 the results for different sets of \( f_D \) and \( f_T \) are given. It is seen that if one reduces the tritium content by 20%, the reaction product \( f_D f_T \) is decreased by only 4%, and provided the plasma physical conditions are kept constant the effect on the energy yield is only minimal. Also if one reduces the tritium content by 40%, the corresponding reduction in the reaction product \( f_D f_T \) will be 16%. A suitable
3. Heavy-ion Inertial Confinement Fusion

<table>
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<th>Number</th>
<th>$f_D$</th>
<th>$f_T$</th>
<th>$f_D f_T$</th>
<th>Reduction in $f_D f_T$ (%)</th>
<th>Reduction in tritium (%)</th>
<th>Energy yield [MJ] per pellet (simul. results [21])</th>
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<tr>
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<td>0.21</td>
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<tr>
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<tr>
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<td>80</td>
<td></td>
</tr>
</tbody>
</table>

Combination may be $f_D = 0.65$ and $f_T = 0.35$, which would lead to a 30% reduction in the tritium content of the fuel mixture. The corresponding reduction in the output energy will be 9%, which may still be sufficient to operate the power plant economically [22].

3.6 Physics of dense plasmas and fast ignition

For the design of an efficient IFE target with high gain the basic processes of beam-plasma interaction and the hydrodynamic response of matter need to be understood on a microscopic level. The detailed knowledge of heavy-ion energy deposition in a target that is partly ionized and in a state of high energy density is obviously a prerequisite for the design of the target as well as for realistic simulations of target hydrodynamics. With the facilities available now, the regime of hydrodynamic expansion of beam-heated matter can be investigated. However, for experiments to measure the conversion of kinetic beam energy into radiation, in the regime of radiation-dominated plasmas, the available beam intensity is not sufficient. Laser facilities such as the projected PHELIX laser facility at GSI will allow us to address beam-plasma interaction phenomena in high-density, high-temperature plasmas [23].

In 1994 a new concept for the ignition of an inertial fusion target was proposed. The new scheme is known to the community as ‘fast ignition’ and allows a fusion target to be ignited with a considerably reduced amount of energy for the primary driver beam [24]. The usual concept of ignition as shown in Fig. 3.1 is based on spherical compression to form an ignition spark in the centre of the shock-compressed fuel. From this spark area a propagating burn wave is launched and propagates throughout the fuel. Fast ignition is based on the idea to decouple the compression and ignition process. As shown in Fig. 3.6 the fuel is first compressed in a standard way by the power of an intense heavy-ion beam. Ignition is then independently achieved in a separate process. Early discussions about fast ignition involved an external laser pulse of ultra-short duration and extremely high intensity [25]. In this case the beam has to propagate through overdense plasma, which is a regime where the plasma frequency corresponding to the plasma density is higher than the laser frequency. Classically an electromagnetic wave cannot propagate under these conditions. Interaction processes of the ultra-short, high-intensity laser beam will create a relativistic electron beam, which travels towards the highly compressed fuel core.

The physics of this process is not yet fully understood but experimental as well as theoretical investigation worldwide is addressing this.

![Figure 3.6: Schematic concept of fast ignition.](image)

In step (a) the fuel is compressed in the standard way by high-power beams, though with lower requirements on spherical symmetry. A channelling beam (b) is then used to drill a hole in the dense plasma environment and in (c) the power of an ultra-short, high-intensity laser pulse is converted into a particle beam penetrating towards the dense core, where the compressed fuel is finally ignited.
problem vigorously, since laser beams with intensities exceeding $10^{19}$ W/cm$^2$ are available now with the advent of ultra-intense short-pulse lasers [26]. Recent experiments have also demonstrated that such laser beams impinging on a curved target surface can create a high-energy, intense proton beam [27], which can be used to propagate towards the highly compressed fuel and finally ignite it. The scenario depicted in Fig. 3.7 combines all ICF schemes that have been treated separately. The power of highly efficient heavy-ion beams is converted to radiation in the converter targets of a hohlraum. The hohlraum is heated to approximately 300 eV and the pellet inside it is compressed. During the final stage of the process an ultra-short, high-intensity laser pulse is fired towards the hohlraum casing producing an intense beam of protons that propagates to the compressed fuel core and achieves ignition. In this scenario laser fusion, light ion beam fusion and heavy ion beam fusion finally unite into one scenario. The next decade will probably see a breakthrough in the physics of inertial fusion based on the development of ultra-short lasers combined with intense particle beams.

3.7 Conclusion

Inertial fusion energy is an area of active basic research while some aspects are already technically feasible. Currently the two big laser facilities under construction in the USA and in France are the main projects towards inertial fusion. A major development has been the arrival of ultra-short pulse lasers in recent years. The detailed knowledge about interaction phenomena of intense fields with matter will certainly also influence the development towards inertial fusion. As pointed out the fast ignitor scenario holds the prospects of combining the advantages of lasers, light-ion and heavy-ion beams into one inertial fusion energy scheme. It reduces the demands on the accelerator with respect to the total beam energy to be delivered to the target.
Bibliography


4. Accelerators for ADS

M. Napolitano

4.1 Introduction

The need for an in-depth study of the constituents of matter and the forces between them has motivated the development of ever more powerful particle accelerators, which are a unique tool for the experimental investigation of nuclear and sub-nuclear matter. Impressive progress on all accelerator components has been continuously achieved since their first realization in the 1930s.

Proton accelerators, with beam powers over one order of magnitude greater than the maximum power of existing accelerators, can now be designed and constructed. This allows the conception, design, and possibly the construction of plants, like accelerator driven systems (ADS) for waste transmutation or powerful spallation neutron sources for material science, which need proton beams of power ranging from several to tens of megawatts (MW). The European Spallation Source (ESS) project is an example of these facilities. A reference design of ESS, based on a 1.334 GeV, 5 MW pulsed H− linac, was published in March 1997 [1]. A definitive plan for ESS with a possible upgrade of the beam power to 10 MW will be ready in May 2002. Another example is the Oak Ridge spallation neutron source (SNS), which was approved in 1998 and is now in the construction phase [2]. The SNS project foresees the realization of a 805 MHz superconducting linac which can accelerate a pulsed H− beam up to 1.25 GeV, with a mean power of 2.65 MW, a peak current of 52 mA and a duty factor of 6% [3].

In the following, we will focus on the proton accelerators able to drive an ADS plant for nuclear waste transmutation. The requirements on beam structure, energy and power will first be discussed and the two possible alternatives for the accelerator, namely a linac and a cyclotron, will be introduced. A discussion on the important issues of reliability and availability will follow and, finally, the ‘state of the art’ and the prospects of both linacs and cyclotrons will be briefly reviewed.

4.2 Beam requirements and options

An ADS for waste transmutation requires a proton beam, possibly ‘continuous wave’ (CW) or with a very high duty cycle, having an energy of the order of 1 GeV and a power ranging from several MW for a low-power demonstration plant, up to a few tens of MW for an industrial burner [4]. The beam energy is essentially determined by two different requirements: the neutron production rate per GeV and per proton, and the energy dissipated in the input window to the spallation target. The first quantity reaches its optimum values at an energy of \( \sim 1 \text{ GeV} \), while the second rapidly decreases with energy (when \( E < 5 \text{ GeV} \)). The beam power is determined by the thermal power of the plant and by the chosen subcriticality of the system. As orders of magnitude, it can be assumed that a demonstration plant of \( \sim 100 \text{ MW}_{th} \) can be driven by a beam of a few MW, while an industrial plant with a thermal power of the order of 1500 MW_{th} needs a beam power of \( \sim 30 \text{ MW} \).

At present, the most powerful proton accelerators are the LAMPF linac at LANL [5] and the PSI cyclotron [6], both supplying a \( \sim 1 \text{ MW} \) beam: the first at 800 MeV and the second at 590 MeV.

Both linac and cyclotron have been considered as possible ADS drivers. In principle, there are no fundamental obstacles to a linac delivering beam currents up to \( \sim 100 \text{ mA} \) at an energy of 1 GeV or more. However, 1 GeV and
10 mA may be considered as limiting values for a (multistage) cyclotron, on the basis of beam dynamics constraints.

An accelerator for an ADS must satisfy the important requirement of extremely high reliability and availability. The target, core and structures of the reactor should not be too frequently exposed to the thermal shock induced by a proton beam interruption. Moreover, the activation of the accelerator structure caused by the beam particles lost along the machine must be minimized in order to allow ‘hands-on’ servicing and keep the downtime as low as possible. Therefore, the design and construction of an accelerator for ADS must introduce additional safety margins into all accelerator components.

4.3 Reliability and availability

Apart from any energy and current considerations, an accelerator for an ADS must have characteristics of reliability and availability which are unheard of in the field of nuclear and subnuclear physics, where accelerators have been developed and are largely used. These characteristics represent a major challenge for designing and building an accelerator for an ADS for waste transmutation and constitute the main item still requiring R&D activities.

The operation of the accelerators used for nuclear and subnuclear physics research has shown that sudden beam interruptions (beam trips) occur very frequently, with duration varying from very short times (of the order of milliseconds) up to relatively long times, as needed for the repair or replacement of faulty accelerator components.

The reliability requirements for an ADS driver are substantially related to the number of allowed beam trips. Frequently repeated beam trips may damage the fuel bars, the target and the structure of the reactor, and also reduce the overall availability of the plant [7].

The preliminary studies of the ADS dynamics performed so far [8] seem to suggest that short beam trips, with duration less than one second, can be tolerated by the subcritical system due to its rather big thermal inertia. The number of allowable beam trips of duration greater than one second depends on the technological details of the target and the reactor. In any case, this number can be estimated of the order of hundreds per year. Long-term beam interruptions affect the availability of the plant. In the perspective of industrial application, it is estimated that the number of unexpected shutdowns should not exceed ten per year [4]. For a conventional nuclear power plant this number is in the range of 1–2 per year.

The above numbers are normally exceeded — in many cases by orders of magnitude — in almost all existing accelerators operating for nuclear and subnuclear physics [9]. This means that operating an accelerator at a high beam power and requiring, at same time, relatively few beam trips of short duration and a negligible amount of time lost for longer beam interruptions poses new challenges in the overall accelerator design. However, while reliability and availability are primary concerns for a nuclear plant, in nuclear and particle physics — where particle accelerators have been developed and where accelerator technology is continuously advancing — priorities usually favour characteristics like higher energies, higher currents, and higher energy resolution. There should therefore be considerable potential for improving accelerators in terms of reliability and availability.

The strategy to be adopted should consider the following.

- Very advanced control systems, based on fast electronics. They must ensure immediate detection of beam trips due, for example, to the sparking of high-voltage components in order to start fast recovery actions for the component concerned within a very short time. In this way, most of the short- and medium-term beam trips, which occur at a high frequency rate in present accelerators, could be moved in the millisecond time-scale, with no damage foreseen for fuel, target and reactor structures.
- A certain amount of over-design, which should increase the stability of accelerator
components, reducing the number of beam trips — not only those due to sparking of high-voltage components and quenches of superconducting devices, but also to the failure of other accelerator components, like magnets, power supplies, vacuum and cooling systems, controls.

- Careful design of the accelerator in order to allow fast interchangeability of accelerator components that may fail during operation.
- Availability of ready-to-operate replacement units.
- Careful planning of component tests and strict acceptance criteria.

The modular structure of a linac should allow easy implementation of the above concepts. It should also be possible to envisage a ‘spare-on-line’ strategy using a higher number of modules than strictly necessary: in case of failure, a component may be switched off and the remaining part of the accelerator rapidly re-tuned.

The compactness and the reduced number of components of a cyclotron allow the above strategy to be only partially followed. However, a different approach to reliability and availability could be to drive the ADS by several independent accelerators; in this case, the reduced dimension and the relatively low cost of a single accelerator could play in favour of cyclotrons.

Concerning the ‘hands-on’ servicing and maintenance, a linac guarantees very low losses, even for very intense beams. In contrast, beam losses during acceleration and, in particular, at extraction may prove to be an extremely critical item for a cyclotron because of its weak focusing and the extremely tight packing of the orbits at high energy.

4.4 Linear accelerator: status and prospects

The design of a high-power proton linac benefits from the impressive progress made in recent decades in the field of superconducting (SC), elliptical RF cavities [10]. Most recent progress has been motivated by the construction of big accelerators like CEBAF at JLAB [11], LEP2 at CERN [12, 13], TRISTAN and KEKB at KEK [14], and, for niobium-bulk cavities, especially by the TTF-TELSA project at DESY [15].

Even though elliptical cavities were developed mainly for relativistic particles, R&D work ongoing for several years has demonstrated the possibility to extend the technology of SC elliptical cavities to the acceleration of protons of relatively low velocity, possibly down to $\beta \sim 0.5$.

The scheme of a high-power proton linac generally considered consists of three sections [3],[16]–[19]:

- the injector, which delivers a proton beam with energy of the order of 5 MeV;
- a low-energy section, which brings protons to an energy of the order of 100–200 MeV;
- a high-energy section, which accelerates protons up to the final energy by SC elliptical RF cavities.

The injector consists essentially of a source and a radio-frequency quadrupole (RFQ). The first is normally a microwave source with an accelerating voltage lower than 0.1 MV. The RFQ is made of normal-conducting copper-bulk structures brazed together. Because of its ability to accelerate and focus at the same time, the RFQ is the best tool for initial beam formation, when disruptive space-charge effects are dominant.

LEDA (Low-Energy Demonstrator Accelerator), at Los Alamos, has demonstrated the feasibility of injectors able to produce CW proton beams of 100 mA and 6.7 MeV [20]. The 8 m long, 350 MHz RFQ accelerates a 75 keV, d.c., 110 mA proton beam from the LEDA source with $\sim 94\%$ transmission and a measured normalized emittance of $\sim 0.25 \pi \text{ mm mrad}$.

Similar injectors are under construction in France and Italy. At Saclay the IPHI project [21] aims at producing a CW proton beam of 100 mA and 10 MeV. The general layout of IPHI is shown in Fig. 4.1. An ECR source, delivering a 100 mA proton beam at 95 keV, feeds a normal-conducting RFQ, able to provide
a 500 kW, 5 MeV CW beam; a drift tube linac (DTL) tank accelerates the beam up to 11 MeV. The source (SILHI) has been built for a few years. Currents up to 100 mA are reached on a routine basis with a normalized emittance below 0.2 \( \pi \) mm mrad. A long run of 103 hours uninterrupted operation, with only one beam trip, demonstrated an availability of 99.96\% [22, 23].

In Italy, a lower current (30 mA) injector [16] has been designed in the frame of the TRASCO project [24]. The proton source (TRIPS) provides a 35 mA, d.c. proton beam, with a normalized emittance below 0.2 \( \pi \) mm mrad, at an operating voltage of 80 kV. The source, based on the off-resonance microwave discharge, is a modified version of SILHI [25]. At present, TRIPS is under test. The 7 m long, 352 MHz RFQ [26] will accelerate the beam provided by TRIPS up to 5 MeV, with a transmission of 96\%. The beam loading of 0.15 MW, less than one quarter that of the LEDA RFQ, allowed a ‘relaxed’ RFQ design. The inter-vane voltage has been kept constant along the structure and gives a ‘reasonably’ low power dissipation density of 850 W per cm of structure length. A single klystron will be used to feed the resonator.

The design of the high-energy section of the accelerator is based on all designs on the technology of SC elliptical RF cavities. The main advantage of these cavities is their ability to transfer to the beam nearly all the RF power. This efficiency, approximately equal to one, has to be compared with that, roughly one half, of ordinary normal-conducting copper cavities. This characteristic is of paramount importance in order to accelerate CW beams with currents from a few mA to 100 mA or more. The operation costs of the accelerator are considerably lowered and the technological problems — related to the requirement of carrying away the huge power dissipated on the surface of copper cavities, and which may prove almost impossible to adequately solve in the case of very intense CW beams — are overcome. Superconducting RF cavities also produce relatively high field gradients, allowing the accelerator to be shortened by a factor of 2–3 and thus reducing infrastructure costs. Finally, at the frequencies usually considered (from a few hundred MHz to about 1 GHz), SC RF cavities have relatively large bore radius compared to the normal-conducting structures: a characteristic which is particularly suited for intense beams as it reduces the probability of beam losses and resulting activation problems.

As protons change their velocity from \( \beta = 0.43 \) at 100 MeV to \( \beta = 0.88 \) at 1 GeV, the acceleration over this energy range has to be done using different types of cavity. Three sections made of different cavities, matched at three different beta values, may give an efficient coverage across this energy range and, even, up to about 2 GeV. A larger number would imply increasing costs due to a wider R&D work and small production series, while a smaller one would lead to inefficient use of the cavity transit time factor. As an example, the sectioning adopted for the TRASCO 704.4 MHz accelerator design is reported in Table 4.1 for two possible values of the maximum energy [16].

R&D activities on low-\( \beta \) cavities have been ongoing for several years [16, 27–29]. In the case of niobium-bulk cavities at 2 K, accelerating gradients over 10 MV/m and quality factors \( Q > 10^{10} \) have been obtained in the range of proton velocity \( \beta = 0.5–0.8 \).

Figure 4.2 shows a single-cell \( \beta = 0.5 \) prototype cavity of TRASCO. Even though it was manufactured using low-grade niobium sheet (reactor grade, with minimal RRR = 30), the cavity performed very well above the design specifications of a \( Q = 1 \times 10^{10} \) at the accelerating field of 8.5 MV/m.

Particularly valuable is the experience gained in the ongoing development of the SC proton accelerator for the Oak Ridge spallation neutron source (SNS) [3], as it is the first high-power proton linac using elliptical SC cavities. The 805 MHz SC RF linac accelerates the beam from 186 MeV to 974 MeV, initially, and to 1.25 GeV after a possible upgrade. It has two \( \beta \)-matched sections, corresponding to \( \beta_1 = 0.61 \) and \( \beta_2 = 0.81 \). In the final configuration there will be 11 cryomodules with three six-cell \( \beta_1 \) cavities each and 21 cryomodules with four six-cell \( \beta_2 \) cavities each. Figure 4.3 shows a picture of the six-cell \( \beta = 0.61 \) SNS prototype and its \( Q \) vs. \( E_{\text{acc}} \) performances.
Figure 4.1: Layout of the Saclay high-intensity proton injector (IPHI).

Table 4.1: Sectioning adopted for the TRASCO 704.4 MHz superconducting linac.

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<td>4.2</td>
<td>4.6</td>
<td>8.5</td>
</tr>
<tr>
<td>No. of focusing periods</td>
<td>20</td>
<td>27</td>
<td>13</td>
</tr>
<tr>
<td>Max. gain per cavity [MeV]</td>
<td>3.3</td>
<td>6.0</td>
<td>11.4</td>
</tr>
<tr>
<td>Max. $E_{acc}$ [MV/m]</td>
<td>8.5</td>
<td>10.2</td>
<td>12.3</td>
</tr>
<tr>
<td>No. of cells per cavity</td>
<td>5</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>No. of cavities per section</td>
<td>40</td>
<td>54</td>
<td>52</td>
</tr>
<tr>
<td>No. of cavities per cryomodule</td>
<td>2</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>No. of cryomodules per klystron</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Max. RF per coupler [kW]</td>
<td>66</td>
<td>120</td>
<td>228</td>
</tr>
</tbody>
</table>

Because of the low proton velocity, elliptical SCRF cavities cannot be used for the low-energy section of the accelerator, for which two basic solutions are considered. The first consists of the well-proven room-temperature drift tubes accelerating structure (DTL or similar designs). The second, derived from the experience of SC heavy-ion accelerators, foresees the use of independently phased superconducting cavities (ISCL) of different designs (spoke, $\lambda/4, \lambda/2$, re-entrant cavities). This solution is being studied and R&D on cavities is ongoing \cite{16, 19, 30, 31}. A design using both solutions (say, DTL up to a few tens of MeV and ISCL from there to 100–200 MeV) may also be envisaged.

Considering what has been achieved up to now, a CW proton linac up to several tens of mA and an energy of 1 GeV or more should be within reach, even though several technological challenges remain, especially in order to obtain a very high reliability and availability. First of all, a good accelerator needs a very good injector, able to provide a high-quality, low-emittance, very stable beam. Traditionally, injector failures such as, for example, sparks of the high-voltage system are among the main causes of frequently repeated beam trips. Major problems for the RFQ and room-temperature accelerating structures are related to their thermal management. These structures have to support very high power dissipations on walls and other internal surfaces. The power density may vary greatly as a function of position, producing rather high thermal gradients, resulting in local resonant frequency variations that may result in
beam losses. The cooling system, based on high fluxes of water at high velocity, has to be very carefully studied and designed in order to get the needed thermal and dimensional stability. Particular care has to be taken in evaluating and reducing beam losses in order to allow ‘hands-on’ maintenance of most accelerator parts. This is necessary not only for safety reasons but also to fulfil the requirement of high availability, reducing the time spent on ordinary and special maintenance of accelerator parts. The linear loss of current should not exceed, for example, 50 pA/m at 1 GeV, which means a fractional loss of $5 \times 10^{-9}$ for a 10 mA proton beam. Very low losses demand, for high-quality, low-emittance beam at injector output, excellent matching and stability of all accelerator parts, careful study of beam halo formation and control. In the design and construction of the accelerator, all possible precautions must be taken to avoid or reduce damage that can be produced by even partial beam losses. Moreover, owing to the high beam power densities, the use of only non-interceptive diagnostics is mandatory.

4.5 Cyclotron: status and prospects

As a compact, relatively low-cost accelerator with a good power conversion efficiency (up to $\sim 35\%$), the cyclotron has been considered a possible ADS driver in several studies [32]–[34]. It can be considered as a serious linac competitor when the required beam power is of the order of a few MW, say up to 4–5 MW. As protons should have a minimum energy of $\sim 600$ MeV, in order to get a good spallation neutron rate on a heavy target, 4–5 MW can be obtained with proton energies in the range 600–800 MeV and a beam current in the range 5–6 mA. A cyclotron providing this kind of beam can be considered as a reasonable extrapolation of the PSI accelerator, which is at present the cyclotron that has accelerated the beam of the highest power ($\sim 1$ MW, namely 1.8 mA at 590 MeV) and is usually considered as the reference machine. The PSI accelerator consists of a 590 MeV separated sector cyclotron, injected at 72 MeV by an injector cyclotron, which is, in turn, fed by a Cockcroft–Walton pre-injector delivering a 870 keV proton beam. The main characteristics of the PSI cyclotron are reported in Table 4.2.

The performances of this 30-year-old cyclotron have far exceeded the initial design current of 100 $\mu$A at 590 MeV. The great increase of the beam current, with losses lower than 1 $\mu$A at extraction, has been achieved by three main upgrades:

- construction of a new injector able to deliver up to 2 mA;
- insertion of a flat-topping cavity into the main ring cyclotron;
4. Accelerators for ADS

Figure 4.3: A six-cell $\beta = 0.61$ SNS prototype cavity and its $Q$ vs. $E_{\text{acc}}$ plot.

Table 4.2: Characteristics of the PSI ring cyclotron.

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Injection energy</td>
<td>72 MeV</td>
</tr>
<tr>
<td>Extraction energy</td>
<td>590 MeV</td>
</tr>
<tr>
<td>Beam current</td>
<td>2 mA</td>
</tr>
<tr>
<td>Beam power</td>
<td>1.1 MW</td>
</tr>
<tr>
<td>Injection radius</td>
<td>2.1 m</td>
</tr>
<tr>
<td>Extraction radius</td>
<td>4.45</td>
</tr>
<tr>
<td>Number of sectors</td>
<td>8</td>
</tr>
<tr>
<td>Sector width</td>
<td>18°</td>
</tr>
<tr>
<td>Spiral angle</td>
<td>35°</td>
</tr>
<tr>
<td>Sector gap</td>
<td>50–90 mm</td>
</tr>
<tr>
<td>$B_{\text{null}}$</td>
<td>2.09 T</td>
</tr>
<tr>
<td>Number of cavities</td>
<td>4</td>
</tr>
<tr>
<td>RF frequency</td>
<td>50.6 MHz</td>
</tr>
<tr>
<td>Harmonic mode</td>
<td>$h = 6$</td>
</tr>
<tr>
<td>Maximum voltage</td>
<td>730 kV</td>
</tr>
<tr>
<td>Flat-topping cavity</td>
<td>1</td>
</tr>
<tr>
<td>Energy gain per turn</td>
<td>2.46 MeV</td>
</tr>
<tr>
<td>Turn separation</td>
<td>$&gt; 7$ mm</td>
</tr>
<tr>
<td>$\Delta\varphi_{\text{ext}}$</td>
<td>$8^\circ$</td>
</tr>
<tr>
<td>Energy spread</td>
<td>0.1%</td>
</tr>
</tbody>
</table>

- increase of the accelerating voltage of the four RF cavities from the design values of 350 kV up to 730 kV.

To date, the maximum current has been limited by the accelerating voltage of the RF cavities and by losses at injection. In the near future two new RF cavities will be installed in the injector to push the deliverable beam current up to 4 mA. A prototype cavity for the main ring, able to achieve 1 MV, is also under construction.

There is a general consensus among experts that it should be possible to design and build a machine able to provide a 10 MW beam at 1 GeV. A few conceptual studies exist [35, 36], based on a few cascading stages, at least the last of which should be a separated sector cyclotron. A general scheme foresees three different stages and is similar to the PSI scheme:

- a pre-injector consisting of an intense source coupled to a compact cyclotron or a Cockcroft–Walton accelerator or an RFQ (beam energy can range from less than 1 MeV up to a few MeV);
- a separated-sector intermediate cyclotron, with at least four sectors, that brings protons up to roughly 100 MeV;
- a final separated-sector cyclotron, which accelerates protons up to 1 GeV and should contain the order of 12 sectors.

The beam extraction is one of the most critical aspects of a cyclotron. For a 1 GeV, 10 MW beam extremely efficient beam extraction is needed to minimize beam losses that could damage the apparatus and activate the components, preventing hands-on servicing and maintenance.
Single-turn extraction is mandatory for high efficiency; this requires well-separated orbits for which a high energy gain per turn and a well-defined narrow beam are necessary. The first is obtained using very high accelerating potentials. With regard to the second, the dimensions of a high-energy, high-intensity beam are mainly determined by the energy spread, which, hence, has to be low in order to obtain a narrow beam. Then, owing to the lack of any longitudinal focusing, the beam has to be bunched with a relatively low phase acceptance (10–20° RF) and has to reach the extraction after a relatively low number of turns, which still requires high accelerating potentials. High potentials and large dimensions make the accelerating cavities, together with the extraction, a very critical part of the design and construction of the cyclotron.

Even though a 1 GeV, 10 mA cyclotron can be seen as an achievable goal, the RF, injection and extraction systems require intense studies and R&D.

Concerning extraction, in order to reduce beam losses, acceleration of $\text{H}_2^+$ instead of protons, and extraction by stripping has been proposed [37]. Extraction by stripping does not require well-separated orbits. The energy gain per turn may be lower, allowing lower accelerating potentials and lower power losses in RF cavities. Preliminary studies of a superconducting ring cyclotron, able to accelerate a $\text{H}_2^+$ beam to 1 GeV/amu, have been published [38].

Only preliminary conceptual studies have been done up to now on high-energy, high-current cyclotrons. The extrapolation of one order of magnitude of the highest achieved cyclotron beam power might not be ‘within reach’ in the same way as it is for a linac, for which the intrinsic characteristics and intense R&D activity, ongoing for many years, make feasible a several tens of MW beam accelerator.
Bibliography


5. The Physics of Subcritical Multiplying Systems

M. Salvatore

5.1 Introduction

Accelerator-driven systems (ADS) have been recognized world-wide [1]–[3] as a highly relevant tool with the potential to transmute very large amounts of radioactive waste and, consequently, to lower the burden on deep geological storage.

Both the two major variants of the strategies of transmutation that make use of ADS [4] call for subcritical cores with a fast neutron spectrum and a fuel dominated by mixtures of plutonium and minor actinides (MA), essentially fertile-free.

These cores are characterized by a very low fraction of delayed neutrons and by a low (even near zero) Doppler reactivity coefficient. In principle, the subcriticality will help to reduce (or to eliminate) the negative consequences of these characteristics on the safety of the multiplying medium. This is one of the points that will be discussed in this chapter.

The physics of the ADS and of its subcritical core is well understood, and there are several publications which deal extensively with the subject (see for example Refs. [5] and [6], among many others). However, several concepts are new and their understanding requires experimental validation.

Here we will focus on a description of the basic physics phenomena in the subcritical multiplying core, with reference to the coupling phenomena and their impact on the subcritical core (SC), when needed.

We will also indicate the areas that need particular care for experimental validation and we will quote some ongoing experimental programmes and preliminary results.

Finally, the inspection of some ‘visual’ images of SCs as they are being studied in several laboratories today will be the occasion to point out some relevant design-oriented problems of SCs and their integration in an ADS.

5.2 Subcritical multiplying core in stationary regime

5.2.1 Flux distribution

In a critical system, the condition of balance of neutron production and neutron disappearance at each point of the phase space \((E, r, \Omega)\) is expressed by the Boltzmann equation, which can be expressed in matrix form:

\[
A\phi = P\phi
\]

where \(A\) is the ‘disappearance’ operator and \(P\) the ‘production’ operator, and \(\phi\) the flux.

In the same system, made subcritical, the condition to have a stationary system is to have an external source \(S(E, r, \Omega)\) such that, e.g., Eq. (5.1) can be written as

\[
A\bar{\phi}_{in} = P\bar{\phi}_{in} + \bar{S}.
\]

\(\bar{\phi}_{in}\) is the solution of the inhomogeneous equation (5.2). The distribution in space, energy and angle of \(\bar{\phi}_{in}\) is obviously different from that of \(\phi\). Of course, \(\bar{\phi}_{in}\) approaches \(\phi\) as the level of subcriticality becomes smaller, approaching the critical configuration.

For an ADS, once the material properties, the geometry of the system, the relevant cross-sections and the source intensity (in units of neutrons per second) have been defined, the distribution of the inhomogeneous flux is fully determined by Eq. (5.2).
Relevant integral parameters characterizing the SC, such as reaction rates, can be easily calculated. This allows us to evaluate the power deposited in each point of the system, the damage rate, the breeding ratio, etc. This is done exactly as in critical systems, characterized by $\bar{\phi}$.

### 5.2.2 Reactivity of the subcritical core

It is formally possible to describe a subcritical system with the introduction of a parameter $K_{\text{eff}}$ which allows us to 'restore' the balance equation (5.2):

$$A\bar{\phi} = \frac{1}{K_{\text{eff}}} P\bar{\phi}. \quad (5.3)$$

Since $\bar{\phi}$ has the same distribution as the 'critical' flux, this equation is obviously an approximation of the real case, as described by Eq. (5.2).

In order to improve the definition of subcriticality and to take into account the change in shape of the flux, a different definition of the subcriticality, by means of a 'K-source' $K_S$, has been proposed. The procedure is to apply the formal balance condition (5.3) to the inhomogeneous flux equation (5.2):

$$A\bar{\phi}_{\text{in}} = \frac{1}{K_S} P\bar{\phi}_{\text{in}}. \quad (5.4)$$

Integrating and recalling that $A\bar{\phi}_{\text{in}} = P\bar{\phi}_{\text{in}} + \bar{S}$ one obtains

$$K_S = \frac{\langle P\bar{\phi}_{\text{in}} \rangle}{\langle P\bar{\phi}_{\text{in}} \rangle + \langle \bar{S} \rangle}. \quad (5.5)$$

### 5.2.3 The $\varphi^*$ parameter

In a subcritical system, the evaluation of the relative importance of the source neutrons with respect to the fission neutrons generated in the SC is of relevance in understanding the behaviour of the source-driven SC.

One introduces a parameter $\varphi^*$, which is the ratio of the average 'importance' of the source neutrons to the average importance of fission neutrons. It can be shown that this parameter $\varphi^*$ is related to $K_{\text{eff}}$ through the following relation:

$$\frac{K}{\bar{\nu}} \varphi^* = \frac{1}{K_{\text{eff}}} - 1 \quad (5.6)$$

where $\bar{\nu}$ is the average number of prompt neutrons per fission and $\Gamma$ the average source neutrons per fission. Relation (5.6) is given in Ref. [7], where the experimental determination of $\varphi^*$ is discussed.

The $\varphi^*$ parameter plays an important role in the ADS performance-parameter assessment. In fact, in Ref. [8] it is shown that the relation between the proton beam current $i_p$, the power in the SC and its subcriticality is given by

$$i_p = \frac{\bar{\nu} \left( \frac{1}{K_{\text{eff}}} - 1 \right) W}{\varphi^* Z \epsilon_f} \quad (5.7)$$

where $W$ is the power of the SC in watts, $\epsilon_f$ the energy per fission (MeV) and $Z$ is the number of neutrons per incident proton.

It can be seen from Eq. (5.2) that a value of $\varphi^*$ higher than 1 can reduce proportionally the proton beam current requirement, for a given subcriticality level. Measurements of $\varphi^*$ are made in the CEA facility MASURCA in Cadarache, in the frame of the MUSE programme [11], described briefly in Section 5.4.1.

### 5.3 Kinetics of a subcritical system

#### 5.3.1 Asymptotic behaviour

The equations which give the kinetic behaviour of a system driven by an external source are of the type [9]

$$\begin{align*}
\frac{dW}{dt} &= \frac{\varphi - \beta_i}{\epsilon_{\text{eff}}} W + \sum \lambda_i C_i + S \\
\frac{dC_i}{dt} &= \frac{\beta_i}{\epsilon_{\text{eff}}} W - \lambda_i C_i
\end{align*} \quad (5.8)$$

where $C_i$ are the precursors of delayed neutrons with decay constant $\lambda_i$. $\beta_i$ is the fraction of the

---

1. 'importance' of a neutron in (E, r, $\Omega$): this function, which defines the contribution of a neutron born in (E, r, $\Omega$) to the asymptotic power level, is the solution of the equation adjoint to Eq. (5.1).

2. This relation is directly related to the 'energy gain', as defined in Ref. [5].
total number of delayed neutrons emitted per fission ($\sum \beta_c = \beta$) due to precursors $C_i$. $\ell_{eff}$ is the neutron generation time and $\rho$ is the reactivity ($\rho = (1/K) - 1$).

In steady state (i.e. if $dW/dt = 0$ and $dC_i/dt = 0$), we have

$$\rho W = \ell_{eff} S.$$  \hspace{1cm} (5.9)

A decrease by a factor $h$ of the reactivity ($\rho' = \rho/h$) or an increase by a factor $h$ of the source ($S' = hS$), induces an instantaneous increase of the power $W' = hW$. For example, if the system is subcritical by $-10\beta$, a reactivity insertion of $+5\beta$ produces a doubling of the power (see Fig. 5.1). This of course is totally different from the behaviour of a critical system (which becomes prompt critical).

In more general terms, the kinetic behaviour of a critical system is characterized by delayed neutrons and their time constants (about 10 s), while the kinetic behaviour of an SC is determined by the time constants related to the external source, in the sense that an instantaneous variation of source has an effect on the time-scale of the prompt neutron lifetime (typically of the order of microseconds).

The evolution of the power with time and the related variation of the temperature are associated with the variation of the reactivity (Doppler reactivity effect, fuel expansion reactivity, reactivity due to the material concentrations in the core, including the coolant, etc.). These reactivity effects (feedback reactivity effects) are essential for the safety of a critical reactor. In an SC, the relevance of the feedback reactivity depends on the level of subcriticality. In fact, for a deeply subcritical core, the dynamic behaviour is dominated by the external source and its variation with time. Closer to criticality, the feedback effects become more important and the behaviour of the core approaches that of the corresponding critical core.

In a very simplified way, if the core is subcritical by $-10\beta$, a feedback reactivity equal to $\pm 1\beta$ induces a $\pm 10\%$ variation of power. The corresponding variation is $\pm 5\%$ if the system is subcritical by $-2\beta$. In a critical reactor, a $+1\beta$ reactivity insertion makes the reactor prompt critical and $-1\beta$ stops the chain reaction. In view of the definition of an ‘optimal’ level of subcriticality, it is of high relevance to verify the transition of the behaviour of the SC from a ‘source-dominated’ to a ‘feedback-dominated’ regime.

### 5.3.2 Reactivity and flow-loss accidents

Fast external reactivity insertions give rise to different consequences in critical and subcritical cores. Examples have been given in Refs \cite{10,11}. In Ref. \cite{10} a 0.55 $\beta$/s reactivity insertion in a PHENIX fast reactor type core, critical or subcritical at $K_{eff} = 0.95$, gives rise (at constant external source level) to the power and average-temperature evolutions given in the following table.

In Ref. \cite{11}, a reactivity of 170 $\beta$/s is injected in a critical core ($W_o = 1$ GW), or in the same core made subcritical at $-1\beta$, $-2\beta$, $-3\beta$. The

![Figure 5.1: Core power evolution for different situations (beam shut-down, reactivity insertion).](image-url)
results show that prompt criticality is reached in the critical core after 6 ms with a first power peak of 700 GW at 8.5 ms and a second peak of 500 GW at 13.2 ms. In the subcritical mode, the peaks are 530 GW at $-1\beta$, 6 GW at $-2\beta$ and 2.2 GW at $-3\beta$ (t = 16 ms) (see Fig. 5.2).

The increase in power is considerably slower in a subcritical system, and the total energy deployed is much smaller.

In the case of loss-of-coolant-flow accidents, Refs. [10] and [11] give simple examples which show that, in the case of no shut-down of the source, the behaviour of a $-10\beta$ subcritical system is less favourable, since in a critical system the increase of the coolant temperature is slower and lower because of the feedback effects. Again, the choice of the level of subcriticality is relevant if one takes into account the potentially beneficial effects of the intrinsic characteristics of the core.

This, of course, has to be verified for each type of core and associated fuel and coolant. It is obvious from these considerations that the accelerator beam intensity must be coupled to the power level of the SC, so that it can immediately be shut down in case of a power excursion.

### 5.3.3 Cores with low Doppler effect

In the case of an ADS dedicated to transmutation, the fuel will be dominated by minor actinides, which will have a low Doppler effect owing to the absence of U-238.

The dynamic behaviour of the core will be differently affected by this according to the level of subcriticality (see Fig. 5.3). At large subcriticality, the calculations of the effect of reactivity insertion performed with a ‘standard’ Doppler coefficient $K_D$, or with a ‘low’ Doppler ($K_D = 0.1 K_D$), show no difference in the power or reactivity behaviour. In contrast, close to criticality the effect can be significant.

![Figure 5.2: Fast reactor power excursion benchmark (as defined in a comparative NEACRP exercise) assuming a rod ejection accident. The reactivity insertion rate is 170 $\beta$/s during a period of 15 ms. The power increase from a critical reactor is compared with 1$\beta$ to 3$\beta$ subcritical accelerator-driven systems of the same initial power. (From Ref. [11])](image)

![Figure 5.3: Insertion of one-third reactivity in one second: Behaviour of a PHENIX-type ADS at three different levels of subcriticality [10].](image)

### 5.3.4 Choice of subcriticality level

No final criteria have yet been established for defining an ‘optimal’ level of subcriticality. However, previous considerations indicate the relevance of finding a compromise between the source- and feedback-dominated regimes.

More quantitatively, in the case that no control rods are foreseen in the SC, the level of
subcriticality should be such that the core stays subcritical when going from a 'hot' state (i.e. normal operation) to a 'cold' state (i.e. reactor shut-down). Since thermal feedbacks induce generally (e.g. in standard fast reactors) a positive reactivity effect (\(\Delta K_{FB}\)) going from a hot to a cold state, one can require that the cold core stay subcritical even in the case of an accidental reactivity insertion (\(\Delta K_{AC}\)) resulting, for example, from coolant voidage. In that case the required \(K_{eff}\) should respect the following relation:

\[
K_{eff} + \Delta K_{FB} + \Delta K_{AC} < 1. \tag{5.10}
\]

During operation, the maximum reactivity insertion (\(\Delta K_{AC}^{M}\)) can be higher than \(\Delta K_{AC}\). In that case one has the requirement that

\[
K_{eff} + \Delta K_{AC}^{M} < 1. \tag{5.11}
\]

Moreover, during the reactor operation, the reactivity varies because of the irradiation (burn-up) of the fuel and its isotopic evolution. In general this reactivity variation \(\Delta K_{BU}\) is negative, but in some cases (e.g. a fuel made essentially from minor actinides, which act as 'fertile' materials since they are transmuted to more 'reactive' elements, as is the case for example for Am-241), \(\Delta K_{BU}\) can be positive. In that case, if the core has no control rods and one does not want to modify the external source, e.g. by changing the current intensity, one should have

\[
K_{eff} + \Delta K_{AC}^{M} + \Delta K_{BU} < 1. \tag{5.12}
\]

Looking for a compromise among the different criteria indicated above, one has also to consider that a very large subcriticality may not necessarily be the optimal solution. In fact, besides obvious considerations on the 'cost' of a strong external source, a largely subcritical core has a peaked power distribution, dominated by the source distribution and therefore very far from the flat behaviour in space, required to optimize the fuel irradiation (and consequently the fuel transmutation).

### 5.3.5 Reactivity control and monitoring

The control of reactivity and of the power level in a critical reactor is made essentially using control rods. In principle, the control in an ADS can be made with only the external source. As an example, the variation of reactivity with the fuel burn-up can be compensated for with an appropriate change of the beam current intensity. A similar system can be conceived to control the reactivity change between hot and cold states. However, major variations of the current would be necessary. For example, in an SC without control rods — which has \(K_{eff} = 0.99\) in the cold state, \(K_{eff} = 0.98\) in the hot state at the beginning of the irradiation cycle and \(K_{eff} = 0.95\) at the end of the irradiation cycle — the source intensity should change by a factor of approximately five to account for both the trip towards nominal power and the operation cycle. In this context, it is clear that the use of control rods to ensure at least some of the functions of reactivity control should be carefully examined.

Moreover, if it is true that in an SC, in particular in source-dominated mode, the shutdown of the source has an instantaneous power-reducing effect, the inverse effect (i.e. an 'overshoot' due to a sudden increase of the external source) results in an instantaneous power increase. Although more limited than the potential power increase in a critical reactor, such an accidental situation should be examined.

Also, when the reactor is shut down, the consequences of the insertion of the full 'reserve' of beam current should be analysed. In fact, if the insertion of the full beam-current reserve cannot be excluded, this accidental event could lead to a power variation given by [12]

\[
\frac{W'}{W} = 1 + \frac{\Delta \rho}{\rho + \beta} = 1 + \frac{\delta_{IP}/\rho}{1 + \beta/\rho}. \tag{5.13}
\]

If \(W_{Max}\) is the maximum allowable power in a short time interval, one can deduce the maximum allowable subcriticality level such that \(W' \leq W_{Max}\).

Finally, we should mention that in principle long-term variations of the reactivity can be
achieved by an appropriate variation of the $\varphi^*$ parameter. This can be obtained, for example, by changing the geometrical arrangement of the buffer (or of the buffer material) surrounding the spallation source.

As for the monitoring of the subcriticality level, different methods can be envisaged and experimentally validated. Below are two examples.

- Use of a pulse mode of the source. The recording of the time evolution of the counting rates of in-core neutron detectors allows the reactivity to be measured. In fact, the point kinetics predicts the prompt decay of the neutron population after a pulse to be of the type $\exp(-\alpha t)$ with $\alpha = (\rho - \beta)/\ell_{\text{eff}}$. For known $\beta$ and $\ell_{\text{eff}}$ one can deduce $\rho$ from the decay of the neutron population obtained experimentally.

- If control rods are foreseen, the modified source multiplication method (MSM [13]) can be used provided the calibration of control-rod reactivity is performed at near-critical level.

5.3.6 Beam trips

As far as the coupling of the accelerator to the subcritical core is concerned, one significant point which has been raised [14] is the effect of frequent beam trips on the SC. Since we have seen that the time-scale for power variation (due to source variation) is very short, the heat transfer time from fuel to coolant being of the order of 0.1/1 s, the heat is stored in the fuel for $\sim 1$ s making high-thermal-conductivity fuels a possible requirement. In a similar way, thermal stresses in the core structures can be expected (due to the difference of time constants between power increase and temperature variations in the structures), and in the case of frequent beam trips fatigue failures of the structures could occur and cause safety concerns (see also Chapter 4).

5.4 Experimental validation

The physics characteristics and the predicted behaviour of an SC, as outlined in previous paragraphs, need an experimental validation in order to calibrate the calculation tools and to gain confidence in the prediction of the basic safety features of an eventual future ADS, which will be fuelled with very innovative fuels.

The following are the main fields which require experimental validation.

- The effects of the relative contributions of source neutrons and neutrons generated by fission. $\varphi^*$ measurements should allow this objective to be achieved in stationary conditions.

- Kinetic experiments performed at different subcritical levels, with or without feedback effects, can be essential to understand the transition between a source-dominated and a feedback-dominated regime.

- Space and energy distributions of neutrons, and their variations close to the external source.

- Subcriticality level assessment and monitoring.

- Relation between the external source and the power in the core.

5.4.1 MUSE experimental programme

Physical principle

A first experiment to check the physical principles of an ADS was performed by C. Rubbia at CERN (FEAT experiment [15]). A proton beam did hit directly a natural uranium block, and the 'energy amplification' was experimentally verified.

Since 1995, at the MASURCA facility of CEA in Cadarache, a series of MUSE experiments (Multiplication avec Source Externe) has been performed (see Table 5.1) in a collaboration between physicists from Cadarache (CEA) and ISN-Grenoble (IN2P3). The principle of
Table 5.1: The MUSE experiments at MASURCA

<table>
<thead>
<tr>
<th>Type of source</th>
<th>Range of subcriticality</th>
<th>Diffusing buffer around the source</th>
</tr>
</thead>
<tbody>
<tr>
<td>MUSE-1 (1995)</td>
<td>$-1.5% \frac{\Delta K}{K}$</td>
<td>none</td>
</tr>
<tr>
<td>MUSE-2 (1996)</td>
<td>$-3.0% - -3.5% \frac{\Delta K}{K}$</td>
<td>sodium steel</td>
</tr>
<tr>
<td>MUSE-3 (1998)</td>
<td>$-0.5% - -6.0% \frac{\Delta K}{K}$</td>
<td>sodium steel</td>
</tr>
<tr>
<td>MUSE-4 (2000–2001)</td>
<td>$-1% - -0.4% \frac{\Delta K}{K}$</td>
<td>lead</td>
</tr>
</tbody>
</table>

Figure 5.4: The MASURCA installation for the MUSE programme.
these experiments [7] is to make the hypothesis of the separability of the effects of source and multiplication in the SC.

Intuitively, one might think that a source neutron will lose ‘memory’ 1–2 mean free paths after entering the SC and behave like any other neutron produced by fission in the SC.

This hypothesis is of course made for an SC not too subcritical (e.g. with $K_{eff} \geq 0.95$). Under this hypothesis, it is possible to study the neutronics of the source-driven SC, using a well-known external source instead of a true spallation source. The first MUSE experiments were performed with a Cf-252 spontaneous fission source, located at the centre of an SC [7].

The present MUSE experiments (MUSE-4) use a pulsed 14 MeV neutron source called GENEPI, built at INS-Grenoble. A deuton accelerator has been coupled to the MASURCA facility, and a deuterium or a tritium target located at the centre of the SC (see Fig. 5.4). These targets are surrounded by a lead buffer to simulate the neutron diffusion inside an actual lead (or lead–bismuth) target. Numerical simulations have shown the validity of the basic hypothesis of the experiments, namely that using a spallation neutron source or the neutrons issued from the (d,d) or (d,t) reactions, the neutron spectrum in the core close to the buffer region is very much the same, whatever the neutron source energy distribution.

This result is shown in Fig. 5.5, where the neutron spectra are shown at the buffer–core interface and 10 cm from that interface. Only at the former are some differences observed.

**Results and techniques**

Experimental results of relevance have already been obtained. For example, in Figs. 5.6 and 5.7 the flux distribution inside the SC of the MUSE-1 configuration is shown in terms of the measured U-235 fission rate, in the presence of the Cf-252 source.

Figure 5.6 shows the radial flux distribution in the core with and without external source. The presence of the source gives a more ‘peaked’ distribution, as expected.

Figure 5.7 shows the axial distributions when the source is at the core–upper reflector interface (+25 cm from the core midplane). Three axial distributions of the fission rates are shown. Far away from the source, the axial profile becomes less sensitive to the asymmetrical position of the source.

Moreover, $\varphi^*$ measurements have been performed and Table 5.2 gives a comparison of
Table 5.2: $\varphi^*$ measurements in MUSE-1 [7].

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Calculated $\varphi^*$</th>
<th>Measured $\varphi^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cf-252 source at core centre</td>
<td>1.25</td>
<td>1.19 ± 4%</td>
</tr>
<tr>
<td>Cf-252 source at core-blanket axial interface</td>
<td>0.91</td>
<td>0.90 ± 4%</td>
</tr>
</tbody>
</table>

(MUSE-4, see Fig. 5.4). Different experimental techniques (transfer function, MSM method, Rossi-$\alpha$ and Feynman-$\alpha$ [16] will be used to measure the subcriticality level, but also $\ell_{eff}$ and $\beta_{eff}$ and control rod worths in the SC.

5.5 Some ADS ‘Images’ and technological problems

Several countries and leading research laboratories are actively working on the development of ADS, in particular in the frame of radioactive waste minimization strategies.

Conceptual designs have been developed. A typical example is the Energy Amplifier proposed by C. Rubbia [17] and mentioned in Chapter 2.

Conceptual designs have also been developed for experimental ADS, in the power range 80–100 MWt. Figure 5.9 indicates two of these configurations: one lead-cooled, developed by Ansaldo (Italy); one gas-cooled, developed by the Novatome division of Framatome (France).

All these conceptual layouts are of a preliminary nature and some relevant technological problems are still to be accounted for in a satisfactory way.

This is the case, for example, for the shielding configurations in the upper part of the systems. The shielding in fact should account for the potential deep penetration of high-energy neutrons ($E_n > 100$ MeV) issued from the spallation of protons (typically $E_p = 0.6 – 1.5$ GeV).

High-energy neutron penetration experimental studies performed in Japan confirm the very large thickness of material (like concrete or stainless steel) needed in order to reduce to an acceptable level the doses around the structures.

The beam entrance configuration is also a
assembly
(e.g. SNR-300 S/A with MOX fuel)

Figure 5.9: Sketches of liquid-metal cooled (right) and gas cooled (left) ADS (not to scale), representative of the European X-ADS proposals (Ansaldo and Framatome).

matter of concern. In fact, a simple vertical beam entrance might imply a very complicated fuel loading-unloading system and also might not be optimal for preventing the intrusion of back-scattered neutrons into the beam tube void, as is required.

These are just a few examples of technological problems that can have an impact on the coupling of the different components of an ADS, and which might require substantial effort in order to develop a robust ADS design.

However, its specific features have never been fully experimentally demonstrated.

Several steps have been undertaken in that direction and planned experiments (like the MUSE experiments) should give most of the demonstrations needed in order to proceed to a sound design of an experimental ADS, as proposed, for example, in the European roadmap towards an ADS demonstration [1].

5.6 Conclusions

The physics of a subcritical core, driven by an external neutron source, is generally well-understood.
Bibliography


6. Basic Nuclear Data at High and Intermediate Energy for Accelerator-Driven Systems

P. Armbruster, J. Bendlivre

6.1 General considerations on spallation reactions needed for ADS

Nowadays it is well established that spallation reactions constitute an optimum neutron source to feed a subcritical reactor in an accelerator-driven system (ADS). However, the present knowledge about this reaction mechanism is not accurate enough for any technical application. Two main aspects will play a major role in the design and construction of the target assembly of the spallation neutron source used in an ADS: the neutron yields and the residual nuclei produced in the reaction.

The neutron production should be characterized in terms of the neutron multiplicity and their spatial and energy distributions. The neutron multiplicity will determine the current and beam energy of the proton-driver accelerator, while their energy and spatial distribution shapes the geometry of the spallation target and the shielding to high-energy neutrons.

Spallation reactions produce not only neutrons but also residual nuclei. Most of these nuclei are radioactive, therefore activation problems should be considered in the design of the target. Figure 6.1 represents the simulated activity induced in a cylindrical lead target by a 1 mA proton beam after one year of irradiation. As can be seen in the figure, both the cooling times and the total activities induced in the target are not easy to handle. In addition the residual nuclei will contribute to the corrosion and to the radiation damage in the target, accelerator window and structural materials.

The main consequence of the present qualitative understanding of spallation reactions is that most of the existing codes used to describe these reactions have a limited predictive power. Therefore, a large experimental programme was initiated in Europe a few years ago in order to improve our knowledge on these reactions. These experiments are expected to provide accurate data to benchmark more reliable model calculations. In the following sections we will describe the highlights of this experimental programme.

![Graph showing total radioactivity](image)

**Figure 6.1:** Calculated radioactivity induced in a cylindrical lead target (120 cm long and 46 cm diameter) by a 1 GeV proton beam of 1 mA after one year of irradiation. Calculations done with the Lahet code system [1].

6.2 Interaction of relativistic protons with thick targets

Spallation reactions are induced by light projectiles at relativistic energies impinging on a heavy target. These reactions can be described as a two-stage process.

First the incoming projectile interacts in quasi-free nucleon-nucleon collisions with the nucleons of the target nucleus. These collisions lead to the prompt emission of few high-
energy nucleons while a fraction of the kinetic energy of the incoming projectile is transferred to the target nucleus as excitation energy (e.g. a 1 GeV proton is expected to deposit on average 200 MeV in the target nucleus, leading to the emission of around five prompt nucleons). These fast nucleons will play an important role in the development of an internuclear cascade process inside the target.

In a second step the residual nuclei produced in the collisions will de-excite by evaporation of low-energy neutrons and protons or by fission. Neutron emission is favoured since to evaporate protons or to fission extra energy is needed to overcome the Coulomb or fission barriers. Around 10 additional neutrons are expected to be produced in this second stage of the reaction. The energy of the evaporated nucleons is determined mainly by the temperature reached by the residual nucleus in the primary collisions and will be in the range of a few MeV.

To describe the full interaction of a relativistic projectile with a target material we should recall that the most probable interaction with the target material will be governed by electromagnetic processes. The main consequence of this electromagnetic interaction will be the slowing down of the projectile and the heat load of the target.

The nuclear interaction between the projectile and the target is determined by the total nuclear reaction cross-section, which for collisions induced by 1 GeV protons on lead corresponds to a mean free path of protons in lead of about 15 cm. In contrast, the mean free path for electromagnetic interaction is much shorter, consequently the incoming projectile will be slowed down before any nuclear interaction. The electromagnetic interaction is characterized in terms of the range of the incoming particle in the traversed medium. The range of a proton with 1 GeV in lead is around 55 cm.

The internuclear cascade inside the target will be determined by the energy balance of the interaction of the projectile with the target. Considering that on average the nuclear interactions take place at 15 cm inside the target, the mean energy loss of the incoming particle before the reaction will be 200 MeV. In addition the energy dissipated in the first spallation reaction is around 200 MeV. This excitation energy leads to a large population of different residual nuclei. The remaining kinetic energy ($\approx$ 600 MeV) will be shared between about five prompt nucleons emitted during the first stage of the reaction. The prompt neutrons will lead mainly to secondary reactions in the target (internuclear cascade). Consequently, a 1 GeV proton impinging on a lead target will induce on average two spallation reactions. The first one at high energy will determine mostly the residual nuclei produced in the target. The second reaction at lower energy ($\approx$ 200 MeV) will produce residues very close in mass and atomic number to the target nucleus, but will contribute to the multiplication of the neutrons and soften their spectrum.

6.3 Neutron production in spallation reactions

The neutron fluxes produced in spallation reactions will depend strongly on the projectile-target combination. In principle, the heavier the target nucleus the larger the neutron excess and the larger the neutron yield. The gain factor between heavy and light targets is around a factor of five. However, the radioactivity induced in the spallation target could be drastically reduced when using lighter targets [2]. In addition to the neutron yields, reliable information on the energy and spatial distributions of the neutrons is required. Different experimental devices are needed to characterize the neutron production in spallation reactions.

6.3.1 Measurement of neutron yields

Neutron multiplicities can be investigated using liquid-scintillator based detectors with a large angular acceptance. Clear examples are the detectors BNB (Berlin Neutron Ball) [3] and ORION [4] used by the NESSI collaboration (Berlin–GANIL–Jülich). This collaboration has performed a large experimental programme to determine the neutron yields produced in thin and thick targets for a large range
of primary projectiles and energies. To fulfil this programme experiments were done at GANIL (France) [4], COSY (Germany) [3] and CERN [5].

Figure 6.2 shows representative results obtained by the NESSI collaboration at Jülich with the BNB detector. This figure shows the measured average neutron multiplicity per incident proton as a function of the target thickness and beam energy for Pb, Hg and W as target materials. For the different target materials, the neutron multiplicity saturates at a given target thickness which increases with the proton energy. This saturation, at about 30 cm, underlies the previous conclusion that an incident proton of 1 GeV originates on average two nuclear collisions.

![Figure 6.2: Average neutron multiplicity per incident proton as a function of target thickness and beam energy for Pb, Hg and W materials obtained by the NESSI collaboration [3].](image)

6.3.2 Energy and spatial distribution of neutrons

Specific experimental set-ups are needed to measure the spatial and energy distribution of the neutrons produced in spallation reactions. The experiments performed by the ‘transmutation’ collaboration at Saturne (France) constitute a clear example. These measurements use two different experimental techniques to cover the full energy range of the neutrons produced in the reaction. The detection of neutrons with energies below 400 MeV was based on a measurement of their time of flight: the time difference between the incident proton, tagged by a plastic scintillator, and a signal from a neutron-sensitive liquid scintillator [6]. Neutrons with higher energies were measured using (n,p) scattering on a liquid-hydrogen converter and reconstruction of the proton trajectory in a magnetic spectrometer [7]. An additional collimation system allowed the angular distribution of the neutrons to be determined.

The neutron production in reactions induced by protons with energies between 0.8 and 1.6 GeV on thin and thick lead targets was investigated [8]. Figure 6.3 shows the results obtained on a two centimetre thick lead target with a 1.2 GeV proton beam, characterizing the spallation process to be applied in an ADS. High-energy neutrons emitted at low angles are representative of the first stage of the collision, while low-energy neutrons emitted isotropically correspond to the evaporation phase. Measurements done with thicker targets will provide information about the internuclear cascade.

6.4 Residue production in spallation reactions

Residue production in spallation reactions can be investigated using two different experimental approaches. In the standard one, the reaction is induced in direct kinematics and the light energetic projectile hits a heavy target. In this case, the recoil velocity of the residues produced in the reaction is not sufficient to make them leave the target and γ-spectroscopy or mass spectrometry techniques are used to identify those residues. The main limitation of this technique is that for most of the residues the measurement is done after β-decay and consequently only isobaric identification is possible.

Better suited to the measurement of the spallation residues is the investigation of the reaction in inverse kinematics. In this case the heavy nucleus is accelerated to relativistic energies and impinges on a light target. Because of the
kinematical conditions, the reaction residues leave the target easily and using the appropriate technique can be identified in a short time.

6.4.1 Measurement of residue production in inverse kinematics

Outstanding experiments have been performed by a German–French–Spanish collaboration at GSI (Germany). The technique used in these experiments takes advantage of the inverse kinematics and gives the full identification in mass and atomic number of the reaction residues as well as their velocities.

The experiments have been performed at the SIS synchrotron at GSI. Primary beams of $^{197}$Au, $^{208}$Pb and $^{238}$U accelerated up to an energy of 1 A GeV impinged on a liquid hydrogen or deuterium target. At this energy all residues of the reaction are predominantly fully stripped, bare ions. The achromatic high-resolution magnetic spectrometer FRS [11] equipped with an energy degrader, two position-sensitive scintillators and a multisampling ionization chamber allowed the identification in atomic and mass number of all the reaction residues with half-lives longer than 200 ns. Resolving powers of $A = \Delta A \approx 400$ and $Z = \Delta Z \approx 150$ were achieved with this technique. The final production cross-sections are evaluated with an accuracy not far from 10%. In addition, the high resolving power of the magnetic spectrometer allows the recoil velocity of the reaction residues to be determined. This information is relevant for the characterization of the damage induced by the radiation in the accelerator window or the structural materials. More details about these experiments can be found in Refs. [12]–[15].

In Fig. 6.4, all residues measured in the reaction $^{208}$Pb (1 A GeV) + p are presented on top of a chart of the nuclides. More than 850 different nuclei were identified in this reaction. As can be seen in this figure, the spallation residues populate two different regions of the chart of the nuclides. The upper region corresponds to the spallation–evaporation residues which populate the so-called evaporation–residue corridor. The second region corresponds to medium-mass residues produced in spallation-fission reactions. Both reaction mechanisms, fission and evaporation, inherently different, contribute to the production of residues.

The measured isotopic production cross-sections for some selected elements are presented in Fig. 6.5. This figure shows clearly the quality of the measured data that can be used to benchmark any model calculation.

Figure 6.6 shows the average kinetic energy in the centre-of-mass frame of fragmentation (closed symbols) and fission (open symbols) residues produced in the reaction $^{208}$Pb + p at 1 A GeV as a function of their atomic number [15]. These results clearly show an increase of the recoil velocity of the fragmentation residues for the most violent collisions leading to the production of lighter residues. The large kinetic energies of the fission residues are a key parameter in evaluating the heat load of the spallation target.
Figure 6.4: Two-dimensional cluster plot of the isotopic production cross-sections of all the spallation residues measured at GSI in the reaction $^{208}\text{Pb} (1 \ \text{A GeV}) + p$ shown on top of a chart of the nuclides [15].

Figure 6.5: Isotopic production cross-sections for some of the elements produced in the reactions $^{208}\text{Pb} + p$ at 1 A GeV measured at GSI [15]. The data are compared with two model calculations: the red lines correspond to the results obtained with the Lahet code [16] while the green lines were obtained with the intranuclear cascade model of Cugnon [10] combined with the evaporation–fission code ABLA from GSI [17, 18].
6.4.2 Measurement of residue production in direct kinematics

Gamma-ray spectroscopy allows the production of spallation residues in direct kinematic reactions to be investigated. Although this method is restricted to isobaric identification after $\beta$-decay, it is possible for some shielded isotopes to determine their primary production cross-sections. This technique consumes less beam time than the inverse kinematics method. Therefore, full excitation functions can be established for selected isotopes, as shown in Fig. 6.7. The method can also be applied to thin and thick targets at low and high energies, and in this sense it is complementary to the inverse kinematics technique.

At present, two main experimental programmes in Europe use this technique. The group of R. Michel at the University of Hannover performed experiments mainly at Saturne (France) and PSI (Switzerland). In these experiments different target materials were irradiated with protons in the energy range 20–2000 MeV [19]. Figure 6.7 shows representative results of these investigations. In this figure, excitation functions of the production of some shielded isotopes in collisions induced by protons on lead are shown [20]. These results clearly indicate that the low-energy reactions produce mainly residues close to the target nucleus, while most of the reaction residues further away from the target are produced by energetic particles. Similar experiments are also performed at the ITEP in Moscow [21].

6.5 Reactions in 20–200 MeV energy range

Reactions induced by neutrons and light-charged particles in the energy range 20–200 MeV are representative of the internuclear cascade in the spallation target. As discussed, these reactions play a major role in the multiplication and moderation of the neutrons. The energy dissipated in the target nucleus leads to the emission of few nucleons and consequently only to residual nuclei close in mass and atomic number to the target material.

The experiments performed in this energy range intend to measure the double-differential production cross-sections of neutrons and light-charged particles in reactions induced by protons and neutrons with energies in the range 20–200 MeV. Fission is also investigated. Measurements of the total fission cross-sections and mass distributions of fission residues for specific targets are also in progress. These measurements will allow ENDF libraries to be extended up to 200 MeV.

Many European laboratories contribute to this experimental programme, which takes advantage of a large network of European facilities delivering protons and neutrons in the investigated energy range: KVI (Netherlands), Louvain la Neuve (Belgium) and Uppsala (Sweden). Most of them contribute to the HINDAS project of the fifth framework programme of the European Commission [22]. This European project also includes the measurements of residue production in inverse kinematics done at GSI and light charged-particle production experiments in COSY, both in the 200–2000 MeV region.

Another important programme supported by the European Commission is the n_TOF project, based on a time-of-flight neutron facility recently set up at CERN. After moderation, the neutrons produced in a spallation lead target fed with 20 GeV protons from the PS cover an energy range between 1 eV and 200 MeV [23].
The experimental programme foreseen for this facility intends to cover a large number of experiments related to neutron capture and neutron-induced fission reactions. The expected neutron fluxes will allow radioactive targets to be used for some of these measurements. Results from these experiments are expected in the coming years.

### 6.6 Model simulations

The primary interaction of relativistic protons in a spallation target is mainly described in terms of semiclassical nucleon–nucleon collisions (intraneutron cascade) followed by the statistical de-excitation of the hot residue. The main inputs of the intraneutron cascade (INC) are the elastic and inelastic nucleon–nucleon cross-sections and the distribution in position and momentum space of the nucleons inside the target nucleus. The statistical evaporation of particles is generally based on the Weisskopf formalism, while fission should be described considering dissipation effects leading to the hindrance of that channel at high excitation energy. In this case the main parameters are the description of the level densities, the barriers for charged-particle emission and the fission barriers. Another critical parameter is the coupling time between the intraneutron cascade and the de-excitation stage.

The internuclear cascade in the target assembly will lead to reactions induced by light particles, mainly neutrons, at energies below 200 MeV. At these energies the semi-classical approach of the intraneutron cascade is no longer valid and codes based on pre-equilibrium models and direct reactions are mainly used.
The last model intercomparison done by NEA [24] revealed important deficiencies in most of the existing codes for describing spallation reactions. In fact these deficiencies can be explained by the lack of high-quality data when the codes were developed. The new data provided by the present experimental programmes will help to improve this situation. In Fig. 6.5, the measured isotopic production cross-sections for some of the elements produced in the reactions $^{208}$Pb + p at 1 A GeV at GSI [15] are compared with two model calculations. In this figure, the red lines correspond to the results obtained with the Lahet code (Bertini + Dresner) [16], while the green lines were obtained with the intranuclear cascade model of Cugnon [10] coupled to the new evaporation–fission code ABLA from GSI [17, 18]. As can be seen, the new INC and evaporation models provide a much better description of the experimental data.

Similar improvements are obtained with the new codes developed in the energy region below 200 MeV. The TALYS computer code system [25] created by NRG Peten and CEA Bruyères-le-Châtel represents one of the most promising tools including nuclear models for direct, compound, pre-equilibrium and fission reactions.

6.7 Outlook

The present experimental programmes investigating the nature of spallation reactions are expected to provide accurate data on both neutron and residual nuclei production. Although the high-energy interaction of protons with lead has been intensively investigated in recent years, experiments at lower energies are still in progress. In that sense, programmes like HINDAS or n_TOF will provide interesting data in the coming years.

The new sets of data will allow ENDF libraries to be extended up to 200 MeV but will also contribute to the development of new simulation codes. These new codes and libraries will constitute a key input in the design of an ADS target or window. In particular, target activity, radiation damage or gas production could be calculated with high accuracy. In order to accomplish these aims coordination programmes between the different communities contributing to the design of an ADS are in progress, like the French GEDEON project on a national scale or the BASTRA cluster in a European context.
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PART 2: LIFE SCIENCES

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1. Introduction

G. Kraft

The development of life science has always been strongly interconnected with physical science. This interaction has occurred in various ways, including the exchange of people, ideas, machines and techniques. For instance, the use in biology and medicine of new techniques and apparatus originating from physical science (microscopes, X-ray machines, centrifuges, computer homographs, etc.) becomes dramatically apparent if a power failure cuts off all the high- and low-tech facilities in a biology institute or hospital. But the connection between life science and physical science was more than just a simple exchange of sophisticated machines: It was also a vivid exchange of ideas and knowledge. One of the most outstanding and successful examples was the application of the knowledge and methodology of quantum physics to biology by Erwin Schrödinger in his book ‘What is life?’, in which he analyses radiobiology experiments in terms of quantum physics [1]. Schrödinger was the first to postulate the existence of genes as biochemical entities and was able to estimate their size from the radiation response within reasonable physical uncertainties. It was the influence of his ideas that led James Watson and his colleague Francis Crick to the study of genetics and to the discovery of the molecular structures of DNA.

Inversely, the stimulation of physics research by a broad interest of medicine was prompted by W.C. Röntgen’s application of X-rays. The primary article on the ‘new kind of radiation’, published in some proceedings of the Wuerzburg Medical-Physical Society, would not have received worldwide attention within a few weeks without the parallel publication of the medical application, i.e. the X-ray photography of the hand of Röntgen’s wife, in many newspapers [2]. Although the physical nature of the new radiation was not known at the time — Röntgen named them X-rays after the X used in mathematical equations for the unknown — X-rays were applied all over the world within a few days of publication for medical diagnosis and later on also for therapy. This application stimulated the physical research effort to understand the nature and action of the ‘unknown’ X-rays.

However, more common than the interest of different people in the same subject was the opposite, i.e. the broad interest of one person in different fields. In this sense, it is quite remarkable that the first radiobiological experiments were performed by Pierre Curie. He tested whether the skin inflammation and ulceration found by H. Becquerel under a pocket containing the newly discovered radioactive material was caused by radioactivity. Curie confirmed Becquerel’s findings and extended his biological experiments with radioactivity. At the beginning of the last century, he reported to the French Academy of Science anomalies in tadpole embryos that had been exposed to radium emanation, i.e. to radon. This was the very first experiment on the influence of ionizing radiation on embryonic development and became important much later, after the bombing of Hiroshima and Nagasaki.

A very powerful tool for biology can be attributed to G. de Hevesey, who was the first to use natural, and later on artificial, radioactive elements as tracers in order to study the pathway of stable elements in biological systems. It is frequently reported that Hevesey invented the tracer principle when he added some radioactivity to the leftovers of a dinner and tested his goulash soup with a Geiger counter in the same restaurant the next day [3]. Although it is impossible to find reliable confirmation of this anecdote in Hevesey’s publications, it illustrates
very well the tracer principle. An amount of radioactive elements — much less than is detectable by chemical analysis — can easily be followed in biological systems such as a human body or a plant.

In the biology of today, the analysis of DNA uses large quantities of radioactive isotopes for sequencing. With radioactive isotopes, the biomolecules are labelled and their decay is detected with a photographic film after the separation of complex DNA fragments on a gel. Radioactively labelled molecules have also been used for the elucidation of the photosynthesis of plants or for diagnostic purposes in nuclear medicine.

In nuclear medicine, a large variety of labelled compounds have been developed specifically for almost each individual organ in the human body. Measuring uptake and secretion, it is possible to determine the organs' functions. As it is also possible to label tumours with specific drugs, tumours can be located by Single Photon Emission Computed Tomography (SPECT), as well as by Positron Emission Tomography (PET) in which positron-emitting isotopes that, for instance, attach preferably to active brain areas are used. SPECT and PET are standard in tumour diagnostics as well as in function studies of the normal healthy brain.

Today, more and more of these measurements of organ function and of diagnostics can be performed with Nuclear Magnetic Resonance Imaging (MRI). The big advantage of MRI is that no radioactivity need be used and the magnetic field applied to the patient does not produce any damage in the tissue. Thus, these measurements can be extended and frequently repeated if necessary.

Another example of the use of stable isotopes instead of radioactive material is Accelerator Mass Spectroscopy (AMS). Because of their high sensitivity to isotopic distribution, accelerators can be used to detect the tiniest amounts of exotic but stable isotopes. This technique can be employed to follow biological pathways in the body without any risk for the patient.

Of the radioactive elements, iodine plays an exceptional role in medicine because it attaches to thyroid tissue with an extreme affinity and selectivity. Therefore, it can be used in low doses for diagnostic purposes, and a thyroid cancer can be destroyed by ionizing radiation emitted from radioactive iodine exclusively attached to the thyroid tissue. Iodine also attaches to and kills metastases of thyroid cancer that are dispersed throughout the body.

This exceptional affinity of an element or molecule to a malignant cell represents an optimum model for tumour therapy because these ‘cell seeking’ drugs obviously conform the dose to the tumour volume in a perfect way. This is not the case for other deep-seated and inoperable tumours, which have to be treated with external radioactive sources or other ionizing radiation. Using electromagnetic radiation like the most advanced Röntgen-bremsstrahlung from high-energy electron linearis, a reasonable amount of dose is always deposited in front of and behind the target volume in the healthy tissue. Therefore, the tumour dose is mostly limited by the tolerance of the surrounding normal tissue.

An essential improvement in external radiation therapy can be achieved using high-energy beams of heavy particles like protons or carbon ions. Ion beams exhibit an inverted depth-dose profile: the dose increases with penetration depth up to a maximum at the end of the particle range. In addition, for ions heavier than protons, like carbon, the radiobiological efficiency increases towards the end of the particle range. Thus, ion beam therapy allows a higher and more effective dose to be deposited in the tumour than any other type of external therapy. Up to now, more than 30,000 patients have been treated with protons worldwide and more than 1000 patients with heavy ions. Together, they reach an outstanding tumour control rate. As a result, numerous initiatives for the promotion of ion beam therapy have emerged in many countries in recent years.

Table 1.1 outlines a brief history of the application of nuclear techniques in life science. It is remarkable to see how many techniques in nuclear medicine, in addition to ion beam
1. Introduction

<table>
<thead>
<tr>
<th>Year</th>
<th>Invention</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>1895</td>
<td>W.C. Röntgen</td>
<td>X-rays</td>
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<tr>
<td>1896</td>
<td>H. Becquerel</td>
<td>Natural radioactivity</td>
</tr>
<tr>
<td>1898</td>
<td>M. and P. Curie</td>
<td>Radium</td>
</tr>
<tr>
<td>1923</td>
<td>G. de Hevesey</td>
<td>First radiobiological experiments, application of radium in therapy</td>
</tr>
<tr>
<td>1927</td>
<td>Blumgart/Weiss</td>
<td>Tracer principle</td>
</tr>
<tr>
<td>1931</td>
<td>E.O. Lawrence</td>
<td>Cyclotron</td>
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<td>1934</td>
<td>E. Fermi</td>
<td>128-iodine</td>
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<tr>
<td>1937</td>
<td>R. Stone</td>
<td>Neutron therapy</td>
</tr>
<tr>
<td>1938</td>
<td>Hertz, Roberts, Evans</td>
<td>Thyroid studies with iodine</td>
</tr>
<tr>
<td>1939</td>
<td>J. Lawrence</td>
<td>Artificial radioisotopes for therapy</td>
</tr>
<tr>
<td>1942</td>
<td>Hertz and Roberts</td>
<td>Treatment of thyroid hyperfunction</td>
</tr>
<tr>
<td>1946</td>
<td>R. Wilson</td>
<td>Proposed proton and carbon therapy</td>
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<tr>
<td>1951</td>
<td>Wrenn/Brownell, Sweet</td>
<td>Positron emission tomography</td>
</tr>
<tr>
<td>1954</td>
<td>J. Lawrence</td>
<td>Proton therapy</td>
</tr>
<tr>
<td>1958</td>
<td>H. Anger</td>
<td>Scintillation camera</td>
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<tr>
<td>1970s</td>
<td>G. Hounsfield/A. Cormack</td>
<td>Computer-assisted tomography</td>
</tr>
<tr>
<td>1972</td>
<td>Damadian</td>
<td>Patent of NMR</td>
</tr>
<tr>
<td>1974</td>
<td>C.A. Tobias/J. Lawrence</td>
<td>Heavy ion therapy</td>
</tr>
<tr>
<td>1997</td>
<td>GSI/PSI</td>
<td>Beam scanning and tumour-conform ion treatment</td>
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</table>

Therapy, originate from Berkeley. Ernest O. Lawrence, inventor of the cyclotron and founder of the Berkeley laboratories had a brother, John Lawrence, who was a physician. Therefore, the medical and biological applications of isotopes and beams were pushed from both sides — nuclear physics and medicine. In addition, the Lawrence brothers' success is a remarkable example of close personal relations between two outstanding scientists being much more effective than any administrative regulation.

It also shows that science is driven by individuals and justifies the connection between people and projects as given in Table 1.1. Of course, not all projects in this table can be discussed here in detail. It is the purpose of this part of the report to give some examples of the positive impact of nuclear science on life science. The selection of topics is somewhat arbitrary and surely influenced by personal affinities. We have tried to give a stimulating overview and to demonstrate the benefits of nuclear science but also the problems regarding the application of ionizing radiation.

The risk of the application of nuclear methods using ionizing radiation is summarized in Chapter 12. Most of the available data originate from the Nagasaki and Hiroshima victims and from patients treated too extensively with diagnostic procedures using ionizing radiation. Moreover, the results from the Chernobyl accident were analysed. A low incidence of genetic mutations and of cancer induction is reported. That is why these data have large error bars. However, the confidence limits are narrow enough to calculate solid values for the risks associated with the application of nuclear techniques. This is very important because the benefit of a procedure has always to be weighed up against its potential disadvantage, as is the case for other techniques used in everyday life.

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2. Nuclear Magnetic Resonance

M.O. Leach

2.1 Introduction

Nuclear magnetism was detected in solid hydrogen by Lazarew and Schubnikow in 1937. The first observations in bulk material were reported independently by Bloch and by Purcell in 1946. This provided insight into the properties of nuclei, with different nuclei having characteristic resonant behaviour. The first in vivo observation of a nuclear magnetic resonance (NMR) signal occurred shortly afterwards, when a strong hydrogen signal was observed from Bloch’s finger. While remaining an important means of probing nuclear structure, it was soon found that NMR spectra contained characteristic information on molecular conformation, and a major role for NMR developed in chemical analysis and in the study of molecular binding. In the 1970s, development of larger superconducting magnets allowed these techniques to be extended to the non-invasive investigation of tissue biochemistry.

The potential of using NMR for imaging hydrogen atoms was first proposed by Lauterbur, based on spatially localizing signals by adding small magnetic-field gradients, which changed strength linearly with spatial displacement, to the homogeneous main magnetic field. Images could be reconstructed from sets of projections, in much the same way as images were reconstructed in X-ray computerized tomography (CT). This approach was used in the first commercial MR system using a superconducting magnet. In parallel, Mansfield developed imaging approaches from a background of solid-state physics, where techniques based on crystallographic principles could be applied to sample signals spatially. This approach led to the development of echo planar imaging (EPI), allowing a full image to be acquired in a single excitation. Further developments, particularly in research groups at the Universities of Nottingham and Aberdeen, saw development of a wide range of imaging methods, which eventually culminated in the modification of Kumar’s Fourier zeugmatography technique by Edelstein to provide the spin-warp imaging technique which is now almost universally used. As well as progressive development of imaging methods, there has been a continuing development in magnet design, with superconducting magnet designs providing considerable stability and allowing increases in field strength up to a current maximum of 8 T for whole-body systems. Recently a range of electromagnets and permanent magnets has been developed to address growing low-field requirements. In clinical applications, NMR imaging is commonly referred to as magnetic resonance imaging (MRI).

In the 1980s the first clinical NMR spectroscopy instruments were developed, using high field (1.5 T or greater) magnets with specialized hardware. As clinical MRI systems at 1.5 T became widespread, magnetic resonance spectroscopy (MRS) used similar techniques to MRI for spatial localization and was incorporated in to some commercial MR systems, requiring broad band capability to allow $^{31}$P, $^1$H, $^{19}$F and $^{13}$C MRS studies. Recent developments have involved integrated studies combining imaging and spectroscopy, often using specialized radiofrequency coils and acquisition methods. Either single voxel acquisitions, or 2D or 3D spectroscopic imaging methods, where an MR spectrum is obtained in each of an array of spatially localized voxels, are employed.
2.2 Applications of magnetic resonance imaging and spectroscopy

MRI has rapidly developed to provide measurement methods that can address a wide range of clinical problems in diagnosis. The scope of these methods, and the continued rate of development, has been unprecedented in clinical imaging. MRI measures the distribution of hydrogen atoms in the body, with the signal (or image contrast) being affected by the freedom of molecules to rotate or move (relaxation times) or by the way in which the signal is manipulated by the MR pulse sequences. These control radio-frequency pulses, magnetic field gradient switching, and signal detection. Cross-sectional images can be obtained directly in any orientation through the body, aiding detection and coverage of specific anatomy. Usually a series of 2D images is acquired, but complete 3D volume images can also be obtained, allowing retrospective reconstruction of any plane, or indeed non-planar surfaces that follow a particular structure. While a set of images usually takes a few seconds or minutes to acquire, images can be obtained in 50 ms using fast imaging techniques. MRI provides unrivalled soft tissue contrast, allowing pathology to be identified, and normal anatomy to be displayed with excellent definition of tissue structures. Bone is not seen directly, as hydrogen atoms in hydroxyapatite are bound too tightly to give a signal detectable on clinical apparatus. Flowing blood provides a signal that can be manipulated to allow MR angiography — images specifically showing vascular structures. In addition to the measurement of the intrinsic signal from tissues, a range of MR contrast agents is under development. These are compounds with intrinsic magnetic properties that affect the relaxation properties of hydrogen atoms. Gadolinium, with seven unpaired electrons, has a very strong electronic magnetic moment, and is used in the form of a chelate such as DTPA. In the form of Gd DTPA, the contrast agent has a major use in identifying areas where the blood–brain barrier, which protects neuronal cells from blood-borne toxins, is broken down. This is a common feature of lesions in the brain. Tumours elsewhere in the body also exhibit high contrast uptake. Other contrast agents utilize ferromagnetic or super-paramagnetic properties, and a range of new applications is developing, including imaging gene function.

The first major application of MRI was in imaging brain disease, where the soft tissue contrast rapidly eclipsed CT. Figure 2.1 shows examples of brain images, showing how contrast can be altered by changing the measurement sequence. More recently, angiography studies have allowed identification of arteriovenous malformations, carotid artery stenosis and, increasingly, stroke. MRI can measure the self-diffusion of water, and the restricted diffusion coefficient is a sensitive measure of the early impact of stroke. It is being used to evaluate protective interventions. Brain perfusion can also be assessed using a range of specialized quantitative imaging techniques. Related methods can be used to measure the rate of transport of contrast agents through the vascular endothelium, providing information on delivery and permeability. A major advance was the discovery that some MRI sequences were sensitive to brain activation — the changes in blood oxygenation and local perfusion that result from the increased energy demands of neurological activity. This is a powerful tool in neurological and psychiatric research, providing high-resolution maps of activity that can be superimposed on excellent images of brain structure. $^1$H MRS can be used to characterize different types of brain cancer, promising a non-invasive means of diagnosis and staging. $^{31}$P MRS has been of value in the evaluation of neonates, allowing identification of brain asphyxia. MRI is also the method of choice for investigating spinal cord disease, where excellent contrast between nerve and cerebrospinal fluid can be obtained without the invasive procedures associated with X-ray myelography.

Orthopaedic applications are another major area of development. Here 3D imaging techniques provide excellent high-resolution images of joints and the associated soft tissues and synovial fluid, allowing investigation of meniscal tears, cartilage injuries, degenerative disease and sports injuries. A new generation of low-field open magnets, allowing imaging of
Figure 2.1: Axial images of a patient with a low-grade glioma. (a) A T1 weighted image showing the tumour as a dark region relative to the brain because of its longer T1. Grey matter appears darker than white matter, and fluid (in the eyes and cerebrospinal fluid) is also dark owing to a very long T1 relaxation time. Bright areas are fat. (b) T2 weighted images showing areas of tumour as bright because of their long T2 relaxation times. Fluid is also bright, and grey matter appears brighter than white matter in the brain. Fat has relatively low signal. (c) FLAIR (fluid attenuated inversion recovery) T2 weighted image. Contrast is similar to (b), but signal from fluid has been nullified by applying an appropriate inversion pulse and recovery period. This allows margins of tumour that might be obscured by bright cerebrospinal fluid signal to be more easily detected.
movement, has increased the availability of these approaches. Peripheral angiography allows whole body surveys of vascular patency. Recently, methods for surveying whole body marrow status, allowing identification of metastatic disease from cancer, have been demonstrated. Figure 2.2 shows an example of a coronal slice through the tibia of a ten-year-old patient, showing a Ewing’s sarcoma in the marrow cavity, extending outside of the bone.

Cardiac measurements have been the focus of considerable attention, leading to the development of techniques for imaging coronary arteries, heart valve function, cardiac muscle perfusion and metabolism (using $^{31}$P spectroscopy), cardiac ejection rate and cardiac wall motion. This combination of high-resolution imaging and cardiac function measurement provides an exceptionally powerful tool. Figure 2.3 is a coronal image through the chest, showing the heart. Figure 2.4 provides an example of body imaging in the abdomen, showing a sagittal view through a cervical tumour. Lung measurements have until recently been a major limitation of MRI. Rapid imaging is now overcoming the loss of signal due to air–tissue interfaces, allowing measurement of lung perfusion and the identification of pulmonary emboli. A new development has been the application of hyperpolarized gases ($^3$He and $^{129}$Xe), where the spin population of the gases can be pumped to give very high polarizations, allowing the gas to be imaged in lung spaces, with applications in chronic obstructive lung disease and in asthma.

In some cases in vivo MRS can be used to directly monitor drug pharmacokinetics and metabolism in tumour and tissues. In these cases the primary drug is identified due to the inclusion of an NMR sensitive atom, such as $^{19}$F (e.g. 5-fluorouracil) or $^{31}$P (ifosfamide or cyclophosphamide). Figure 2.5 shows an example of a set of localized $^{19}$F spectra from a patient receiving 5-fluorouracil, superimposed on a proton image of the liver. The localized spectra showed that the signal was all concentrated in the gall bladder (bright on the proton image), and the frequency of the observed peak showed that the compound observed was a bile acid conjugate of fluoro-beta-alanine, a catabolite of 5-fluorouracil. In other cases an isotopically enriched label such as $^{13}$C can be substituted for a non-NMR sensitive high abundance atom ($^{12}$C).
2. Nuclear Magnetic Resonance

Figure 2.3: A coronal section through the chest, showing the heart, liver and lung cavity. The image is T2 weighted (turbo spin echo) and was obtained during a breath hold to avoid respiratory motion.

Figure 2.4: A sagittal T2 weighted turbo spin echo through the abdomen of a patient with a cervical tumour. The tumour is the large dark area between the sacrum (shown by the relatively bright marrow in the vertebral bodies) and the cervix (above the very bright bladder).

Figure 2.5: An array of $^{19}$F spectra obtained using 2D chemical shift imaging from a patient receiving 5-fluorouracil chemotherapy. Significant signal is only found in one voxel, centred over the gall bladder (bright on the underlying proton image through the liver). The chemical shift of the peak identified it as the bile acid conjugated form of fluorobeta-alanine, confirming hepatobiliary recirculation of this catabolite. (Reproduced with permission from Dzik-Jurasz et al., Magn. Reson. Med. 44 (2000) 516.)

Figure 2.6: A 3D $^{31}$P chemical shift imaging examination of a patient receiving treatment for a sarcoma. A plane of the spectral data is shown superimposed on a proton image of the same tissue slice. The spectrum from the voxel indicated is enlarged to show the characteristic tumour spectrum. PME: phosphomonoester, PE: phosphoethanolamine, PC: phosphocholine, P: inorganic phosphate, PCR: phosphocholine, NTP: nucleotide tri-phosphate.
The major advantage of MRS is that the chemical form of the parent compound can be observed, and in many cases the metabolism of the drug to active or non-active compounds can be simultaneously identified in real time. This is a unique advantage of MRS compared with other imaging methods such as positron emission tomography (PET). Figure 2.6 shows an example of a proton decoupled $^{31}$P MRS spectrum from a sarcoma, showing a characteristic tumour spectrum, and the separation of the characteristic phosphomonoester (PME) signal into phosphocholine and phosphoethanolamine signals. A proton localization image is also shown, with the superimposed chemical shift imaging spectra. Measuring the metabolites in the PME peak gives information on tumour prognosis and response.

MRI measurements can also provide information about the properties of the blood supply to tumours. By employing contrast agents and using rapid imaging techniques, images can be obtained where the contrast is either affected by the rapid magnetic susceptibility changes arising from the first pass of the concentrated bolus of contrast through the vasculature — providing information on blood volume and perfusion; or by the change in signal due to contrast leaking out of the vascular space into the extracellular space — providing information on contrast delivery and vascular permeability. These measurements are, for example, allowing the effects of new therapies directed at manipulating vascular function to be assessed, and they are also relevant to assessing drug delivery.

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3. Single Photon Emission Computed Tomography

R.J. Ott

Images of the biodistribution in the body of injected radiotracers created using a gamma camera (Fig. 3.1) can provide information about the function of tissues. Such 2D images, known as planar scintigrams, are degraded by the superposition of non-target activity from the 3D body, which restricts the measurement of organ function and prohibits accurate quantification of that function. Single photon emission computed tomography (SPECT) is a technique whereby cross-sectional images of tissue function can be produced allowing the removal of the effect of overlying and underlying radioactivity (Larsson 1980, Williams 1985, Croft 1986, Ott 1986).

Reconstruction using methods similar to those used in X-ray computed tomography (CT) provides 3D data sets allowing the tracer biodistribution to be displayed in orthogonal planes. The advantages of SPECT over planar scintigraphy can be seen in the improvement of contrast between regions of different function, better spatial localization, improved detection of abnormal function and, importantly, greatly improved quantification. In general SPECT images have poorer spatial resolution than the 2D images used to produce them.

Clinical applications of SPECT include evaluation of function of the brain, heart, skeleton, liver, lungs and kidneys.

Figure 3.1: Schematic diagram of the production of radionuclide images using a gamma camera.

SPECT involves the use of radioisotopes such as $^{99m}$Tc, where a single gamma-ray is emitted per nuclear disintegration. This is in contrast to positron emission tomography (PET) which makes use of radioisotopes such as $^{18}$F, where two gamma-rays, each of 511 keV, are emitted simultaneously when a positron from a nuclear disintegration annihilates in tissue. SPECT images are produced from multiple 2D projections by rotating one or more gamma cameras around the body to achieve complete 360° angular sampling of photons from the body.

In the brain SPECT is used to measure cerebral blood flow and brain patency in patients with stroke and tumour. Figure 3.2 shows the uptake of the blood flow tracer $^{99m}$Tc Ceretech in the normal brain, indicating the differential blood flow in grey and white matter which is known to be $\sim 4 : 1$. In a patient with reduced blood flow after stroke, the size of the abnormal region and the level of residual blood flow may help to differentiate between ischaemic and infarcted tissue.

Figure 3.2: Transaxial sections through a $^{99m}$Tc Ceretech scan of a normal brain.
viability of the heart muscle to help differentiate between ischaemia and infarction. Both these tracers mimic the perfusion of the left ventricle and the use of SPECT highlights regions of poor flow during stress. These regions of the heart will reperfuse post-stress if the tissue is ischaemic rather than infarcted. Figure 3.4 shows uptake in the left ventricle of a normal heart (bottom) and in a heart showing poor perfusion in the posterior wall of the ventricle (top). This type of information is important in aiding the treatment of patients with cardio-vascular disease — especially to determine if a bypass grafting is warranted for those patients with ischaemic disease, in contrast to transplant which might be indicated for the seriously infarcted heart. These studies are also used in patients with suspected stenosis, hypoxia, aneurism or cardiomyopathy.

Skeletal radioisotope imaging is most often performed as anterior/posterior whole-body scans as the tracer used, $^{99m}$Tc MDP, provides excellent contrast for 2D imaging. However, there are many areas where 2D contrast is insufficient to make a diagnosis and SPECT can then help the diagnosis by providing the necessary improved contrast. Radioisotope imaging is indicated for a wide range of skeletal diseases including Paget’s disease, spondylolisthesis, sacroiliitis, ankylosing spondylitis, avascular necrosis, metastases, synovitis, arthritis and osteomyelitis. Whilst the images are not specific to any particular skeletal disease the sensitivity is excellent. Figure 3.5 shows how SPECT can provide accurate anatomical location within the vertebra of a patient with a suspected stress fracture. Similar results can be obtained if the patient has metastatic disease in the skeleton. Such scans are used to stage the treatment of patients with breast or prostate cancer.

Liver disease can be imaged using SPECT to determine the existence of sarcoma, hepatic tumour, haemangionia, metastases, cyst, glycogen storage disease, Menetrier’s disease, etc. In Fig. 3.6 we see the uptake of $^{99m}$Tc colloid in the liver of a patient with suspected metastatic disease. This tracer is extracted by normal Kupfer cells in the liver. In the planar image there is an indication of reduced uptake of the colloid in
the right lobe of the liver. This is clearly delineated in the transaxial SPECT images which also show the existence of disease extending into the left lobe. The same tracer can be used to examine the status of the spleen which is shown on the right side of the images in Fig. 3.6. By using an alternative tracer such as labelled red-blood cells it is possible to visualize increased uptake in liver lesions, indicative of haemangioma.

In the lung, the most common radioisotope imaging is carried out to measure the ventilation and perfusion status of the lung using $^{99m}$Tc labelled tracers. These scans can be used to differentiate space-occupying disease from other pulmonary abnormalities such as obstructive airways or embolism. SPECT can sometimes help the diagnosis and can also be used to quantify treatment outcome.

Other applications include the evaluation of tumour in the lung to determine the perfusion status which may help treatment. Figures 3.7 and 3.8 show the uptake of $^{99m}$Tc Ceretech in the lungs of two patients with metastatic disease. In Fig. 3.7 the SPECT image shows high perfusion in a right-lung lesion which is barely visible in the planar scan.

Figure 3.8 shows the perfusion of a large tumour again in the right lung. In this case the lesion has a well perfused periphery with poor perfusion in the central region, possibly indicating necrosis. In both these patients the SPECT images provide a much more detailed picture of the functional status of the disease, which may well affect the way the patients are treated.
There are many other applications of SPECT which can help in the diagnosis of disease and in some cases be an aid to therapy. Targeted radionuclide therapy is a method of expanding interest for cancer treatment and SPECT has a very important role to play in quantifying radionuclide-tissue concentration, which cannot be achieved as well using planar imaging. An example of this is the use of SPECT to image the biodistribution of $^{123}$I-mIBG in patients with neural crest tumours such as childhood neuroblastoma (Fig. 3.9). In this case the images are used to determine if the uptake of the tracer in the tumour warrants treatment with $^{131}$I-mIBG. The quantitative information from the $^{123}$I-mIBG image can be used to estimate the activity of $^{131}$I-mIBG needed and subsequent $^{131}$I images can be used to estimate radiation dose to the tumour and normal tissues.

Figure 3.9: Transaxial SPECT image of the uptake of $^{123}$I-mIBG in an abdominal neuroblastoma.
4. Positron Emission Tomography

K. Wienhard

4.1 Introduction

Positron Emission Tomography (PET) is the most sensitive method to image trace amounts of molecules in vivo. Therefore this technique is used to measure in man or in the living animal biochemical and physiological processes in any organ with three-dimensional (3D) resolution. The last 25 years have seen a rapid and still ongoing development in the production of positron emitters, radiochemical labelling techniques, tomograph technology and image reconstruction algorithms. This is driven by the manifold research applications of PET in the study of human disease and its possible treatment. Because of the possibility to see and measure quantitatively physiological disorders in an early stage, before permanent morphological damage has occurred — which will only then be visible in X-ray or magnetic resonance computed tomography — PET is finally finding its way from a sophisticated research tool into routine clinical diagnosis.

4.2 The method

The use of positron emitters for radioactive labelling offers important advantages over single photon emitters. Some of the physiologically most interesting chemical elements like carbon, nitrogen and oxygen have only positron emitting short-lived isotopes. This makes a detection device which is sensitive to positron decay highly desirable. The positron travels only a short distance in tissue, then it annihilates with an electron into two 511 keV gamma-quanta or photons which are emitted back to back. When these two gamma-quanta are detected by two detectors in coincidence the decay event can be localized to the line connecting the two detectors. This 'electronic collimation' allows the construction of highly efficient detection systems because there is no need for heavy collimators. Since both photons have the same energy the detection probability is almost independent of the point between the two detectors where the decay event originated. Therefore the corrections for attenuation in tissue, which are generally huge in these measurements, can be done very accurately.

Commercia tomographs consist of thousands of detectors, which are arranged in many rings surrounding the object. Each detector is connected in coincidence with opposing detectors. First this was restricted to opposing detectors in the same ring or in neighbouring rings, with septa shields between the detector rings to eliminate unwanted scattered radiation. In order to increase sensitivity and make better use of the applied activity the septa were then made removable and coincidences between many rings were registered. Now the most advanced systems have no shielding at all between the detectors. The accepted solid angle is increased by extending the axial field of view with more detector rings, and tightly packed detectors with a high stopping power are arranged in a compact geometry. This 3D or volume data acquisition, where all possible coincidence lines inside the detector volume are registered, induces, besides higher count rates of true coincidences, a large increase of scatter coincidences and random coincidences. A scatter coincidence is a detected event where one or both photons were deflected by Compton scattering and are therefore assigned to a wrong coincidence line. Scattered coincidences may be as numerous as true events and very elaborate correction algorithms have become necessary to subtract scatter contributions for accurate quantitative results. A random coincidence is an event were two pho-
tons from different decays are detected during the short coincidence time window. Random coincidences can be subtracted online using a second delayed coincidence window.

4.3 New developments

From the outset it was a primary objective in PET to improve the spatial resolution of the tomographs. The early tomographs used NaJ(Tl) crystals as scintillation detectors. These were soon replaced by BGO (bismuth germanate) crystals which have higher detection efficiency and are not hygroscopic. Commercial tomographs using BGO reached a spatial resolution of approximately 4 mm. Today, the first tomographs with a new LSO (lutetium oxyorthosilicate) scintillator are being built. LSO has a similar high detection efficiency to BGO, but with a five-fold higher light output and an eight times faster light decay time. This allows spatial resolution to be pushed close to its physical limit of 1–2 mm, which is determined by the positron range in tissue and the small noncollinearity of the annihilation photons. Increased spatial resolution requires smaller scintillation crystals. When the size of the photomultiplier tubes (PMT) became the resolution limiting element, the one-to-one coupling between crystal and PMT was given up and an Anger camera principle was applied in the block detector concept. For example, 8 × 8 crystals were cut into a single scintillator block forming a light guide to four PMTs. By comparing the signals of the PMTs the individual crystals are uniquely identified. An even more cost-effective scheme, a quadrant sharing arrangement of PMTs (Fig. 4.1), allows the number of crystals per PMT to be further increased. This was quite recently realized in the first PET tomograph for neurological studies in humans with LSO scintillators: the High Resolution Research Tomograph (HRRT), manufactured by SIEMENS/CTI, Knoxville, TN, USA and installed at the Max-Planck-Institute for Neurological Research in Cologne. It consists of 120,000 single crystal elements, each 2.1 mm × 2.1 mm wide, arranged in 104 rings giving a reconstructed spatial resolution of less than 2.5 mm in a 20 cm diameter volume. This made it also necessary to measure the depth of interaction (DOI) of the incident photons in the 15 mm deep detectors to avoid ambiguities when photons penetrate several crystals by oblique incidence. From several ideas how to obtain DOI information, a scintillation phoswich with two crystal layers having different light decay times was chosen. Pulse shape discrimination allows then to separate scintillation events in the two layers.

![Figure 4.1: A LSO phoswich detector block on a set of four PMTs. The scintillator block consists of two LSO layers with different light decay times, which are cut into an 8 × 8 crystal matrix and glued on a light guide.](image)

After correction for scatter and random events, attenuation and dead time an image of the activity distribution can be reconstructed. Originally this was done with standard 2D filtered backprojection algorithms giving a stack of transverse image slices. With 3D data acquisition this was extended to volume reconstruction, which required hours of computing time. Therefore the algorithms were parallelized and run on multiple processors. Only recently, with the rapid developments in computer hardware, has iterative 3D reconstruction become routinely feasible. The PET images can then be superimposed on higher resolution images from other modalities like MRI (magnetic resonance imaging) to relate the information in the moderate resolution PET images to morphology.

4.4 Tracer production

The commonly used positron emitting nuclei in PET are $^{11}$C, $^{13}$N, $^{15}$O and $^{18}$F. The first three are isotopes of the most abundant elements in
organic compounds. This allows the labelling of naturally occurring biomolecules or drugs without altering the compounds' defined courses in living systems. $^{18}$F can be used to substitute hydrogen or hydroxyl groups. Because of their short half-lives, ranging from 2 min for $^{15}$O to 110 min for $^{18}$F, they have to be produced close to their application. Various small, compact cyclotrons specially constructed for the production of those isotopes in a hospital environment are commercially available. There are cyclotrons with external proton and deuteron beams, proton-only self-shielding machines with internal targets down to 3 MeV beam energy for the exclusive generation of $^{15}$O, and negative ion machines allowing simultaneous irradiations on separate target ports. They are equipped with targetry and remote systems for chemical precursor production like $^{11}$CO, $^{13}$CO, $^{13}$NH$_3$, $^{11}$CH$_4$ or H$_2^{15}$O in high yields of several curies. Computer-controlled automated robot systems synthesize in heavily shielded hot cells radiolabelled compounds in optimized short times of a few minutes, including purity controls. For $^{18}$F-labelled compounds, which can be shipped over considerable distances because of its 110 min half-life, a network of distribution centres has been built in several countries to deliver the radiotracers by car or plane to PET installations with no accelerator facility. This has contributed greatly to the recent expansion of PET in routine clinical diagnosis.

4.5 Applications

The study of physiological parameters and their pathological variation with PET has found broad clinical applications in neurological disease, brain and body tumours and cardiac tissue viability. In addition the unique possibilities of PET methodology are being used to investigate brain function in normal volunteers, to develop experimental models for various human diseases, to study the in vivo pharmacokinetics of drugs and the efficacy of treatments of disease. Several hundred molecules have been labelled with positron emitters, only a few of which have yet found their way to clinical application. Figure 4.2 gives a schematic overview of common tracers and some neurological applications.

Short-lived $^{15}$O is mainly used to label water for blood-flow studies or oxygen gas to study oxygen metabolism. Because of its short half-life $^{15}$O-water injections can be applied repeatedly at short time intervals to measure blood flow under various functional activation paradigms in normal or diseased brain (Fig. 4.3). The combination of quantitative values for cerebral blood flow (CBF), oxygen consumption (CMRO$_2$) and oxygen extraction (OEF) give important information about the pathophysiological status in the acute phase of cerebral ischemia and can guide stroke therapy with early reperfusion by thrombolysis (Fig. 4.4).

Fluorodeoxyglucose (FDG) has for more than 20 years been the most widely used PET tracer, and is now also supplied routinely from many distribution centres. Its uptake in tissue can be easily quantified as glucose metabolic rate with a stable autoradiographic model developed by Sokoloff, which needs in addition to a single PET measurement only the time course of radioactivity in arterial blood. It has been broadly applied to image and to quantify metabolic disturbances in cardiology, oncology and in almost every neurological disease or disorder.

The study of the dopaminergic system with $^{18}$F-L-DOPA and raclopride allows disturbances to be imaged on the transmitter and on the receptor side in movement disorders like Parkinson's disease. A labelled receptor ligand like raclopride enables the measurement of regional receptor densities or the observation of changes in endogenous dopamine levels caused by functional stimulations.

Flumazenil (FMZ) is a ligand binding to benzodiazepine receptors, which can be used to image neuronal integrity with applications for example in stroke and epilepsy.

With labelled amino acids, like methionine, amino acid transport, metabolism and protein synthesis can be studied under various conditions. Especially in brain tumours, amino-acid uptake in combination with FDG gives important information about tumour localization, extent and malignancy.
Figure 4.2: Overview of the PET method with brain images for common tracers in clinical applications.
This short overview of some clinical applications shows only a small share of the potential of the PET method. Promising future applications are emerging in drug development, psychopharmacology, visualization of gene therapy and in experimental research with animals.

Figure 4.3: Blood-flow changes under speech activation superposed on MRI brain volume. The location of the speech areas (red areas with increased blood flow) in relation to the brain tumour (shown in green) give important information to the neurosurgeon before surgical intervention.

Bibliography


Figure 4.4: Cuts through the brain volume from MRI overlaid with blood-flow information from PET in colour. The left image shows the infarcted region with decreased blood flow in blue. After trombolysis therapy blood flow has recovered, as shown in the right image.
5. New Advances in *in vivo* Small Animal Imaging

F. Pain and P. Lanièce

5.1 Introduction

The emergence of new animal models that mimic human disorders (neurodegenerative diseases, tumour growth, neuropsychiatric disorders) have enabled new fundamental and therapeutical approaches to these diseases. Mice, rats and guinea pigs have become progressively ubiquitous participants in most areas of molecular biology, toxicology and drug discovery research. Well-characterized models have been thus developed for a wide range of diseases in order to study their fundamental mechanisms and to test potential drugs. The mouse, in particular, has become a key animal model system for the study of development and human disease [1]–[4]. Its genome can be manipulated, allowing accurate models of many human disorders to be produced, resulting in significant progress in the understanding of human diseases. In neurobiology, the possibility to mimic neurodegenerative diseases with rat models has permitted the development and the assessment of new therapeutical strategies based on genetic therapy or cell grafts [5]–[8].

The increasing number of studies performed on animal models has stimulated the development of new imaging tools adapted to the particular constraints of *in vivo* studies in small animals. *Ex vivo* experiments such as autoradiography have indeed shown their limitations when subjected to longitudinal analyses required in the studies involving animal models. They are expensive, time consuming, and not very precise owing to the considerable inter-individual physiological variation. Extension of modern biomedical imaging techniques to the small animal has therefore presented some interesting challenges and opportunities. This has led to the emergence of dedicated systems including radiotomography and magnetic resonance imaging technologies. The aim of this article is to review briefly these systems and to illustrate their performance in biological contexts.

5.2 Radioimaging systems

By their principle, radioimaging techniques do not deliver anatomical images. Combined with specific radiotracers they provide metabolic data of different orders, examples of which include functional and oncological imaging with $^{18}$F-FDG [9]–[11], the evaluation of new radiopharmaceutical for diagnostic efficiency [12], the evaluation of new receptor ligand [13], and more recently the reporter gene expression imaging [14]. Since 1990, different instrumental approaches based on Single Photon Emission Computed Tomography (SPECT) and Positron Emission Tomography (PET) systems have been investigated. The use of a geometry with best compactness and the development of an algorithm with best accuracy have led to the conception of tomographs with a spatial resolution of the order of 2 mm [15, 16]. In practice, the fundamental spatial resolution of PET systems is limited by the average distance that the emitted positron travels before it annihilates and by the residual momentum of the electron–positron pair at annihilation which results in gamma rays being emitted with a small deviation from the assumed 180°. In the case of SPECT cameras, the spatial resolution depends on the geometry of the collimation system. Although, in theory, it should be possible to conceive a camera yielding submillimetric spatial resolution using collimators with holes of very small diameter, the very low efficiency which should result from such a geometry would impose an unrealistic acquisition time and too strong an injected dose. This condition has led to the conception of a SPECT camera which can usually only be
employed for visualizing static tracer distributions, whereas their planar imaging variants can analyse rapidly changing activity distributions albeit with less accuracy. Among the numerous PET scanners which have been dedicated to small animals, some systems have already provided some promising results and are now commercially available.

Figures 5.1 and 5.2 present images obtained from animal PET systems. Figure 5.1 shows a few examples of biological applications performed with $^{18}$F-FDG [17] and in Fig. 5.2 the specific problem of resolution is illustrated through the binding of a radiotracer in a specific region of the central nervous system called the striatum. Figure 5.2a shows the perfect distinction of both lobes of the striatum obtained in primate and rat, discrimination which became more ambiguous in mouse since the distance between lobes is smaller than the intrinsic resolution of the tomograph [18]. Figure 5.2b shows that alternative PET systems can offer optimized images leading to a perfect distinction of the structures in mice [19]. Nevertheless, this can only be achieved by significantly increasing the acquisition time (20 minutes in this example), which precludes the existence of dynamic information on the binding kinetic of the tracer.

Figure 5.1: $^{18}$F-FDG rat images obtained with MicroPET, the animal PET system developed at UCLA. Top: coronal cross-sections through the whole body. Middle: transverse sections through the level of the heart. Bottom: coronal brain sections. Acquisition time varied from 90 minutes (top) to 40 minutes (others). (From Ref. [17].)

Figure 5.2: a) PET images of horizontal (top) and coronal (bottom) brain sections of primate, rat and mouse performed with MicroPET, the animal PET system developed at UCLA. Black areas correspond to striatum revealed after the injection of [11C]-CFT, a tracer of the dopaminergic system. Image time: 50 minutes. Injected doses: 5 mCi (primate), 1 mCi (rat), 180 $\mu$Ci (mouse). (From Ref. [18].) b) PET image of mouse coronal brain section obtained with PET HIDAC, the animal PET system developed by Oxford Positron Systems (UK). Striatum is also revealed after injection of [11C]-CFT. Image time: 20 minutes. Injected dose: 67 $\mu$Ci. (From Ref. [19].)
In addition to SPECT and PET systems, other tomographs have been developed specifically for small animal imaging based on different principles [18]. This is in particular the case of TOHR, a tomograph developed at the University of Orsay (France) with high resolution and small field of view [20]. It is a gamma- or X-ray counter composed of a large solid angle, focusing collimator coupled with a set of radiation detectors. Here, the backprojection is avoided by directly measuring the activity concentration in a small volume assumed as a voxel of the image, and the tomographic image is accumulated by moving the sample relative to the detector to scan the volume of interest. Figure 5.3 shows accumulation of $^{125}$I in a rat thyroid obtained with TOHR. The 1.4 mm resolution allows the visualization of not only both lobes of the structure but also the isthmus connecting them.

![Figure 5.3: Image of a rat thyroid section obtained with TOHR after the injection of free $^{125}$I. Image time: 30 minutes. Injected dose: 60 $\mu$Ci.](image)

5.3 Magnetic resonance imaging systems

One of the major advantages of magnetic resonance imaging (MRI) is that the technique can deliver both anatomical and metabolic images. Since the technique is based on the detection of nuclei having specific magnetic properties, these nuclei can be from an exogenous source (injection of paramagnetic tracer) or from an endogenous source (detection of signal issued from $^1$H protons, from $^{13}$C, $^{19}$F or $^{31}$P nuclei, or even from an endogenous paramagnetic molecule: deoxyhemoglobin).

As with SPECT systems, the extension of MRI to small animals is based on a trade-off between resolution and sensitivity. Indeed, the magnetic field necessary to obtain a resonance (and thus a relaxation of the magnetic nucleus) is applied as a gradient to combine the location information. The spatial resolution is thus fixed by this gradient whereas the sensitivity of the system, i.e. the intensity of measured signals, depends on both the magnetic field intensity and the measured volume fixed by the resolution and the acquisition time. The increase of the spatial resolution is therefore based on the choice of a suitable gradient to preserve detectable signals. This can easily be reached by increasing the intensity of the magnetic field, the acquisition time, or the region of interest: conditions difficult to adapt on human but not on rodent. Within these constraints, the parametric conditions are different according to the type of MRI expected. Anatomical MRI ($\text{MRI}_a$), which is based on the detection of relatively high signals, is obtained by using long acquisition times and low magnetic fields (around 1.5 to 2 T). This allows images with a spatial resolution yielding 100 $\mu$m to be obtained (see Fig. 5.4 [21]) — particularly well suited to rodent tissue dimensions [22]. MRI has for instance great potential for analysing the phenotype changes of genetically modified mice.

Functional MRI ($\text{MRI}_f$), which is based on the detection of low detectable signals, is obtained by using high magnetic fields (from 3 to 10 T) and a trade-off between the probed volume dimension and the acquisition time [23]. To adjust the low sensitivity of the method, it is indeed necessary to realize measurements on large volumes since the dynamic nature of the physiological information is not compatible with a long acquisition time [24]. Techniques using injection of exogenous molecules as enhancer have been recently proposed to increase the sensitivity of the method [25, 26]. Two examples are shown in Figs. 5.5 and 5.6, which demonstrate the possibility to superimpose the dynamic image directly on the anatomical image from the same animal [26, 27].
Figure 5.4: MRI image of the whole body of a mouse with isotropic resolution of $100 \times 100 \times 100 \, \mu m$ acquired at the Duke Center for in vivo Microscopy.

In conclusion, one of the most promising areas of application for imaging in the biomedical sciences is the study of small animal models. Radioimaging and MRI systems — as well as computed tomography, ultrasound systems, echography and optical imaging — are now available for rodent. Biomedical programmes are underway worldwide to use these potential tools on animal models. At present, focus is on the possibility to obtain complementary measurements simultaneously on the same animal by combining the techniques [28]–[30]. This will provide tools able to supply both high spatial and temporal resolution anatomical and dynamic images of the small animal. The capability of these powerful tools will be enhanced by other improvements which are also under development and which concern both the complete monitoring of the animal during experiments (control of physiological parameters) and the possibility to perform studies on awake and freely moving animals [31].

Figure 5.5: Image of a frontal brain section of a rat acquired with a 7 T magnet (from Ref. [27]). In colour: MRI of the olfactory bulb after an olfactory stimulation obtained with a 220 $\mu m \times 220 \, \mu m \times 1 \, mm$ spatial resolution and 36 s temporal resolution. In grey: corresponding high-resolution MRI.

Figure 5.6: Frontal rat brain distribution of a cerebral blood-flow increase after the injection of 1 mg/kg cocaine revealed by MRI. Images were produced with a 2 T magnet after the injection of a paramagnetic enhancer and superimposed with MRI images. Spatial resolution, 0.6 mm$^2$; temporal resolution, 10 s. (From Ref. [26].)
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6. Proton and Heavy Ion Beam Therapy

G. Kraft and J. Debus

6.1 Motivation

Cancer is the second most frequent cause of disease in developed countries. Every year, more than a million people in the European Union are diagnosed with cancer. A general cure is not in sight because the term 'cancer' stands for a large variety of diseases that are characterized by uncontrolled cell growth. Normally, cell growth is regulated by a number of specific genes that stop growth after an organ has reached its normal size. Only a few cells in our body show permanent cell division like the stem cells of the different blood-forming organs, skin cells, and mucosa cells in the intestines. Most of the other cells in our body are not permanently dividing and some of them — such as brain cells — stop dividing completely.

However, if the balance of genetic growth regulation is impaired in a single cell by a genetic mutation, and if this cell is not eliminated, infinite tissue growth will result from this single cell and tumour growth starts.

Most tumours originate from such a single mutated cell. In contrast to a bacterial infection, when foreign cells invade the body, tumour cells are not foreign. Therefore they are almost indistinguishable from other cells of the body except that one of the growth regulating genes, called oncogenes, is out of control.

Oncogenes are also present in any normal cell, but usually their function is strictly controlled. Although our understanding of oncogenes and their regulation has dramatically increased in recent decades, it is impossible — now and probably in the near future — to use molecular therapy, i.e. gene therapy, to cure cancer on a molecular scale. Up to now, macroscopic treatments like surgery, irradiation and chemotherapy have been successfully applied, yielding a cure rate (i.e. the proportion of patients surviving disease-free for at least five years after treatment) of almost 50% (Fig. 6.1).

![Cancer situation as presented by (EC 1991)](image)

Figure 6.1: Distribution of the more than one million new cancer patients in Europe: local disease (red fraction) refers to patients with only one well-defined tumour in the beginning, generalized disease (blue) to those with more than one tumour. Nearly 50% of the patients survived tumour-free for five years by the different treatment modalities, but 18% of patients with local disease in the beginning could not be cured. These are the candidates for particle therapy.

Surgery is the most effective way to remove malignant tissue. However, radiation, alone or in combination with surgery, improves the cure rate to about 40%. For a generalized disease, where the cancer has spread over a larger part of the body, a final cure is much less likely. Even in the fraction of localized tumours, 20% of all patients cannot be cured because their tumour can neither be resected completely by surgery nor — in conventional therapy — be treated with sufficiently high radiation doses. In principle, any tissue can be destroyed by radiation if the applied dose is high enough. In practice, radiation...
therapy is limited by the radiation tolerance of the normal tissue surrounding the tumour. Consequently, the precision of dose delivery is the key to the success of therapy: the more the dose is ‘conformed’ and limited to the target volume and the more the surrounding normal tissue can be spared, the higher can be the dose applied to the tumour and the more likely is the success of the treatment [1].

6.2 Dose delivery

In conventional radiotherapy, dose delivery is limited by the physical parameters of the depth–dose distribution and the scattering of the beam when penetrating a thick layer like the human body. For low-energy X-rays the dose decreases exponentially with depth. For higher energies, as for cobalt gamma rays, a build-up effect shifts the maximum dose below the skin (Fig. 6.2). Finally, for highly energetic bremsstrahlung the maximum dose is located 3 cm below the skin and followed by a slowly decreasing exponential curve, which allows a higher dose to be delivered to deep-seated tumours. Consequently, the trend in conventional radiotherapy with electromagnetic radiation has been towards higher energies because of better depth–dose profiles and reduced scattering.

Today, sophisticated beam delivery methods are developed, whereby a fine beam of high-energy bremsstrahlung is delivered in an intensity-controlled way and is restricted to the target volume by a multileaf collimator. If many fields of such a beam are delivered to the target volume from many (i.e. 6 to 10) directions, a highly conformed dose can be applied even to tumours of complex shape. However, intensity-modulated radiation therapy with photons (IMRT) can only distribute the unwanted dose over a larger area of normal tissue; it cannot increase the ratio of tumour dose to normal-tissue dose.

6.3 Ion beam therapy

The transition from electromagnetic radiation to heavy particles like protons or heavier ions like carbon is the next step to improving the physical properties for external radiotherapy [2].

![Graph showing depth-dose profiles](image)

Figure 6.2: Comparison of the depth–dose profiles of X-rays, $^{60}$Co-gamma rays and Röntgen bremsstrahlung with carbon ions of 250 MeV/u and 300 MeV/u (courtesy of U. Weber).

The main differences between photons and particle beams are the depth–dose profiles and the increased radiobiological efficiency (RBE). Particle beams show an inverse dose profile compared to photons: the dose increases with penetration depth up to a maximum value (Bragg peak) at the end of the range (Fig. 6.2). Normally, the tumour and the target volume are much larger than the unmodified Bragg peak and the beam has to be adjusted to the target volume.

In the early days of particle therapy, the size of the treated field was modulated in the longitudinal and lateral directions by means of collimators, compensators and range modulators. Using these passive systems, the target volume can be shaped according to the tumour volume in such a way that the tumour is completely covered by the high-dose area. But a perfect congruence between irradiated volume and tumour cannot be achieved with passive beam shaping systems. Frequently a large fraction of normal tissue is contained in the high-dose region, causing side-effects later on. In order to improve the
situation a novel technique of irradiation by active beam shaping has been introduced for protons at PSI, Villigen, and for carbon at GSI, Darmstadt.

Figure 6.3: Principle of the three-dimensional conformal beam delivery. The target volume is dissected into layers of equal particle range. These layers are covered with a net of pixels for each of which the intensity has been calculated before. The beam is switched from one point to the next after the precalculated number of particles has been reached. Together with a shift in depth, which is done by energy variation, a three-dimensional target volume can be filled with high precision.

Ions are charged particles, and can thus be easily deflected by magnetic fields. Consequently, the principles of intensity modulation can be reached by beam scanning using two fast driven electromagnets with fields perpendicular to each other and to the beam. This is the same technique as used in every TV set, where an electron beam is scanned over the screen and the intensity is modulated for each spot (Fig. 6.3). However, in the ‘ion TV’ a third dimension is introduced by changing the particle energy and irradiating a target volume layer by layer. Using this technique, a target volume of any shape can be irradiated with a precision of a few millimetres.

Using intensity-modulated delivery for particle beams, conformity of the applied dose to the target volume can be optimized. In Fig. 6.4, a treatment plan of nine photon fields is compared to a two-field carbon therapy, both using intensity-modulated techniques.

The carbon plan has less dose in the normal tissue outside the target volume and steeper dose gradients to critical structures such as the brain stem. This is due to the smaller lateral scattering of carbon beams and — most importantly — to the inversed dose profile, i.e. the increase of dose with penetration depth into the body up to a sharp maximum at the end of the particle range.

6.4 Patient positioning and quality assurance

This high precision of particle dose delivery demands equally high precision in patient positioning. In stereotactic treatment of patients with conventional therapy, patient immobilization techniques have been developed that allow a precision of a millimetre in the head region and a few millimetres along the back bone and in the pelvic region. These are areas of no or negligible internal motion; consequently they have, up to now, been prime candidates for particle treatments. However, it does seem possible to adapt beam scanning to moving organs.

Another important problem is the control system. The actual delivery has to be monitored in each treatment section and compared to the planned beam delivery. For this, two different and complementary methods have been developed: position-sensitive transmission detectors installed directly in front of the patient and gamma cameras as monitors for the positron decay that occurs inside the patient.

From high-energy nuclear physics, the technology of position-sensitive detectors has been
adapted for a fast control system for therapy. At GSI, for instance, two systems of a wire chamber combined with an ionization chamber measure the beam intensity and the centre of the beam 6000 times per second and compare the measurement to the requested data. In case of disagreement beyond a small tolerance limit, the beam is cut off within less than half a millisecond. This fast control system is the basis for safe operation of the beam in a medical treatment.

The particle distribution inside the patient can be monitored in addition by means of positron emission tomography techniques: during the passage of the primary carbon beam a small amount of radioactivity is produced by nuclear reactions. From this activity, the positron-emitting isotopes like $^{10}$C and $^{11}$C are of special interest because they have almost the same range as the primary beam and can be monitored via the positron decay over two coincident gamma quanta. With this novel technique the position and range of the beam can be controlled inside the patient without applying any additional dose.

### 6.5 Radiobiological efficiency

Finally, the use of ions heavier than protons has the advantage that the biological reaction of the cell can be potentiated. Heavy ions have a greater energy loss and consequently a higher density of locally produced electrons. At the microscopic scale of DNA as the sensitive target inside the cell the elevated ionization density correlates with an elevated density of DNA lesions. Single and isolated lesions like single strand breaks can be repaired easily by the cell. Clusters of double strand breaks are frequently connected with information loss at the DNA level and mostly irreparable [2].

Using carbon ions the energy loss, and consequently the lesion density, is distributed in a very favourable manner: At high energies in the entrance channel ionization density is low, increasing towards the end of the track. Thus, the extent of the damage differs along the track. At its beginning at high energies the DNA damage in the tissue in front of the tumour can be repaired to a large extent. At the end of the track, ionization density is high and mostly irreparable lesions are produced in the tumour. Confining the track ends strictly to the tumour, the efficiency for tumour-cell killing can be potentiated by a factor of two or more, which is most relevant for otherwise radioresistant tumours. The choice of carbon ions as the optimum ion is mainly determined by the gain in

![Figure 6.5: Comparison of planned dose (top) with expected (middle) and measured (bottom) positron activity. This comparison shows that no tissue in critical regions like the brain stem is affected.](image-url)
radiobiological efficiency between entrance channel and tumour. Lighter ions like protons lack the elevated RBE at the end of the track. Heavier ions like neon have an additional increase in efficiency also in the entrance channel, thereby causing severe late effects.

Carbon ions strike the best balance between the production of lethal lesion in the tumour on one hand and minimization of damage to normal tissue on the other. Measurements of DNA double strand breaks as well as of cell survival have confirmed the high repair rate in the healthy tissue and the very effective cell killing in the tumour region.

![Adenoidcystic carcinoma. Combined radiotherapy with photons and carbon ions: before (left) and six weeks after radiotherapy.](image)

However, the choice of carbon ion means that, first, the RBE variation complicates the treatment planning to some extent and second, that the beam delivery should conform as far as possible to the tumour and avoid any normal tissue being hit by the very effective stopping ions. These conditions are fulfilled using the scanning systems and the biological optimized treatment planning. The clinical results achieved up to now confirm the theoretical benefits of target conformity for protons and carbon ions and the additional RBE gain for carbon beams.

These very advantageous properties of ion beams have prompted many facilities to introduce ion therapy. From the rather reluctant beginnings of proton beams at Berkeley in 1958, this new radiation modality has now turned into a forefront activity of many advanced clinical units. The basis of this increased activity is the success of ion therapy up to now. Approximately 30,000 patients have been treated, mostly with proton beams. Only for a small fraction of patients are heavy ions like carbon ions used: around 1000 patients in Chiba, Japan, and 100 at GSI, Germany. Patients have been treated with ions in many centres (see Chapter 7 on hadron therapy) and over a period of more than 40 years, during which particle therapy has come a long way.

### 6.6 Clinical results

To date almost 30,000 patients have been treated with ion beams, mainly with protons. At the low-energy proton beams, eye tumours — mainly uveal melanomas — have been treated extremely successfully [3].

These tumours occur in patients of all ages but are more frequent in older people. The tumours are quite large, typically 13 mm in diameter and 15 mm in height, and mostly in close proximity to the optic nerve which would be affected in conventional therapy. Therefore, enucleation (removal of the eye) is the frequent alternative to radiation therapy. This results in the total loss of vision but the radical tumour extirpation prevents a metastatic spread.

![Local control rate as a function of delivered dose. The local tumour control can be substantially raised for tumours in the base of the skull region by the application of ion beams [4].](image)

Treatment with proton beams is in most cases similarly effective. Because of the small lateral spread of the proton beam a dose of 70 Gy can be given to the tumour, typically in five days, without a deleterious effect on the
optic nerves. Because it is possible to visually control the tumour localization in the eye with great precision, an excellent conformity of the treatment can be achieved. In the treatment of more than 10,000 eye patients, a local tumour control of 95% or better was reached, coupled with a high survival rate and a high rate of useful vision. The treatment of eye tumours with proton beams yields by far the best clinical result. Thus, proton treatment is the best choice for these tumours.

The other fraction of tumours treated with protons is less homogeneous and spread over a few large centres like Harvard, Loma Linda and many smaller activities. Due to the very limited access to proton therapy, mostly those tumours where the greatest benefit in comparison to conventional therapy was to be expected have been chosen for proton treatment. These are tumours that grow rather slowly and have little radiosensitivity like chordomas, chondrosarcomas and meningiomas.

Secondly, tumours close to critical organs such as the brain stem and spinal cord have been chosen. These include tumours in the head and neck region, along the spinal cord or in other regions where the target dose is limited by the tolerance of critical organs nearby. Given its better conformity, proton therapy was applied in these cases. Because of the lower contamination of the normal tissue the tumour dose could be increased, yielding significantly better results. Since the escalation of dose was a very slow process — depending on the physical improvement of dose distribution over a period of 40 years — it cannot be characterized by a single number as was the case with eye tumours; but in most cases an improvement of the local control of some 30–40% could be achieved. This increase of the tumour control rate with particle beams is a significant difference, especially if this is not accompanied by a higher incidence of side-effects.

The good results of proton therapy confirm a general rule in radiotherapy: that better conformity of the irradiated volume with the target volume yields better results. This holds true for heavy-ion beams, too, where a superior conformity compared to protons can be achieved. But the main advantage of heavy ions is the increase in biological effectiveness that should provide a greater tumour control probability, especially for radioresistant tumours.

Heavy-ion therapy began in 1974 at the Lawrence Berkeley Laboratory. There, first patients were treated with argon ions at the Bevalac accelerator complex where a limited number of patients could be irradiated in time sharing with physics experiments. After a few patient treatments, however, the observed side-effects proved to be unacceptable. Treatment then shifted to the lighter silicon and further on to neon beams, which were applied to approximately 400 patients. Tumours treated were radioresistant ones close to critical organs; they ranged from brain tumours and other head and neck tumours to tumour incidences all over the human body. Because of the large diversity of the tumours treated a quantitative analysis is not possible, but the results were so convincing that they motivated the construction of a dedicated Heavy Ion Medical ACcelerator (HIMAC) in Chiba, Japan.

Since 1994 about 1000 patients have been treated with carbon ions for a variety of tumours in carefully conducted Phase I/II and Phase II trials. Patients treated with carbon ions at HIMAC include those with head and neck cancer, lung cancer, liver cancer, prostate cancer and tumours of the gastrointestinal tract. At the current time, the most promising data are found in patients with base-of-skull tumours, head and neck cancer, soft-tissue sarcoma and early-stage lung cancer in the Japanese trials. The incidence of treatment-related toxicity was very limited in the Phase II trials.

At GSI patient irradiations started in December 1997. So far 100 cancer patients have undergone carbon-ion irradiations and promising first results can be reported today [5]. At GSI, again, radioresistant tumours close to critical organs are treated but for the first time the active scanning systems are used in connection with a biology-based treatment planning system. This yields a dramatic reduction of dose to the normal tissue outside the tumour and in
consequence a significant decrease in early side-effects. Although the physical dose could be decreased because of a realistic RBE estimation, to date no tumour regrowth within the treated area has been observed in the Phase I/II study. Because of the short follow-up time and the limited number of patients it is too early to draw final conclusions from the heavy-ion treatment, but the results achieved so far are motivating. This reflects the increasing number of new ion-beam units proposed for the near future (see Chapter 7).

6.7 Summary

The use of heavy charged particles like protons or heavier ions was triggered by radiation physicists and radiobiologists. It was and is difficult to promote these new ideas in the medical community mainly because of the larger complexity and technical demand of these therapy units. In the beginning, particle therapy was feasible only with the generous help of the nuclear physics community that has developed the high-energy accelerators and the beam-monitoring equipment that allows safe patient treatment. At present, a separation of the medical application from the nuclear physics experiments is taking place, and dedicated medical facilities can now be found on the industrial market. Just a few operational therapy units of modern standards will make this the greatest success of a spin-off product of nuclear science.
Bibliography


7. Hadrontherapy in the World

U. Amaldi

7.1 Conventional radiotherapy

About one third of the 15,000 particle accelerators running in the world are used in biomedicine: 3% in nuclear medicine and 30% in radiotherapy [1]. Most of them produce X-rays and are discussed in this section. Only about twenty-five are used as sources of hadron beams; they are discussed in Sections 7.2–7.5).

Radiotherapists use mainly electron linear accelerators (linacs) as sources of radiation. About 5000 such accelerators are at present treating patients world-wide. Photon beams (usually called ‘X-ray beams’ by medical doctors) are characterized by an exponential absorption after a maximum which, for beams having a maximum energy of 8 MeV, is reached at a depth of 2–3 cm.

In order to selectively irradiate deep-seated tumours, radiotherapists use multiple beams usually pointing to the geometrical centre of the target. These irradiation techniques are applied by rotating the structure containing the linac about a horizontal axis (Fig. 7.1).

In a typical treatment session 2–2.5 grays (Gy) are delivered to the tumour target, while delivering less than 1–1.2 Gy to any of the organs at risk (1 gray = 1 J/kg). Since the treatment lasts about 30 sessions, usually spread over six weeks, the target will eventually have received 60–75 Gy. It has to be underlined that even a small increase of the dose is worthwhile. For a typical tumour, which is controlled with a 50% probability, a 10% increase of the dose usually improves this probability by 15–20%, so that the control rate increases from 50% to 65–70%. This is a sizeable effect since it corresponds to a reduction of the failure rate from the initial 50% to 30–35%.

The unavoidable doses given to the healthy tissues are a limiting factor. By conforming the dose to the target the dose absorbed by the tumour can be increased. Thus ‘conformity’ is the main goal of all recent developments in cancer teletherapy. Intensity Modulated Radio-Therapy (IMRT) makes use of 6–10 X-ray beams; the beams may be non-coplanar and their intensity is varied across the irradiation field by means of variable collimators (‘multileaf collimators’) that are computer controlled. For planning the treatments very sophisticated codes have been developed (‘inverse treatment planning’).

7.2 Distribution of the dose with beams of charged hadrons

‘Hadrontherapy’ is a collective word that describes the many different techniques of oncol-ogical radiotherapy which make use of fast non-elementary particles made of quarks: protons, neutrons and light nuclei are the hadrons used to locally control many types of tumour.

Neutron beams have an unfavourable depth–
dose distribution very similar to that of photon beams. Consequently they are losing their appeal, though about 20,000 patients have been treated and good results have been obtained by irradiating tumours of the salivary glands and of the parotis.

The depth–dose curves of proton and light-ion beams are completely different from those of photons and neutrons because these charged particles show little scattering when penetrating in matter and give the highest dose near the end of their range, as shown by the ‘Bragg peak’, just before coming to rest. The first proposal to use protons and carbon ions in radiotherapy was put forward in 1946 by Bob Wilson, who suggested exploiting the Bragg peak to obtain a conformal treatment [2]. It is worthwhile noting that the existence of such a peak is a direct consequence of the fact that — below about 250 MeV/u — for all light ions the kinetic energy dependence of the energy loss is well reproduced by a simple power law: $K^{-0.82}$. It follows that all Bragg peaks, i.e. all energy losses plotted as a function of the residual range $r$, are approximately given by the simple formula $r^{-0.45}(0.45 = 0.82/1.82)$. The famous peak of a mono-energetic beam of fully stripped ions — down to a residual range of 1–2 mm of water — is due to divergence of the function $r^{-0.45}$ when $r$ goes to zero.

Protons and light ions are advantageous in Intensity Modulated Hadron Therapy (IMHT) because of three physical properties. Firstly, as mentioned above, they deposit their maximum energy density in the Bragg peak at the end of their range, where they can cause severe damage to the tumour cells while sparing both traversed and deeper located healthy tissues. Secondly, they penetrate the patient practically without diffusion. Thirdly, being charged they can easily be formed into narrowly focused and scanned pencil beams of variable penetration depth so that any part of a tumour can be accurately and rapidly irradiated. Thus, a beam of protons or light ions allows highly conformal treatment of deep-seated tumours with millimetre accuracy, giving minimal doses to the surrounding tissues.

The depth of the Bragg peak depends on the initial energy of the ions and its width on the energy spread of the beam that, in order to make best use of the distal steep drop of the peak, should not be larger than about 0.2%. By varying the energy during the irradiation in a controlled way, one can superimpose many narrow Bragg peaks and obtain a Spread-Out Bragg Peak (SOBP). This can be achieved in two ways: the first is based on the interposition of an absorbing material of variable thickness in the beam path; the second is based on the modulation of the beam energy during irradiation. This modulation is easily feasible in some accelerators such as synchrotrons, but it is more difficult with others (cyclotrons).

In order to reach depths in soft tissues of more than 25 cm — necessary to treat deep-seated tumours — proton and carbon-ion beams must have initial energies not lower than 200 MeV and 4500 MeV, respectively (i.e. 375 MeV/u).

Radiotherapists use rotating linacs to treat patients with X-ray beams (Fig. 7.1) and would like to have the same possibility when using proton (and ion) beams. The magnetic rigidity of 200 MeV protons is such that the magnetic channel capable of achieving this has a typical total radius of 4–5 m. For this reason, fixed (mainly horizontal) proton beams were used world-wide until 1992, when the first hospital-based centre became operational at the Loma Linda Medical Center in Los Angeles (Fig. 7.2).

Since then new facilities have usually included more ‘gantries’: large mechanical structures which rotate around a horizontal axis and rigidly support the needed bending magnets and quadrupoles.

Relatively simple ‘passive spreading systems’ were used in all centres until 1997. In this approach, the protons are diffused by a first ‘scatterer’ and their energy is adapted to the distal form of the tumour by using appropriate absorbers. The transverse form of the irradiation field is defined by collimators. Only in 1997 at the Paul Scherrer Institute (PSI) in Villigen, Switzerland did the first rotating gantry with a 250 MeV proton beam come into operation (Fig. 7.3).
At PSI a novel ‘active spreading system’ has been implemented: the target is subdivided into many thousands of voxels and each one is irradiated in successive steps by sending the proton beam of about 5 mm section with a given energy and direction. By summer 2001 about 80 patients had been treated successfully. As discussed in Section 7.5.1, during the same period an active spreading system of a different design was started on the medical ion beam of GSI (Darmstadt). The new hadrontherapy facilities will all be capable of treating patients with active spreading systems.

For eye melanoma, as for the treatment of macular degeneration, protons of energies in the range 60–70 MeV are enough and passive spreading is sufficient: the accelerators, which are listed in Table 7.1, are much smaller and there is no need for active dose distribution. Contrary to deep therapy, this type of treatment is well developed in Europe. The most important centres are at PSI, the Centre Antoine Lacassagne (Nice, France), the Clatterbridge Centre for Oncology (UK), the Centre de Protonthérapie d’Orsay (France) and the Lisa Meitner Centre in Berlin. In October 2001 patients will be treated at the Laboratori Nazionali del Sud (LNS) of INFN in Catania, where an external source has been added to the superconducting cyclotron that has been running for many years.

In the following, only hospital-based centres having more than one irradiation room devoted to hadrontherapy of deep-seated tumours will be discussed. Typically, between 10,000 and 20,000 irradiation sessions are held in such a centre every year.

### 7.3 Number of patients

To date about 30,000 patients have undergone protontherapy (Table 7.1) and, as far as deep-seated tumours are concerned, very good results
Table 7.1: Patients irradiated with protons world-wide up to summer 2001 [4].

<table>
<thead>
<tr>
<th>Centre</th>
<th>First therapy</th>
<th>Last therapy</th>
<th>Accel.</th>
<th>Beam</th>
<th>Max. energy [MeV]</th>
<th>Clinical energy [MeV]</th>
<th>No. of patients</th>
<th>Date of total</th>
</tr>
</thead>
<tbody>
<tr>
<td>LBL, Berkeley (USA)</td>
<td>1954</td>
<td>1957</td>
<td>S</td>
<td>Horiz.</td>
<td>70-230</td>
<td>70-150</td>
<td>30</td>
<td>Jul. 01</td>
</tr>
<tr>
<td>GWI, Uppsala (Sweden)</td>
<td>1957</td>
<td>1976</td>
<td>C</td>
<td>Horiz.</td>
<td>15-185</td>
<td>100</td>
<td>73</td>
<td>Jun. 30</td>
</tr>
<tr>
<td>HCL, Cambridge (USA)</td>
<td>1961</td>
<td>1976</td>
<td>C</td>
<td>Horiz.</td>
<td>160</td>
<td>160</td>
<td>8906</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>JINR, Dubna (Russia)</td>
<td>1967</td>
<td>1974</td>
<td>S</td>
<td>Horiz.</td>
<td>70-200</td>
<td>70-200</td>
<td>84</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>ITEP, Moscow (Russia)</td>
<td>1969</td>
<td>1974</td>
<td>S</td>
<td>Horiz.</td>
<td>10,000</td>
<td>70-200</td>
<td>3414</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>LINPh St.Petersburgh (Russia)</td>
<td>1975</td>
<td></td>
<td>SC</td>
<td>Horiz.</td>
<td>1000</td>
<td>1000</td>
<td>1029</td>
<td>Jun. 98</td>
</tr>
<tr>
<td>NIRS, Chiba (Japan)</td>
<td>1979</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>70-90</td>
<td>70-200</td>
<td>133</td>
<td>Apr. 00</td>
</tr>
<tr>
<td>PMRC, Tsukuba (Japan)</td>
<td>1983</td>
<td></td>
<td>S</td>
<td>Vert.</td>
<td>500</td>
<td>500</td>
<td>700</td>
<td>Jul. 00</td>
</tr>
<tr>
<td>PSI, Villigen (Switzerland)</td>
<td>1984</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>500</td>
<td>250</td>
<td>3360</td>
<td>Jul. 00</td>
</tr>
<tr>
<td>JINR, Dubna (Russia)</td>
<td>1987</td>
<td></td>
<td>SC</td>
<td>Horiz.</td>
<td>600</td>
<td>70-200</td>
<td>88</td>
<td>May 01</td>
</tr>
<tr>
<td>TSL, Uppsala (Sweden)</td>
<td>1989</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>200</td>
<td>45-200</td>
<td>300</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>Douglas U. Clatterbridge (UK)</td>
<td>1989</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>62</td>
<td>62</td>
<td>1033</td>
<td>Dec. 00</td>
</tr>
<tr>
<td>LLUMC Loma Linda (USA)</td>
<td>1990</td>
<td></td>
<td>S</td>
<td>Horiz.</td>
<td>250</td>
<td>70-200</td>
<td>6174</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>UCL, Louvain (Belgium)</td>
<td>1991</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>90</td>
<td>90</td>
<td>21</td>
<td>Nov. 93</td>
</tr>
<tr>
<td>CAL, Nice (France)</td>
<td>1991</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>65</td>
<td>65</td>
<td>1590</td>
<td>Jun. 00</td>
</tr>
<tr>
<td>CPO, Orsay (France)</td>
<td>1991</td>
<td></td>
<td>SC</td>
<td>Horiz.</td>
<td>200</td>
<td>70-200</td>
<td>1894</td>
<td>Jan. 01</td>
</tr>
<tr>
<td>NAC, Faure (South Africa)</td>
<td>1993</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>200</td>
<td>70-200</td>
<td>398</td>
<td>Jun. 01</td>
</tr>
<tr>
<td>IUCF, Indiana (USA)</td>
<td>1993</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>200</td>
<td>75-200</td>
<td>34</td>
<td>Dec. 99</td>
</tr>
<tr>
<td>UC Davis, Calif. (USA)</td>
<td>1994</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>200</td>
<td>70-200</td>
<td>284</td>
<td>Jun. 00</td>
</tr>
<tr>
<td>TRIUMF (Canada)</td>
<td>1995</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>70</td>
<td>57</td>
<td>72</td>
<td>Jun. 00</td>
</tr>
<tr>
<td>PSI, Villigen (Switzerland)</td>
<td>1996</td>
<td></td>
<td>C</td>
<td>Gantry</td>
<td>800</td>
<td>250</td>
<td>72</td>
<td>Dec. 00</td>
</tr>
<tr>
<td>Berlin (Germany)</td>
<td>1998</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>55</td>
<td>65</td>
<td>166</td>
<td>Dec. 00</td>
</tr>
<tr>
<td>NCC, Kashiwa, (Japan)</td>
<td>1998</td>
<td></td>
<td>C</td>
<td>Horiz.</td>
<td>235</td>
<td>235</td>
<td>75</td>
<td>May 00</td>
</tr>
</tbody>
</table>

**TOTAL** 29852

Figure 7.4: The proton treatment plan on the right is definitely better than the IMRT treatment on the left that uses nine X-ray beams [6]. The active proton distribution system developed at the Paul Scherrer Institute was used.
have been obtained in head and neck cancers. Clinical data on protontherapy, indications, protocols and results are summarized in Ref. [5].

Many protontherapy protocols are well defined and Phase III trials are under way to accurately compare the results with those of conventional radiotherapy. By summarizing a long chain of arguments, one can state that most experts agree that protontherapy is better than even the best IMRT treatments for about 1% of all the patients irradiated nowadays with X-rays. Large tumours in particular are potential targets because with X-rays the surrounding tissues receive unavoidably a much larger dose. This is clear from the treatment plan comparison of Fig. 7.4.

Since in a population of 10 million Europeans about 20,000 patients are irradiated every year with X-rays, this corresponds to about 200 candidates for protontherapy. Moreover, for about 10% of the usual treatments, i.e., for about 2000 patients per year, protontherapy should give a better tumour control, but more clinical data are needed to precisely quantify the advantages.

Carbon beams of about 400 MeV/u are indicated for treatment of deep-seated tumours, which are radioresistant both to X-rays and to protons. The radiobiological arguments can be summarized as follows [7]. The dense column of ionization produced near the Bragg peak of a light-ion track gives rise to many double-strand breaks and multiple damaged sites when it crosses the DNA of a cell nucleus. The effects on the cell are thus qualitatively different from the ones produced by sparsely ionizing radiation, such as X-rays, electrons and protons, which interact mainly indirectly with DNA, mostly producing repairable single- and double-strand breaks. Due to the much larger proportion of direct effects, light ions have — for many 'end-points' and delivered doses — a relative biological effectiveness (RBE) which is about three times larger than that of X-rays and protons. They are therefore suited for clinical situations where the radioresistance — linked to hypoxia or being 'intrinsic' — is a difficult problem to overcome both with conventional radiation therapy and with protons.

A number of results show that these qualitative differences show up at linear energy transfer (LET) values larger than about 20 keV/μm. This is physically understandable, since it corresponds to an average energy deposition of 40 eV in the 2 nm thickness of a crossed DNA fibre: just what is needed to produce on average one ionization.

About ten years ago, radiobiologists and radiotherapists reached the conclusion that the optimal ions are found in the range $Z = 3 – 6$, i.e. between lithium and carbon. There are two main reasons for this. Firstly, LET values larger than 20 keV/μm can be confined to the tumour tissues: in the cases of carbon and lithium this approximate threshold is passed for a residual range in water of 47 mm and 5 mm, respectively. Secondly, some of the incoming ions fragment into lighter ones that have a lower charge and thus a longer range than the parent ions. This effect: increases with $Z$ and produces a 'tail' in the dose distribution downstream of the rapid fall-off of the Bragg peak, often irradiating healthy tissues.

As far as the number of patients is concerned, the starting point is that about 20% of all the tumours currently treated with X-rays are resistant, so that the number of new radioresistant tumours per year is about 4000 within a population of 10 million. Between one third and half of these tumours would benefit from an ion treatment, corresponding to at least 1300 patients per year; but part of this cohort overlaps with the one of the already discussed protontherapy patients. Specific indications thus correspond to about 1000 patients per year for 10 million inhabitants.

Since the usual cellular repair mechanisms have little effect, there is no point in fractionating the dose in the 30 sessions used with X-rays. This shortening of the treatment is an advantage, but the lack of repair may also induce late recurrences in the healthy tissues and requires much care in modelling the RBE of a complex irradiation field. Potential problems can be overcome by using a carbon beam to deliver in 3–4 sessions a 'preboost' to the central part
of hypoxic radioresistant tumours, followed by a conventional treatment (20–30 sessions) with X-rays [7]. This proposal is also interesting for an effective use of the costly infrastructures to be discussed in Section 7.5.

Whatever treatment schedule is used, many more clinical data are needed since only 1000 patients have been irradiated with carbon ions world-wide. Indeed the available clinical results are mainly due to the activity pursued since 1994 at the Heavy Ion Medical Accelerator Centre (HIMAC) in Chiba, Japan — where about 900 patients have been irradiated with passive spreading systems [8] — and to GSI (Darmstadt) [9], where pioneering work in the simulation of the RBE of ions has been done and about hundred patients have been treated with the raster scanning technique described in Section 7.5.1.

7.4 Centres for deep protontherapy

When discussing deep protontherapy, it should be underlined that only two of the facilities operating at present were built as fully dedicated hospital-based centres: the Loma Linda Medical Center (Fig. 7.2) and the Proton Therapy Facility of NCC in Kashiwa, Japan. All the other centres make use of beams produced by accelerators built for fundamental research in nuclear and particle physics and later adapted to protontherapy. Such centres have been running, some for a long time, in Europe, Japan, Russia and USA (Table 7.1). In South Africa there is the well-equipped National Accelerator Centre in Faure, where neutroneurotherapy is also performed and a new proton beam line is being commissioned.

7.4.1 Centres under commissioning or construction

In Europe, deep protontherapy with charged beams is carried out in Orsay (CPO) and Uppsala at two modified nuclear physics cyclotrons. Thus the recent increase of interest in hadrontherapy throughout Europe is quite natural, as in 2002 there will be two dedicated hospital-based centres for deep protontherapy in the USA and four in Japan (Fig. 7.5).

The Loma Linda Medical Center is represented in Fig. 7.2. The second hospital-based centre in the USA, the Northeast Proton Therapy Center at Massachusetts General Hospital in Boston, will treat patients from autumn 2001. It is based on a 230 MeV cyclotron built by the Belgian Company IBA and aims to treat 1000 patients per year in two gantry rooms and one room with two horizontal fixed beams. In 1999 it was decided to upgrade the cyclotron of Indiana University at Bloomington. As a consequence, in 2003 there will be a third American centre for deep protontherapy: the Midwest Proton Radiation Institute featuring two horizontal beams and an isocentric gantry.

Towards the end of 2001 two Chinese private hospitals ordered cyclotrons and gantries from IBA. The hospitals are in the towns of Zibo (Wanjie Tumor Hospital) and Xian (Chang An Information Industry Group) and the treatments should start in 2003.

Figure 7.5: In Japan, there are four protontherapy centres and two centres (HIMAC at Chiba and HARIMAC at Hyogo) featuring synchrotrons that can accelerate light ions. They are discussed in Section 7.5. The centres shown in green were active in summer 2001.

In Europe a new project (PROSCAN) was launched at the end of 2000 by the PSI. A new superconducting protoncyclotron was ordered for the project from ACCEL (Fig. 7.6). The proton beam will serve both the existing isocentric gantry and a new isocentric gantry and, to actively distribute the dose, an improved version of the PSI spot scanning technique will be implemented.
ACCEL has already sold a superconducting cyclotron to Prohealth AG near Munich, where the Rinecker Proton Therapy Centre (RPTC) will have a multi-purpose treatment room and four gantries.

### 7.4.2 Linear accelerators for proton-therapy

Usually proton linear accelerators run at low frequencies, have diameters of the order of one metre and accelerate large currents. Since the currents needed for protontherapy are only a few nanoamperes, one can use high-frequency accelerating structures, which have small apertures and large accelerating gradients. The idea of using 3 GHz structures with gradients of the order of 15 MeV/m is at the basis of the studies initiated in 1993 by the TERA Foundation, in collaboration with ENEA (Frascati) and Istituto Superiore di Sanità (ISS, Rome) [11]. This activity has led to the TOP project of ISS and the LIBO project.

The *Terapia Oncologica con Protoni* (TOP) project is based on the construction in Rome, in collaboration with ENEA and the oncological institute IRE, of a 3 GHz linear accelerator that will be used for eye therapy. The linac, about 10 m long, is based on an accelerating structure of novel design named ‘side coupled drift tube linac’ [12].

The LIBO project (Fig. 7.7) is based on a Linac BOoster: a 13 m long copper structure which, installed downstream of a small cyclotron, will be capable of accelerating the protons extracted from the cyclotron from 50–70 MeV to 200 MeV or more. The chosen structure (side-coupled linac) was designed for lower frequencies in Los Alamos, but it has never been used to accelerate protons at such a high frequency.

In 1998 a collaboration between CERN, the universities and INFN sections of Milan and Naples and the TERA Foundation was set up to construct a 1.25 m long module of LIBO capable of accelerating protons from 62 to 74 MeV. The module — power tested at CERN at the end of 2000 — performs better than foreseen, since in each of the four tanks the gradient is 27 MeV/m instead of 15.5 MeV/m [13]. An acceleration test (from 62 to 74 MeV) — in collaboration with IBA/Scanditronix — will be performed in Catania at the INFN *Laboratori Nazionali del Sud* in early 2002.

![Image of a cyclotron](image)

**Figure 7.7:** The current of a cyclotron is large (50–100 μA) compared with what is needed for protontherapy (10 nA). The acceptance of LIBO can thus be small and still provide the necessary beam for deep protontherapy. The repetition rate is 400 Hz, well suited for a voxel active spreading system such as at PSI.

### 7.5 Centres with beams of light ions

In 1994 the first patient was treated at HIMAC in Japan with carbon ions and with a passive dose distribution system. As already mentioned, at this centre about 1000 patients affected by brain glioma, tumours of the cervico-cephalic area, lung, liver, prostate and uterine cervix tumours have been treated. Very interesting results on some special tumour sites, such as lungs and liver, have been obtained [8]. Since
400 MeV/u carbon ions have a magnetic rigidity three times greater than 200 MeV protons, the construction of rotating gantries poses many technical challenges. At HIMAC the choice was made to have horizontal and vertical beams in one treatment room. The other two rooms feature horizontal beams. Passive spreading systems are used, but active systems of interest will be implemented.

At the Hyogo Centre (Fig. 7.5) the first patient was treated with protons in May 2001. This centre has three rooms for protons (two of them with gantries) and two rooms for ions featuring a vertical beam and an inclined beam.

7.5.1 The GSI pilot project

In December 1997 at the Darmstadt heavy-ion laboratory, for the first time two brain tumours were treated with a carbon-ion beam and an active distribution system. Three main features of this pilot facility are worth mentioning:

- the active ‘raster’ scanning system;
- the fully automatic control of the GSI accelerator complex, which can be handled by a trained operator of standard X-ray equipment;
- the two gamma-ray detectors placed above and below the patient to determine ‘on-line’ the exact location and shape of the irradiated volume because, when penetrating the body, the incident carbon ions fragment into $\beta^+$ radioactive nuclei, mainly $^{11}$C, which can be detected by a standard PET technique.

The pilot project has been a great success: about 100 patients had been treated by summer 2001. Hence, in September 1998 GSI, the Oncological Clinic of the University of Heidelberg and the Deutsches Krebsforschungszentrum (DKFZ) presented to the authorities a project for a hospital-based proton and ion centre capable of treating 1000 patients per year (Fig. 7.8). The approval is expected by October 2001.

7.5.2 The Proton–Ion Medical Machine Study (PIMMS)

At the end of 1995 the TERA Foundation, together with the AUSTRON project, attracted CERN’s interest in the design of an optimized synchrotron for light-ion therapy, to be built by those European countries investing the required funds. At the beginning of 1996 the CERN management agreed on the proposal and a study of such a synchroton was started at CERN under the leadership of Phil Bryant.

PIMMS is a collaboration between CERN, Med-AUSTRON (Austria), Oncology 2000 (Czech Republic) and TERA (Italy). GSI experts brought their competence to the many meetings held in the years 1996–1999 and some of them have been members of the Project Advisory Committee. The study group has published two general volumes [15] and many detailed articles.

For hadrontherapy with active spreading the main issue is the time uniformity of the spill since, mainly because of the magnet ripple, synchrotrons have time structures at many frequencies. This complicates the accurate measurement of the dose during the active spreading
of the clinical pencil beams. Thus the PIMMS synchrotron has been designed starting from the clinical beam and going ‘backwards’. Its extraction is based on a special optics. During extraction, all magnet currents are kept constant and the energy of the circulating particles is slowly increased by the induced electric field due to a ‘betatron core’.

The PIMMS layout is shown in Fig. 7.9. It has to be underlined that the mandate of the group was to design an optimized centre without considering budget and space constraints. The idea was that the European groups interested in constructing their ion therapy centre could adapt the PIMMS proposal to their needs and budget.

In 1998 the Med-AUSTRON project, which concerns the realization of a proton and carbon-ion centre at Wiener Neustadt in southern Austria, proposed to the Austrian authorities a staged construction of the PIMMS project [16]. This staging allows an initial investment that is smaller than the 120 million euros needed for the construction of the centre shown in Fig. 7.9.

The Italian TERA Foundation aims at the construction in Milan of CNAO, the national centre for oncological hadrontherapy, a health-care and research facility equipped with both proton and ion beams so to allow the treatment of more than 1000 patients per year. In 1997 a first proposal was presented to the Italian authorities [17].

In the years 1998–2000 a new design was prepared, based on the PIMMS synchrotron. To reduce the overall cost the surface covered by the two-floor underground bunker was reduced to 2900 m² and a single injector was used for injecting both protons and ions. As shown in Fig. 7.10, the injector was located inside the synchrotron and a very compact configuration of the beam line was chosen. For the 7 MeV/u injector the GSI was adopted (Fig. 7.8). To construct Phase 1 of CNAO 60 million euros are needed. In 200: the Italian Ministry of Public Health allocate 20.6 million euros to a foundation created to construct the centre. The board of the CNAO Foundation is made up of the directors or presidents of two university hospitals (Milan and Pavia), an oncological institute (European Institute of Oncology), the National Institute of Neurology and TERA.

The PIMMS design is at the core of other
hadrontherapy projects. In 1998 the Université Claude Bernard of Lyon asked TERA to produce a preliminary design of a centre for carbon ions. In summer 2000 the university signed a contract with IN2P3 and CEA with the aim of having a preliminary proposal ready by autumn 2001. In spring 2001 the project group decided that the proposal will be based on the PIMMS synchrotron and that, as proposed by TERA, the GSI 7 MeV/u injector will be used.

In 1999 scientists from the Karolinska Institute and Hospital in Stockholm and from TERA decided to prepare jointly a proposal for a light-ion facility to be built very close to the radiotherapy department of the Karolinska Hospital. A recent report [7] explains the rationale of such a centre and includes the plan shown in Fig. 7.11.

7.6 Conclusions

Since 1998, when a similar review was published [18], many hadrontherapy centres have either started treating patients or have been proposed to the local authorities. Japan is the country in which this novel radiotherapy technique is most widespread, but recently interest has been growing in Europe as a consequence of the collaboration established between nuclear research laboratories and oncological hospitals.
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8. The In-flux of Nuclear Science to Radiobiology

G. Taucher-Scholz, G. Kraft, B. Michael and M. Belli

8.1 Introduction

Radiobiology is mostly concerned with uses of X-rays in medical diagnosis, like fluoroscopy or computerized tomography (CT), both of which are products of atomic science. Although nuclear science entered radiobiology rather late, around 1950, it did so with great intensity.

At this time, the long-term consequences of the nuclear bombing of Hiroshima and Nagasaki became evident and triggered public discussion. The catastrophe demonstrated that the nuclear bomb was not just a more efficient explosive than dynamite, but also had unknown biological consequences that had to be studied in radiobiological experiments (see Chapter 12). Thus, it was in a rather indirect way that nuclear physics stimulated radiobiological research. A more direct way followed when particle accelerators became available for radiobiological research and — more importantly — for use in the radiotherapy of tumours in critical regions (see Chapter 6). Finally, the advantageous properties of particle beams as a tool for radiobiological research have been discovered very recently, allowing precise research of the repair mechanism and signal transduction of biological cells.

8.2 Relative biological efficiency

In radiation protection, radiotherapy and basic radiobiological research, a big difference can be observed in the effectiveness of particle radiation compared to X-rays. This difference can potentiate the genotoxic and carcinogenic effects but can also be used for more efficient tumour therapy. In research practice, the concept of Relative Biological Efficiency (RBE) is introduced, defined as the ratio of X-ray dose to particle dose necessary to obtain the same biological effect [1]: \( RBE = D_x / D_p \). Experimental studies found RBE values around 3–4 but higher values have been measured at low doses. It has been demonstrated experimentally that RBE depends on biological reactions as well as on physical parameters of the applied radiation field such as particle energy and atomic number, indicating a difference in the structure of the energy deposition at a microscopic level.¹

For both X-rays and particles the biological effect is due to ionization events, mainly caused by liberated electrons, which can take place in the DNA itself and the water molecules around. Once a free electron is produced, its biological action does not depend on its origin: X-rays or particles [2]. The main difference between the two radiation types, however, is the local distribution of the ionization events as shown in Fig. 8.1. Particles form tracks with high ionization densities corresponding to local doses of thousands of Gys in the centre and a steep decrease towards the maximum radius, with a zero dose outside the tracks. In contrast, for sparsely ionizing radiation, the dose is more or less homogeneously distributed. Biological experiments at cellular and DNA levels, however, have shown that the increase in RBE is most pronounced for biological systems having a large repair capacity, while RBE remains nearly constant (equal to one) for repair-deficient systems. This shows that the processing of the damage by cellular repair systems is of utmost importance for understanding the increased RBE of particles.

¹Radiation protection introduced a quality factor \( Q \) different from RBE. The dose \( D \), given as the energy deposited in a mass unit, has to be multiplied by \( Q \) in order to get the biologically equivalent dose \( H \): \( H [Sv] = Q \times D [Gy] \). The dose is given in gray (1 Gy = 1 J/kg), the equivalent dose in sievert (Sv) and the dimensionless quality factor is defined and fixed by law to values that, for safety reasons, should represent an upper limit [1].
8.3 Induction and repair of DNA damage after heavy-ion radiation

Among the DNA lesions induced by ionizing radiation, mainly base-pair and strand-break damage, the most deleterious one is the DNA double-strand break (DSB). If left unrepaired, this lesion can result in a loss of genetic information, leading either to cell death or — if mis-repaired — to mutations and the induction of cancer. In mammalian cell systems, the repair of DSBs can essentially be observed in two different ways: at the molecular DNA and at the chromosomal level [3]. The DNA in the nucleus is folded and packed with proteins and referred to as chromatin. It gets further compacted and condensed to chromosomes just before cell division. Only in this stage are chromosomes visible under the microscope and DNA damage can be scored as chromosome aberration. Fragmented DNA molecules and incorrect processing of DNA lead to abnormal chromosomes and aberrations. However, many of the most heavily damaged cells are hindered in performing this condensation. As a result, severely damaged cells obtained after exposure to heavy particles are not scored and the ion irradiation was believed to be less dangerous with regard to genetic alterations. Recent experiments, integrating chromosome aberrations expressed over a longer time period, revealed that for particles, a greater RBE can also be observed for chromosomal lesions as for cell killing [4].

At the molecular DNA level, a similar problem arises. Using conventional electrophoretic separation, no increase in the number of DNA DSBs was found that could account for the increased efficiency in cell killing. If the cells are incubated for repair after irradiation the DNA lesions induced by particles are not as well repaired as damage induced by X-rays. In addition, DNA fragment size distributions have recently demonstrated that the correlated production of breaks after exposure to particle irradiation yields a higher proportion of small fragments than an exposure to X-rays. This proximity of DSBs places an additional demand on cellular repair systems. Most likely, the processing of DNA damage after irradiation provides the link to the cellular reaction.

Indeed, there is evidence for a higher fraction of breaks left unrepaired after irradiation with low-energy charged particles, showing a maximum of cell-inactivation efficiency. This finding helps to explain the observed enhanced RBE
for cell inactivation and it has to be concluded that the structure of the particle-produced DNA damage is more complex and less repairable than the damage produced by X-rays. However, the structure of these complex lesions is a subject of research and not known yet. Exposure to ion beams from accelerators can be executed with varying energies and atomic numbers, changing the extent and intensity of the damaged sites. Thus, particle accelerators are used to elucidate the structure of DNA damage and its influence on correct repair, which is a basic problem of the action of ionizing radiation.

8.4 Nuclear dynamics of protein involved in the DNA damage response

The nature of localized dose deposition along the tracks of charged particles is expected to induce complex lesions, representing a challenge to the cellular repair machinery. On the other hand, just this production of DNA damage within well-defined regions of the cell nucleus following exposure to particle radiation provides the means to study the dynamics of protein interactions involved in the response of the cell to this injury. Using fluorescence-labelled antibodies against the various repair proteins, the location of sites of particle-induced damage can be observed under the microscope. In this way, for the first time, the inhomogeneous microscopic dose deposition pattern characteristic to particle radiation could be visualized as a strictly localized, discrete biological response confined to the ion tracks traversing the cell nucleus. This is shown in Fig. 8.2 for the p21-protein (CDKN1A), one of the key proteins involved in the inhibition of cell proliferation after exposure to radiation to allow the repair of DNA damage.

8.5 Modelling the particle response

There are many theoretical approaches to correlate these inhomogeneities in the microdistribution of the dose deposited by particles directly to the biological result [5, 6]. In a recent theory, the Local Effect Model (LEM), the cell killing by particle exposure is calculated from X-ray efficiency in a way that fully includes repair
and microdosimetric dose distribution [7].

In LEM, the increased biological effect of particles is calculated on the basis of three measurable quantities: the size of the cell nucleus, the X-ray dose effect curve and the radial dose distribution. The effect of changes in repair is implicitly contained in the non-linearity of the X-ray dose effect curve. There, the same increment of the dose is more efficient in cell killing at higher than at lower doses if repair is predominant, yielding a linearquadratic dose response curve. In case of no or little repair this difference disappears, X-ray dose effect become linear and RBE remains constant. In a Monte Carlo calculation, tracks traverse cell nuclei and the dose distribution of the overlapping tracks is folded with the X-ray dose effect curves to determine the damage probability (Fig. 8.3).

Finally, these probabilities are integrated over the complete cell nucleus. Using this approach all the dependences on dose, energy and atomic number can be reproduced very well. For cell inactivation, LEM has been very successfully used in carbon radiotherapy to calculate the RBE for cell killing over the target volume and for late effects in the surrounding normal tissue. Numerous tests with cultured cells prior to the therapy also confirmed the LEM approach. Although the model calculation can be used to transfer the X-ray efficiency to the response to particle exposure very successfully, LEM does not answer questions regarding the molecular nature of primary damage, the pathways of repair or the involved proteins that were posed in the earlier models [5, 6].

8.6 Bystander effects of radiation

8.6.1 A new mechanism for radiation damage

The mechanisms and modelling described above relate to what is called the ‘classical model’ of radiobiological effects. One of the concepts on which this model is based is that each cell individually presents a target which either responds or does not respond to irradiation independently of its neighbours. In many situations, the classical model continues to serve well as a descriptor and a predictor of the biological actions of various types of radiation field. However, over the past decade it has become clear that it does not account for certain types of response, particularly at low doses.

Up until the early 1990s, it was generally thought that the damaging biological effects of radiation occurred only very close to the tracks along which energy is deposited. Thus it was believed that damage was only induced within nanometre distances of where the primary and secondary charged particles passed through the cell. It was also believed that all of the significant biological effects of radiation derived from deposition of energy within the nucleus of the cell and not in the cytoplasm and that genomic DNA was much more sensitive than any other constituent. In particular, DSBs were
considered to be the key initial lesions which, if not correctly repaired, could lead either to cell death or to permanent changes in the irradiated cell and in its descendants. These changes included chromosomal aberrations, mutations and malignant transformation. There had been some indications of radiation effects that were not consistent with the classical model, for example evidence for the induction of chromosomal damage in cells that had not been exposed to radiation if they were placed in contact with plasma from irradiated individuals. There was also evidence from radiotherapy of changes arising in tissues that were distant from the treated field (‘abscopal effects’). However, such effects were considered not to be of major importance and, in general, to contribute very little to the overall biological actions of radiation.

In 1992, a report was published by Naga-sawa and Little [8] which appeared to challenge the established classical model of direct damage. The authors reported that when Chinese hamster cells in culture were exposed to low doses of alpha-particles such that on average only 1% of cell nuclei were actually traversed, about 30% of the cells subsequently showed chromosomal damage in the form of sister chromatid exchanges. Thus, their data showed that DNA damage had been induced in many more cells than the fraction that had had energy deposited in their nuclei. This was a surprising finding and appeared to conflict with the direct damage model. Over the next few years, several further reports from Little’s and other laboratories confirmed that cells do indeed incur damage as a consequence of being in the neighbourhood of irradiated cells. The novel concept of a ‘bystander effect’ of radiation gained acceptance. Cellular responses induced via bystander mechanisms have been shown to include the induction of chromosomal aberrations, mutations, cell death, apoptosis (or programmed cell death), malignant transformation and genomic instability. Figure 8.4 illustrates the bystander effect.

8.6.2 What is the mechanism?

The bystander effect has been found to occur in a number of different cell systems and several types of signalling mechanism appear to be involved. In some systems, where there is direct cell-to-cell contact, there is evidence that signalling occurs via the gap junctions which certain cell types use to communicate. Evidence from other studies, where cells are not in direct contact, points to the involvement of a factor transmitted via the culture medium. There is evidence for the involvement of cytokines, which are small proteins or biological factors that are released by cells and have specific roles in cell-to-cell communication. Other evidence points to the role of reactive oxygen species (ROS). Currently, much of the research on the bystander effect is concerned with identifying the signalling mechanisms. Most of the information gained so far has come from in vitro systems and there is a pressing need to study bystander responses either directly in vivo or in model tissue systems.

8.6.3 Comparison with direct effects

Research has already revealed several important differences between bystander and direct effects of radiation. Perhaps the most striking difference is found in the shapes of the dose–effect relationships, illustrated in Fig. 8.5.

With direct effects, for example cell kill, as the dose of radiation is increased the
Proportion of cells showing a response continues to increase progressively until virtually all cells in the exposed population are affected. This can be explained in terms of the particulate nature of radiation and each cell behaving as a sensitive target with an equal probability of being inactivated if hit by a particle track. Thus the survivors are those that by chance escape a lethal hit. In contrast, bystander effects are characterized by a low threshold dose above which the response exhibits a plateau. Further increases in dose produce little or no increase in the fraction of cells that respond. There are two main theories to explain the plateau. One is that a pre-existing fraction of the cell population is sensitive to the bystander signal. The other is that the cell population as a whole responds to the bystander signal, but also generates an inhibitory signal which limits the response to a certain fraction.

8.6.4 Potential importance

There are two main areas in which bystander effects may have a significant role. The first is in relation to the risks associated with low-dose exposures. This is because at low doses the arrival of radiation tracks at the level of cells in human tissues is, on average, well separated both in space and time. For example, natural background exposure amounts to an average of about one electron track per cell per year and one alpha-particle per cell per century. Conventional estimates of radiation risk indicate that natural background radiation contributes about 3% of all human cancers, but whether this is an over- or underestimate depends in part on assumptions about the biological actions of single tracks.

Bystander effects may also have a role in some forms of radiotherapy. For example, cancer treatments using targeted radionuclides may fail to reach all parts of the tumour, in which case any transmission of lethal effect from hit to non-hit tumour cells would be beneficial. The same consideration may apply in boron neutron capture therapy (BNCT).

8.7 Microbeams for research in radiation biology

Over the last decade, there has been a surge of activity in the development of microbeams for research in radiation biology. More than a dozen high-tech microbeams have either been constructed or are being planned for research centres around the world. The stimulus for their development stems from many of the questions outlined above. In particular, they promise to help answer the fundamental question, “What are the effects of single charged-particle tracks delivered to specific regions of the cell?” This may seem a rather esoteric question, but the answers to it will provide a Rosetta stone which will unlock many of the remaining secrets of the biological actions of radiation.
8.8 Modern microbeam developments

The concept of radiobiological microbeams is not new [9], but the current generation of systems [10]–[12], has been able to take advantage of modern developments in technology, particularly with regard to image capture and processing and computer control. Today's microbeams have targeting accuracies ranging from several microns to the sub-micron level and operate under programmed control to irradiate up to about 10,000 cells per hour. Ions ranging from protons to argon nuclei are available at the various installations. It is standard to count the number of ions and to interrupt the exposure of each cell when it has received the intended number of traversals. In this way, it is possible to deliver strict numbers of particles to cells and defeat the confounding effects of random (i.e. Poisson-distributed) particle arrivals that occur in experiments using conventional sources. The main elements of a charged-particle microbeam are shown in Fig. 8.6.

The ability to deliver strictly single particles to individual cells is of direct relevance to the determination of RBE for different types of radiation, as outlined above. For radiation protection purposes, radiation quality factors, which are decided from our knowledge of RBE, need to be estimated for the lowest possible dose level and this corresponds to the biological actions of a single track.

Another important feature of microbeam systems is that they are able to revisit individual cells or their progeny after division. This means that the effects of irradiation on cells that have been targeted and on others that have not can be followed clearly and distinguished, for example in experiments on the bystander effect. It also means that in studies of the nuclear dynamics of proteins involved in the DNA damage response, referred to above, it is possible to visualize the localization of response proteins in direct microscopic relation to the path of the radiation track that initiated the damage to which they have responded.

There is also currently much interest in the application of microbeam methods using focused soft X-rays or electrons. The aims here are to exploit the high targeting accuracy achievable with focused X-rays (about 30 nanometres) and to explore low-dose effects of low-LET radiations. Here the aim is ultimately to be able to determine the actions of the passage of a single electron track through the cell, a topic of particular relevance in understanding and predicting low-dose radiation risk.

Figure 8.6: A charged-particle microbeam for individual cell irradiation showing the microcollimator used to shape the beam, the particle detector used to count the ions delivered, the cell dish, microscope and imaging system.
Bibliography


9. Radiobiology for Space Research

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9.1 Introduction

Life on earth has developed under gravity, humidity and atmospheric air pressure. In space, this environment has to be reproduced artificially in order to provide living conditions for astronauts. The radiation environment, however, is present in both cases but differs in quality and intensity. On earth, the natural and artificial radiation is mainly caused by sparsely ionizing radiation like X- or γ-rays. Additionally, densely ionizing radiation is created by a few α-emitters like radon. For both radiation qualities combined, the European average mean dose is 1 mSv per year. In space, the radiation burden is much bigger and depends on altitude and inclination of the space orbit. For high-altitude flights beyond the magnetic shielding of the earth, cosmic radiation, i.e. high-energy particles from protons to iron ions, predominate. Space radiation reaches its maximum values at a solar particle event where lethal doses can be delivered within an hour or less. These events, however, are rather unlikely. Much more important is the permanent, i.e. protracted, exposure to the low-dose radiation of the heavy charged particles of galactic cosmic rays. Because of their high local dose these particles are able to create local damage in bio-molecules that can manifest itself in long-term alterations like genetic mutation and cancer induction. It is the induction of these biological changes that determines the general risk of long-term missions. For low-altitude flights such as MIR or space-station orbits, trapped electrons and protons of solar origin predominate. To study the biological radiation response, especially genetic alterations and cancerogenesis, X-ray experiments can be performed in order to mimic sparsely ionizing electrons. And in order to gain more insight in the biological action of galactic cosmic rays, accelerator experiments are necessary.

9.2 The radiation field

Depending on the distance from earth the composition of the radiation field varies because particles are trapped in the geomagnetic field. At low earth orbit (LEO), trapped protons and electrons from the radiation belt predominate.

Apart from the galactic cosmic rays there are also the protons of solar eruption events, the solar flares. These solar particle events are statically distributed and it cannot be predicted whether or not they will occur during a specific space mission. But in the long run, a stable period of eleven years for maximum solar activity has been observed. During solar particle events, a dose of a fraction of one gray up to the gray region can be delivered in a short time of a few hours or even less. Therefore, solar particle events could be life threatening in extreme cases, but viewed over the long term they are so rare that they contribute only a small fraction of the overall radiation burden.

The main contribution of dose during a space mission outside the magnetic shielding of the earth originates from galactic cosmic rays (Fig. 9.1), which are heavy charged particles from the most frequent protons up to iron ions. Ions heavier than iron are several orders of magnitude less frequent since they originate from supernova explosions and cannot be synthesized by exothermic nuclear fusion reactions like iron and the elements lighter than iron.

The galactic cosmic radiation (GCR) has an energy spectrum with a broad maximum at a few hundred MeV per nucleon and a steep and continuous decay towards higher energies (Fig. 9.2). This high-energy tail has been
measured up to the TeV region and is a big problem for effective shielding. Because the cross-sections for nuclear fragmentation are still significant for the ions of these extremely high energies, any shielding layer introduced to stop the low-energy particles also produces showers of nuclear fragments from the high-energy particle impact.

Transport calculations for the GCR spectrum in various shielding materials like aluminium showed that after a small benefit in the first thin shielding layers, thicker absorbers do not produce a net reduction of the biological effect that correlates reasonably with the increasing mass of the shielding material. The decrease in the amount of low-energy particles is almost compensated for by the increase in nuclear fragmentation. For details of these very complex but important ion transport avalanches we ask the reader to refer to the comprehensive article by Wilson et al. [1].

The fluence distribution of protons is three orders of magnitude higher than that of iron ions but the energy deposition, i.e. the dose of a single particle, depends on the square of the atomic number. Therefore, the difference between protons and iron in their frequency contribution is nearly compensated for by the dose. Taking the change in the relative biological efficiency into account the fraction of iron particles becomes as large as that of protons. For low earth orbit (LEO), the total dose per day is about 1 mSv behind a shielding of 1 g/cm² of aluminium. This is the average dose per year on earth. Spaceflights are on average 300 times more exposure-intensive than our daily life. But the actual value of a space mission very much depends on the altitude of the flight and on the inclination of the route: shielding is drastically reduced at the magnetic poles, so an orbit over the poles results in a greater exposure than an equatorial flight.

Outside the geomagnetic shielding the particle composition of the GCR becomes more relevant and the estimation of the biological effect is determined rather by the hit probability of the critical target inside the cells, i.e. the cell nuclei, than by a dose averaged over a long time. For example, a three-year mission to Mars was calculated to produce 400 proton and 40 helium-ion traversals through the nucleus of each cell of the human body [2]. This number decreases for heavy ions like iron to a hit.
probability of 3%. According to these statistics, every cell nucleus will be hit by a proton once every three days and by a helium ion once every thirty days. Considering the high hit frequency and the large number of cells at risk it is very evident that a non-protracted exposure would be lethal while the distribution over a period of three years allows repair with a good chance of the traversed cells surviving. However, the long-term consequences like cancer induction or genetic mutation determine the risk. It is well-known that a macroscopic tumour may originate from only one transformed cell. If a single mutated cell survives cancer may develop. Consequently, cancer is determined more by the biological processing than by mere induction statistics of DNA damage.

9.3 Health effects of cosmic radiation

Depending on dose, acute or long-term effects can be induced by radiation exposure. Acute effects like nausea, vomiting, skin irritation, and depletion of white blood cells occur at doses of about 1.5 Gy or more. These high doses are produced by solar storms. Astronauts can be protected in storm shelters from the low-energy particles because these events are preceded by an enhanced solar activity.

Long-term effects are genetic alterations, cancer induction, damage to the central nervous system and peripheral neurons, and accelerated aging. Among these effects, cancerogenesis and neural damage seem to be the most important. The uncertainties for determining the risk of long-term effects are large and the risk estimation is mainly based on epidemiological data from the atomic bomb survivors (see Chapter 12). However, the radiation quality in these historical data differs greatly from that in the space radiation environment. Our current knowledge about the effects of low space-radiation doses is compiled from different sources. There are the observations of astronauts returning from long-term space missions such as at MIR [3, 4] or the previous moon expeditions. In addition, accelerator experiments are performed with human blood cells and various other cell types. In both cases, the induction of chromosome aberration is studied because it is believed to be the most accurate and sensitive indicator of genetic mutations in general and for cancer induction in particular.

Additional information is gained from the few accidental exposures of nuclear technology workers. But again, these cases are rare and have another radiation quality than space radiation. A special problem is the continuous or protracted irradiation with low doses in space. This situation is completely different from an acute exposure in nuclear accidents and it is not possible to simulate a many-month protraction in cell experiments. Normally, the experiments are performed at high doses delivered in a short time and model assumptions have to be made in order to extrapolate to low-dose protracted exposure.

9.4 Biological dosimetry

Chromosomes become visible when the cells divide and the DNA is distributed in equal proportions to the daughter cells. In this stage, DNA condenses to small bodies, the chromosomes, that are visible under the microscope. Pertinent DNA damage is expressed in chromosomal aberrations [3–5]. Chromosome damage is therefore an indicator for DNA mutation and genetic alteration.

In the past, so-called non-transmissible aberrations like rings or dicentrics have been used to measure chromosomal changes because such aberrations are easy to score. However, cells carrying these aberrations have only a small chance of surviving the next cell divisions and are not representative for DNA damage leading to long-term consequences. More recently, new techniques like chromosome painting [3, 5] have allowed us to study the induction of transmissible chromosome aberrations like translocations. Translocations originate from at least two DNA double strand breaks that are misrepaired in one or two chromosomes (Fig. 9.3). Since no genetic information is lost, cells carrying this new DNA configuration have a high chance of survival and pass the aberrations on to further cell generations. In addition, translocations are very
frequently involved in the initiation of cancer, i.e. when a strong promoter is linked to an oncogen stimulating infinite cell growth.

The recent development of the multicolour fluorescence in situ hybridization (M-FISH) allows us to paint each chromosome in one specific colour and to observe the exchange of material between chromosomes, as shown in Fig. 9.3.

![Figure 9.3](image)

Figure 9.3: Originally, each chromosome pair is painted in one colour. Chromosomes that are multicoloured result from the interchange of parts of two or more different chromosomes. Cells showing translocations normally remain viable but may induce cancer if oncogenes are connected to strong promoters.

9.5 Biodosimetry of exposed persons

Men returning from extended space missions, such as the moon expedition or extended stays at MIR, carry chromosome aberrations in their blood cells as illustrated in Fig. 9.4 and described in detail in Refs. [3, 4]. However, it is almost impossible to deduce dose effect curves from these measurements. First, because the aberration rate is fortunately low and second, because the dose and particularly the composition of the radiation environment, i.e. the radiation quality, is not known in detail. It is possible, however, to analyse these data in model calculations using X-ray dose effect curves and quality weighting factors — and other plausible assumptions — in order to calculate expected values for chromosome damage. Then, the prediction can be compared with the findings obtained from the blood analysis of the cosmonauts. Basic assumptions in these estimations have to be made for the extrapolation of partial or inhomogeneous exposure to total body exposure, from high dose rates to low dose rates and from X-rays to particles.

![Figure 9.4](image)

Figure 9.4: Lymphocyte chromosomes of a Russian cosmonaut clearly show aberrations after a six-month MIR mission. Arrows indicate the chromosome aberrations detected in these cells.

At least the last extrapolation, i.e. the change in radiation quality, can be studied in accelerator experiments where the differences in efficiency of particle radiation can be assessed. However, these experiments are very tedious. Owing to the complexity of the particle-induced aberrations, the time-scale of their expression is drastically increased (see Chapter 8) and it is difficult to obtain in vitro experimental data that can be used for risk estimation. Still, these accelerator experiments are the only source of reliable data that can help to understand the mechanics of genetic mutations and cancer induction. Therefore, the accelerators for nuclear and high-energy physics will always have a small but important experimental activity in radiobiology.
9.6 Conclusions

Human beings in space are exposed on average to a radiation dose that is 300 times higher than on earth, thus receiving a daily exposure equal to the annual radiation burden on earth. In addition, the two radiation environments differ in quality and space radiation is dominated by high-energy particles. It has been proven that this radiation exposure produces genetic alterations including cancer. Other stress factors of space travel like microgravity, gas environment and artificial nutrition also generate biological and physical impairments. But physiological recovery is reached shortly after landing. With regard to genetic damage, transmissible chromosome aberrations are pertinently present during and after the space mission. At present, the mechanisms and the extent of this damage are not known in detail. Today, only high-energy particle accelerators for nuclear physics experiments allow these questions to be studied in detail, being the only source of information that allows us to optimize the shielding conditions for humans in space.
Bibliography


10. Isotopic Tracers in Biomedical Applications

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10.1 Historical perspective

At the beginning of the 1910s experiments conducted by F. Soddy gave the first demonstration that most of the elements in nature are composed of atoms identical from the chemical point of view but slightly different in weight. These characteristics were later explained by the fact that each element is identified by the number of protons in each atom, although the number of neutrons may vary. The term ‘isotope’ was introduced to account for this variation. In 1934, H.C. Urey was awarded the Nobel Prize in Chemistry for the discovery of deuterium. Shortly after, the idea of using stable isotopes in kinetic/dynamic investigations found useful applications in the early studies by R. Schoenheimer and D. Rittenberg on fat metabolism in mice with deuterium, and continued with studies in nutrition by using $^{15}\text{N}$, $^{13}\text{C}$ and $^{18}\text{O}$. Over the following three decades, with the advent of scintillation counting and the availability of radioactive isotopes, the use of stable isotopes was replaced by the radiotracer technique. The principle of use of radiotracers was actually conceived by G. de Hevesy in the early 1910s; however, their widespread use in clinical/biomedical applications really started after the development of the cyclotron as a source for massive production, and quickly expanded with the production of radionuclides in nuclear reactors.

The stable-isotope approach remained confined to light elements such as $\text{H}$, $\text{O}$, $\text{C}$ and $\text{N}$ (in particular through lack of suitable radioisotopes of $\text{N}$ and $\text{O}$) in food science with some tentative applications towards other elements, such as the investigations by Lowman and Krivit in the 1960s on plasma clearance of iron. Only in the mid-1970s was there renewed interest in stable isotopes. This was due to a greater sensibility of the scientific community towards the use of radioactive substances in healthy volunteers, as well as to the availability of new and improved analytical techniques such as inductively coupled plasma mass spectrometry (ICPMS) and accelerator mass spectrometry (AMS).

Stable isotopes are now mainly used in the fields of nutrition and physiology for investigations on microminerals and essential trace minerals, but applications to selected issues can be found. For example, the biokinetics in humans of fallout radionuclides is studied by combining the use of stable isotopes as tracers and complementary analytical techniques such as charged particle activation analysis (CPAA), thermal ionization mass spectrometry (TIMS) and ICPMS.

This chapter will exclusively deal with non-imaging applications of isotopic tracers, and particularly with metabolic and molecular studies in the life-science field.

10.2 Principles of tracer use

Tracer methods find applications in nearly every field of science, be it in life-science fields such as medicine, biology, physiology, nutrition, toxicology and biotechnology, or more technical areas such as physics, chemistry, agriculture, geo-science and engineering which have now become an integral part of everyday life.

The common issues for all these applications concern the possibility to trace the entity of interest, called ‘tracee’, which may be a substance, or a component of a substance, like a radical, a molecule or an atom. An ideal tracer has the same physical, chemical or biological properties of interest as the tracee, but it presents some peculiar characteristic that enables its detection in the system where the tracee is also present.
The production of an isotopic tracer involves the substitution of one or more naturally occurring atoms in specific positions in the tracee molecule with an isotope of that atom with a less common abundance. Either stable or radioactive isotopes can be used as tracers. Mass differences of isotopes are due to different numbers of nuclear neutrons, so that the chemical properties are not affected. Both stable and radioactive isotopes of an element take part in the same chemical reactions of the element.

The use of a labelled tracer requires the assumption that the labelled molecule, or atom, will not be discriminated from the unlabelled and will trace the position or movement of the unlabelled molecules. Some isotopic effects like evaporation processes or root uptake in plants can however be observed, especially for light elements, and should be taken into account.

10.3 Choice of tracer approach

The principles of the use of stable and/or radioactive isotopes in tracer studies are similar; however fundamental differences exist which must be kept in mind in the design of a tracer experiment or in the choice of a tracer application. The optimization process should also take into consideration safety aspects, measurement conditions and equipment requirements in view of the expected or required sensitivity.

The first obvious difference is that stable isotopes are non-radioactive; so in case of in vivo application, there is no radiation hazard for the investigated subjects, with undeniable advantages for particular groups of the population such as pregnant women and newborns. At the same time, this characteristic demands measuring techniques which are usually, though not always, more complex than those used for radioactive ones. For in vitro or non-human applications (e.g. molecule labelling, DNA sequencing, environmental studies) the use of stable and radioactive tracers can be seen as a complementary approach, although the protection of the working personnel exposed to radiation hazard should be taken into account.

The choice depends also on the isotopic availability. Monoisotopic elements, like Be, F, Na, Al, P, Mn, Sc, Co, clearly cannot be used as they are indistinguishable from the intrinsic content. When more stable isotopes are present, as for the five elements of main interest in the life-science field (H, C, O, N, S), the possible tracer will present a modified isotopic composition, with the abundance of one isotope highly enriched with respect to the natural one. Multiple labelling studies, as required for example in nutrition and physiology, can be performed only when a sufficient number of isotopes and suitable techniques for their simultaneous discrimination are available.

Most of the chemical elements present one or more radioisotopes, either naturally occurring or artificially produced. Artificial radiotracers may be considered free from any interference due to intrinsic content in the sample, in the other cases a non-null blank value can be expected. Although this contribution may significantly affect the results, it can be evaluated on the basis of isotopic ratio measurements, which require more accurate and time-consuming techniques.

Further factors affecting the choice of radiotracers may be the availability of isotopes with too short (28Mg, 64Cu) or too long (65Zn, 46Ca, 55Fe, 63Ni) half-lives in relation to the planned study, or the high costs of the infrastructure and obligatory licences required by the regulations. Stable isotopes, on their side, might be quite expensive (depending on the degree of enrichment desired) and the lower sensitivities of the analytical techniques may demand the administration of significant masses, which might not strictly fulfill the tracer assumption.

In summary, for in vitro studies radioactive isotopes may be the tracers of choice; for environmental studies (soil, water, plants, animals) the advantage of radiotracers should be balanced against possible risks for the exposed personnel (including environmental release); and in human studies the superior diagnostic information they provide must be justified by a soundly recognized benefit for the exposed subject or for the community as a whole. Otherwise, stable isotopes should be preferred, and particularly in three cases:
when the information they provide, combined with a sufficiently refined data analysis, is satisfactory for the study aim;

- when the exposed subjects (volunteers) are not the ones directly benefiting from the study (tracer kinetic and metabolism research studies in physiology, nutrition, etc.);

- when no suitable radiotracers are available.

10.4 Tracer measurement

Radioactive tracers can be detected on the basis of their decay properties, as described in Section 10.4.1. When the half-life is too long or the amount too small to enable a statistically significant measurement, techniques like those used for stable isotopes are needed, which make use of isotopic nuclear properties.

10.4.1 Intrinsic radioactive characteristics

The radiotracer is generally the only source of radioactivity present in the sample, apart from the natural background. The decay counting technique, properly corrected for background activity, is therefore sufficient to obtain relative or absolute determinations. The most appropriate radiometric method will obviously be chosen according to the type and energy of radioactive emissions, as well as the method’s sensitivity. If more accurate information is required, such as activity distribution in an inhomogeneous sample or in a living organism, autoradiography or external imaging (e.g. using gamma-camera as in nuclear medicine) can be employed.

If two or more radioactive tracers are simultaneously present in the sample, simple counting is of no use. The different contributions can be discriminated by repeated counting measurements on the basis of the different half-lives, or by spectrometric measurements on the basis of the characteristic emissions. Routine methodologies involve predominantly the use of gas-filled detectors, organic or inorganic scintillators. Because of the negligible background matrix, the high-resolution properties of solid-state detectors are not required for gamma spectrometry.

10.4.2 Stimulated response

For the detection of a stable isotope or of a radioisotope with a low activity not suitable for measurement, a characteristic (isotopic) response can be stimulated:

- through application of a series of electrical and/or magnetic fields to the ionized sample in order to discriminate between atoms or molecules with different mass-to-charge ratios, thus distinguishing the tracer on the basis of its nuclear mass;

- through induction of nuclear reactions after bombardment of the sample with projectiles of given type and energy, thus distinguishing the tracer on the basis of the decay emissions of the radioactive reaction product.

In the first case, the tracer content in the sample may be determined by measuring the current intensity corresponding to the tracer ions using detectors such as Faraday cups or secondary electron multipliers.

As for the induction of nuclear reactions, all sample constituents can be activated (each with its own probability) and therefore the reaction product of interest may be accompanied by a series of other radioisotopes which can cause interference, thus requiring high-resolution spectrometric techniques, typically gamma spectrometry with semiconductor diode detectors. The necessity of standard nuclear techniques for a successful application of the tracer methodologies is immediately evident.

For mass spectrometry this is particularly valid when accelerators and detection techniques such as time-of-flight measurements, developed for basic nuclear research, are employed.

The peculiarity of using such machines lies in the high energy attained by the accelerated ions (in the MeV order of magnitude), which improves significantly the resolution and therefore the accuracy and sensitivity of the technique.

AMS was developed mainly for radiocarbon dating in geochronology studies with a sensitivity many orders of magnitude larger than the
usual counting techniques, and more recently applications for the detection of stable or long-lived radioactive isotopes of elements such as H, Be, C, Al, Cl, Ca, Ni, I, U of interest for bio-analytical tracing in the life sciences have emerged. The accelerators employed for AMS range from simple Van de Graaff or Tandem machines with terminal voltages of a few MV to more energetic cyclotrons or linear accelerators.

Another nuclear technique which has been recently applied in bioracera studies is neutron scattering analysis for the determination of shapes and dispositions of complex molecules labelled with deuterium. This has been made possible by recent advances in sources and instrumentation for neutron scattering as well as in biotechnology, which have made sample preparation and deuterium labelling easier and cheaper. Also the understanding of the coordinated functions of interacting biomolecules could benefit from a broader application of scattering techniques.

The great potential of these techniques with stable isotopes, however, has not been yet fully exploited, mainly because of the complexity and costs associated with the use and maintenance of the large facilities required. Therefore, simpler techniques with radiotracers are often preferred, although they imply an exposure to ionizing radiation which is absent or negligible for stable or long-lived radioactive tracers.

10.5 Selected applications

The two examples shown below give an idea of the different fields which can be investigated using tracer techniques and of the extent of the implications of the results obtained.

10.5.1 Molecular and cellular biology

Radioactive isotopes such as $^3$H, $^{14}$C, $^{32}$P, $^{35}$S, $^{86}$Rb and $^{125}$I have played and continue to play a key role in understanding the metabolic aspects of cells or bacteria, yeasts, plants and animals (including humans), and in the elucidation of the fundamental properties of genetic material. The radioisotopically labelled metabolites trace the corresponding stable molecules, and autoradiographic or counting measurements provide the information of interest. So, for example, $^{32}$P-deoxy-adenosine triphosphate ($^{32}$P- dATP) is used in the phosphorylation of a protein in order to evaluate its kinasic activity. Similarly, $^{35}$S and $^{125}$I label proteins in order to evaluate specific expressions. $^{35}$S-, $^{33}$P- or $^{32}$P-dideoxynucleotides are used for the synthesis of families of DNA-labelled molecules (template DNA) in DNA sequence analysis, and as probes for the detection of specific genes (Fig. 10.1).

Recently stable isotopes in combination with mass spectrometric techniques have been used for such labelling, especially when the molecules have to be injected into patients for in vivo studies.

The incorporation of $^{86}$Rb into glial and neuronal cells is a method of tracing the potassium entering the cell via the Na+/K+ pump and therefore to monitor the Na+/K+ ATPase activity in these cells, which is an important information in the study of neurodegenerative diseases. The value of the use of tracers in genetics will continue to improve, particularly as gene therapy is now beginning to find its way into ordinary clinical application.

10.5.2 Elemental kinetics

The biokinetics of essential elements, trace elements, micronutrients or any other elements of interest in nutrition, physiology or toxicology can be studied by tracer method. As already pointed out, the use of stable tracers is the ethically justifiable choice when dealing with healthy volunteers, although this could mean less information and/or more cumbersome measurements than with radiotracers. Therefore a sound experimental design is required. The double (or multiple) tracer technique, for example, consists in the simultaneous administration of two (or more) tracers through different pathways (typically, one oral and one intravenous tracer), and thus permits us to obtain dynamic pictures of relevant processes such as intestinal absorption or the main excretion pathways (Fig. 10.2).
Figure 10.1: Radioactive in situ hybridization of normal and tumour colon tissue sections. RNA probes containing $^{35}$S UTP were used to detect the presence of the transcript of a specific gene in the two samples. The incorporated radioactivity is detected by autoradiography using silver salt grains to impregnate a photo film. Panels (a) and (c) show the morphology of normal and cancer tissue, respectively, viewed in bright field under the microscope. In the dark-field panels (b) and (d), where luminescence of grains is the only source of light, the distribution of the specific RNA in the tissues can now be observed. In the case reported the gene is heavily expressed in the cancer cells, as shown by the intensity of the (white) signal.

Figure 10.2: Simplified scheme of the distribution of an element in the organism, as can be studied using the double stable tracer technique. One tracer is administered orally, the other is injected intravenously. Blood, urine and faeces samples are then collected and analysed, in order to characterize the processes of interest.
Many analytical approaches are used for the determination of the stable tracers in biological samples, and many are the works which can be found in literature about the study of biokinetics of nutrient and non-nutrient elements. For some elements such as iron, molybdenum, ruthenium and zirconium, the combination of stable isotopes, charged particle activation analysis and thermal ionization mass spectrometry has enabled us to obtain, for the first time ever, a detailed picture of the kinetics in blood plasma and of the renal elimination process. It has therefore been possible to revise the existing models, in order to provide a more realistic description of the biokinetics of ingested material. Radiation protection is, for example, a field which may greatly benefit from this improved realism, since the revised models may enable a more correct interpretation of control measurements in persons suspected of contamination and provide a sound support for the implementation of effective protective actions in case of radiological emergencies.

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11. Radiocarbon Dating of the Iceman Ötzi with Accelerator Mass Spectrometry

W. Kutschera

11.1 Discovery of the Iceman

On 19 September 1991 an extraordinary archaeological discovery was made in a high-altitude mountain pass (Tisenjoch, 3210 m) of the Ötztal Alps near the Austro–Italian border. Two mountain hikers from Nuremberg, Erika and Helmut Simon, after having scaled the Finail Peak (3516 m) that day were on their way back to the Similaun mountain hut (3019 m) located at the lowest part of a mountain ridge connecting the Finail Peak with the Similaun (3607). This ridge forms the border between Austria (to the north) and Italy (to the south). As the hikers approached a shallow ice-filled depression along the ridge, they were startled by seeing the body of a man sticking half-way out from the ice. Unusual climatic conditions in the summer of 1991 (including dust from Sahara resulting in enhanced melting of snow) had partly freed the body from its icy grave. The Iceman was later nicknamed ‘Ötzi’, after the mountain range where he was found. Two days after the first discovery, Hans Kammerlander and Reinhold Messner, two famous mountain climbers from South Tyrol happened to arrive at the site, and Fig. 11.1 shows them inspecting the Iceman. Messner’s initial guess was that he might have died some 500 years ago. Two days later on 23 September 1991 the body was recovered from the ice by Rainer Henn from the Institute of Forensic Medicine at the University of Innsbruck, and was flown to his institute by helicopter. Next day, when Konrad Spindler from the Institute of Pre- and Protohistory of the University of Innsbruck saw the unusual pieces of equipment found together with the body (in particular an axe with a bronze-like blade), he estimated that it was a 4000 year old find. This immediately created great excitement for both scientists and the public, resulting in many ‘colourfu’ events in the ensuing weeks.

One of the more serious events was the determination of the exact location of the find as it was very close to the Austro–Italian border. According to the Treaty of Saint Germain from 1919, the border was supposed to run along the water divide between the river Inn to the north and the river Etsch (Adige) to the south. After an official remeasuring of the border line it was established that the Iceman had been found 92 m inside Italian territory. According to international regulations, the Iceman therefore belonged to Italy, even though the meltwater from the discovery site was now draining towards the Inn. When the border was fixed originally, the site was filled with ice and snow, and it was not possible to determine the exact location of the water divide. However, the Iceman remained more than six years at the University of Innsbruck, from where most of the scientific investigations were organized. In January 1998, Ötzi was brought to his final home at the newly established Archaeological Museum in Bolzano, South Tyrol, Italy, where he is on display to the public. There he is safely stored in a glass vitrine with controlled temperature (−6°C) and humidity (98%) under glacier-like conditions. An impressive collection of his clothing and equipment is also on display. Popular write-ups of the Iceman story are available in German [1] and in English [2]. Scientific investigations of the Iceman are published in a series of monographs, with the latest one concentrating on paleobotanical results [3].
11.2 \(^{14}\)C dating of the Iceman

The accelerator mass spectrometry (AMS) laboratories in Zurich and Oxford performed the first \(^{14}\)C measurements on milligram amounts of bone and tissue from the Iceman [4, 5]. In Fig. 11.2 the determination of the calibrated date from the measured radiocarbon age is depicted. Although this procedure looks complicated, it is well established among the international radiocarbon community. It is apparent that the calibrated date covers a much larger time range than the uncalibrated radiocarbon age, which is obtained directly from the results of the AMS measurements. This is due to the ‘wiggles’ in the calibration curve, which results in a 95.4\% (2\(\sigma\)) confidence range of 3370 to 3100 BC. (For more details on the calibration issue see Section 11.4 below). Nevertheless, the \(^{14}\)C dating result unambiguously established that the Iceman lived before the Bronze Age (2400–800 BC), at the end of the neolithic period. Besides the body of the Iceman itself, a lot of equipment and other material was recovered from the site, apparently belonging to the Iceman as evidenced from \(^{14}\)C dating at the AMS facilities of Uppsala, Gif-sur-Yvette and Vienna [6]. In addition, some 500 kg of sediment was collected from the discovery site, and botanical and other remains were extracted by the Institute of Botany of the University of Innsbruck for \(^{14}\)C dating at the Vienna Environmental Research Accelerator (VERA).

11.3 \(^{14}\)C dating of materials from the site

There exists strong evidence that early in the Holocene (which covers the 10,000 years since the end of the last ice age) there were periods considerably warmer than today. Such changes in temperature can be most sensitively traced at high altitudes, where the vegetation reaches
its limit of existence. For example, tree logs set free by the rapidly receding Pasterze Glacier of the Grossglockner, the highest mountain in the Austrian Alps, have been $^{14}$C dated to the period between 8000 and 6900 BC [7]. These finds indicate that trees must have grown during that time at locations still covered by glaciers today. Similarly, one might expect that the high-altitude pass where Ötzi was found enjoyed ice-free periods also at other times. Figure 11.3 summarizes the results of 64 $^{14}$C measurements, most of them performed at VERA [8].

The materials are grouped into different species. It is apparent that they are spread over a large time range from approximately 5000 to 2000 BC. The upper two groups in Fig. 11.3 fall within the time period determined from body

Figure 11.2: The determination of the age of the Iceman from $^{14}$C measurements at the AMS laboratories of Zürich [4] and Oxford [5]. The combined radiocarbon age from these measurements is 4550 ± 19 years BP (Before Present = 1950 AD). The error is the 68.2% ($\sigma$) confidence value. The uncalibrated age is translated into a calibrated age with the help of the computer program OxCal using the INTCAL98 tree-ring calibration curve [12]. (a) Calibration curve from 4000 to 2000 BC (Before Christ). The straight line at 45° indicates a 1:1 transformation of the radiocarbon age into an uncalibrated calendar date. The intersection of the radiocarbon age with this line and the tree-ring calibration curve shows that the calibrated date is approximately 650 years older. (b) The enlarged ‘wiggly’ section of the calibration curve leads to three different solutions for the calendar date spanning 250 years. The small rectangular brackets beneath the peaks indicate the distribution of the 68.2% ($\sigma$) confidence ranges into three sections of 3360–3300 BC (29.3%), 3210–3190 BC (19.8%), and 3160–3130 BC (19.1%). The large brackets indicate the 95.4% ($2\sigma$) confidence ranges of 3370–3320 BC (34.3%), and 3230–3100 BC (61.1%).

Figure 11.3: Comparison of the ages for various materials collected at the discovery site of the Iceman, determined from $^{14}$C measurements at the AMS facility VERA in Vienna [10]. These results indicate that the site of the Iceman was probably ice-free also at considerably earlier and later times, and used as a high-altitude pathway across the Alps. See text for a more detailed discussion of the results.
samples of the Iceman himself [4, 5], and are thus most likely part of his belongings. The group labelled 'grass' includes several different species. Those falling within the Ötzi period are species which cannot grow at these altitudes and must have been brought to the place, probably by Ötzi himself. The grasses labelled _poa alpina_ and _poa laza_ today grow up to altitudes of 3000 and 3100 m, respectively. Since they apparently grew also at 3210 m (where Ötzi was found), a warmer period than today may have existed during the relevant time period. Mosses are less sensitive indicators of temperature changes, since they grow at these altitudes in a variety of climatic conditions. The group labelled 'other plants' includes _salix herbacea_, which grows in shallow depressions filled with snow for most of the year, whereas _saussurea moschata_ can be found today up to 50–120 m below the site of the Ötzi find. The other two samples are needles from trees which do not grow at these altitudes, probably blown up by winds across the surface of the glacier. Dates from animal dung of caprine origin (ibex, goat, sheep, etc.) spread over a large time range. It is perhaps not surprising that such remains are found there, as these are typical mountain animals. The lack of samples from this group during the Ötzi period looks curious, but may be explained by the observation that animals actually prefer to lie down on snow to cool off in summer time, and do not like bare rock as was probably present during Ötzi's time. The 'wood' group comprises samples which must have been brought up by man. The two samples, ax-1 and bow, clearly belong to the Iceman. Among the older samples, the most significant find is a piece of charcoal, which indicates that a thousand years before the Iceman a human being may have visited the place, making fire right there or bringing the remains of a fire to the site. The youngest sample, green alder, falls into the Iron Age (Hallstatt period), and shows signs of being cut and worked on by man. It is the first sample of this period found in this particular region of the Alps. Finally, two samples of soil have been collected in a spot close to the Ötzi site by two Italian scientists, and the total organic content of this material was _14C_ dated [9]. The older sample came from a somewhat thicker layer of soil, indicating possibly a warmer climate than for the younger one.

The new _14C_ dates raise hopes that climate indicators are present at this unique site. Combined with information from other regions in the Alps about climatic changes during the Holocene, this may allow one to link the presence of the Iceman to some climatic condition which favoured his appearance at this high altitude. Although, at this time, we can only hint at such a connection, it may be another piece in the puzzle that solves the mystery of the Iceman's origin, and his death at this lonely site high up in the mountains.

### 11.4 The radiocarbon dating method

Carbon forms the basic building blocks of organic compounds and therefore is an essential part of all life on Earth. As a consequence, the human body with an average weight of 70 kg contains approximately 16 kg of carbon. Almost all of this carbon is formed by the two stable isotopes, _12C_ (98.9%) and _13C_ (1.1%). However, a minute fraction of the carbon consists of the long-lived radioisotope _14C_ (1.2 × 10⁻¹²%), originating from cosmic-ray interaction in the atmosphere (see below). Since the half-life of _14C_ is 5730 years, the total _14C_ activity of our body is 3700 becquerels (1 Bq = 1 decay per second). This follows from the basic law of radioactivity:

\[
\frac{d}{dt}^{14}C_t = -\lambda \times ^{14}C_t = -\frac{\ln 2}{t_{1/2}} \times ^{14}C_t.
\]

\[\text{(11.1)}\]

Here, _14C_t_ denotes the number of radiocarbon atoms present at time _t_, _λ_ is the decay constant, ln 2 the natural logarithm of 2 (ln 2 = 0.693), and _t_{1/2_} the half-life. In our example _14C_t_ = 9.6 × 10¹⁴.

When we die, the supply of fresh carbon from the environment stops, and the radioactivity of the body decreases exponentially with time:

\[
^{14}C_t = ^{14}C_o \times e^{-\lambda t}.
\]

\[\text{(11.2)}\]
By knowing the initial $^{14}$C content, $^{14}$C$_0$, and measuring $^{14}$C$_t$, we can determine the time $t$ from

\[
t = -\frac{1}{\lambda} \times \ln \left(\frac{^{14}\text{C}_t}{^{14}\text{C}_0}\right) = -\left(\frac{t_{1/2}}{\ln 2}\right) \times \ln \left(\frac{^{14}\text{C}_t}{^{14}\text{C}_0}\right).
\]

Equation (11.3) is the basis for age determination by the radiocarbon method developed by Willard Libby in the late 1940s [10, 11]. This earned him the 1960 Nobel Prize in Chemistry “for his method to use carbon-14 for age determination in archaeology, geology, geophysics, and other branches of science”.

Accelerator mass spectrometry (AMS) measurements of $^{14}$C$_t$ in bone and tissue of the Iceman Ötzi revealed that the original $^{14}$C content ($^{14}$C$_0$) had decreased to 53% [4, 5]. From Eq. (11.3) one then calculates that the Iceman lived 5200 years ago, i.e. at the end of the Stone Age. However, for a correct determination of the age we need to know the actual atmospheric $^{14}$C$_0$ value at the time when Ötzi lived. In contrast to Libby’s original assumption, the $^{14}$C content of the atmosphere was not constant in time, and thus cannot be inferred for the past by measuring present-day $^{14}$C. We now know that both the earth and the solar magnetic field change with time. This has a varying shielding effect on the cosmic rays impinging on the atmosphere, and thus on the $^{14}$C production rate. In addition, climatic effects can also change the atmospheric $^{14}$C content by variations in the exchange of $^{14}$C between the global reservoirs of $^{14}$C (see below). For the past 12,000 years, a $^{14}$C$_0$ calibration was obtained by measuring $^{14}$C$_t$ in tree rings whose absolute age (calendar year) was determined from dendrochronology (tree-ring dating) [12]. For earlier times, other objects such as corals, stalagmites, and lake sediments can be used [12, 13]. It is important to note that the uncalibrated ‘radiocarbon age’ must not be confused with the calibrated ‘calendar date’, since there can be considerable time differences between the two (see Fig. 11.2).

11.5 $^{14}$C production via cosmic rays

High-energy protons originating from the sun and from outside the solar system continuously bombard our atmosphere and produce secondary neutrons by smashing atomic nuclei of nitrogen, oxygen and argon, the main constituents of the air. The neutrons are slowed down by elastic collisions with other air nuclei, and are eventually captured by nitrogen producing $^{14}$C through the nuclear reaction $^{14}$N + n → $^{14}$C + p. The freshly produced $^{14}$C atoms are chemically very reactive and immediately form carbon monoxide through the reaction $^{14}$C + O$_2$ → $^{14}$CO + O. After an atmospheric residence time of 2–6 months, $^{14}$CO molecules react with the extremely aggressive OH radical to form carbon dioxide through the reaction $^{14}$CO + OH → $^{14}$CO$_2$ + H. After a mean atmospheric residence time of several years, where $^{14}$CO$_2$ is thoroughly mixed with the stable CO$_2$ content of the atmosphere, it exchanges with the biosphere (through photosynthesis), and with the hydrosphere (dissolution in oceans and other water systems). It is interesting to note that approximately one fifth of the total atmospheric CO$_2$ inventory is cycled through these reservoirs per year. As a consequence of these processes, a well equilibrated distribution of the global $^{14}$C inventory is reached, with ~ 93% of $^{14}$C residing in the ocean, ~ 5% in the biosphere, and ~ 2% in the atmosphere.

11.6 Measurement of $^{14}$C with AMS

The $^{14}$C content of a sample can be measured through decay counting (radioactivity, see Eq. (11.2)) or through atom counting ($^{14}$C/$^{12}$C isotope ratio). In the latter measurement one need not wait for the infrequent decays of $^{14}$C. Since the archaeologist in general wants to preserve as much material as possible from a precious find, it is important to use only a little material for the age determination. In this respect, accelerator mass spectrometry (AMS) has an enormous advantage over decay counting. From the example above, one can calculate that one milligram ($10^{-3}$ g) of carbon from our body still contains 60 million $^{14}$C atoms. However, because of the long half-life of $^{14}$C, the radio-
activity of this material is only $2.3 \times 10^{-4}$ Bq, or about one decay per hour. On the other hand, with AMS it is possible to measure about 2% of all $^{14}$C atoms in one hour, i.e. 1.2 million. One thus gains a factor of one million in the detection sensitivity of $^{14}$C! This is comparable with the gain in light collecting power when using a 5 m telescope (e.g. on Mount Palomar) rather than the naked eye which has an aperture of about 5 mm (the light collecting power is proportional to the square of the diameter). For $^{14}$C measurements this means that instead of using several grams of carbon in several days of beta counting, an AMS measurement can be performed with 1 mg of carbon in about one hour, achieving the same statistical accuracy.

AMS determines the isotopic composition of a sample material by first producing a negatively-charged ion beam, which is then subjected to a series of extremely selective filtering procedures in order to find $^{14}$C, ‘the needle in the haystack’. $^{14}$C/$^{12}$C ratios in the range of $10^{-12}$ to $10^{-15}$ can be measured in this way. Details of the measuring procedures, which at essentially all AMS facilities involves a tandem accelerator, can be found in Refs. [14]–[16].

An important part of $^{14}$C dating is the sample preparation, i.e. the extraction of genuine carbon from the raw sample material. For AMS measurements there are four distinct steps: i) a precleaning procedure to remove all non-indigenous carbon, ii) the complete combustion of carbon to CO$_2$, iii) the reduction of CO$_2$ to elemental carbon with H$_2$ using Fe or Co as a catalyst (graphitization), and iv) the pressing of small pellets containing typically 1 mg of carbon for use in the Cs-beam sputter source to produce negative ions. At the AMS facility in Vienna, the Vienna Environmental Research Accelerator (VERA), forty carbon samples can be loaded into the ion source, usually thirty unknowns together with eight calibration samples of known $^{14}$C/$^{12}$C ratios, and two background samples.

With careful consideration of all steps in sample preparation and isotope ratio measurements, overall uncertainties around ±35 years are achieved at VERA for uncalibrated radiocarbon ages less than about 10,000 years BP. However this uncertainty can increase considerably through the ‘wiggliness’ of the tree-ring calibration curve (see Fig. 11.2). The $^{14}$C dating limit lies at about 50,000 years BP. This limit is not determined by the counting statistics, but by the finite background correction, which lies in the same time range. It means that the unavoidable contamination with modern carbon in the sample preparation procedures must be kept below one per mil (i.e. 1 µg out of 1 mg). There are many other factors which have to be taken into account (isotope fractionation, reservoir effects, the ‘old wood’ problem, etc.) in order to arrive at a reliable date. All things considered, it is wise to follow the Libby rule: ‘Radiocarbon dating is something like the discipline of surgery — cleanliness, care, seriousness, and practice’.

11.7 Conclusion

AMS measurements of $^{14}$C in small samples has grown into an extremely useful method in a variety of different fields [16]. Besides numerous applications in archaeology, such diverse fields as oceanography, ground-water dating, atmospheric science, forensic medicine, biomedical science, glaciology, sedimentology and meteoritics all benefit from the extreme sensitivity of the method and the smallness of the required sample size. Although other cosmogenic radionuclides are being measured with AMS, $^{14}$C is by far the most used one. More than 90% of all AMS measurements world-wide are devoted to $^{14}$C. The variation of the atmospheric $^{14}$C content with time is a serious problem limiting the achievable precision of $^{14}$C dating, as shown by the example of dating the Iceman. One is well advised not to push the precision beyond the limits set by these natural variations, even though under certain circumstances one can improve the precision of the age determination by performing ‘wiggle matching’ calibrations. This is possible if a series of samples is available, where a relative chronology of the samples can be deduced from other considerations (e.g. stratigraphy in an archaeological find). Notwithstanding this caveat, $^{14}$C is a true gift of nature to man, allowing us to look at our world in a way not possible by any other means.
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12. Invisible Threat: The Risk of Ionizing Radiation

M. Kellerer

12.1 Introduction

The destruction of Hiroshima and Nagasaki by atomic bombs, the nuclear arms race during the Cold War, and the Chernobyl reactor accident have created deep concern with regard to radioactivity and ionizing radiation. In the public perception, radiation has thus become one of the major risk factors, but this perception is often abstract and frequently not backed up by an informed judgement of the real nature and magnitude of the risk, in particular the risk of low-dose radiation exposures. The following synopsis is intended to outline basic facts and figures that put radiation and radiation risks into a more meaningful perspective and thus facilitate a realistic judgement of the merits and dangers of nuclear technology and of the various applications of radioactivity and ionizing radiation.

Health effects of ionizing radiation, specifically skin damage, had been observed early, when X-ray tubes were widely used without precaution. But such effects were explained as the results of gross negligence and continued high doses. For decades, radiation protection was not an issue. There was one notable exception: In New York a few days after Röntgen’s announcement Thomas Alva Edison pushed his mechanics to produce X-ray tubes, keeping them awake through day and night shifts by a hand organ set up in the workshop. Yet he was the one person to react and abandon at once all work with radiation when one of his assistants developed serious burns after exposure to the X-rays [1].

Most of the health effects that were observed in this first period were, indeed, the result of high exposures, but even before the nature of X-rays was recognized in the diffraction studies of Friedrich and Laue in 1912, a cluster of leukaemia was noted among Berlin radiologists, perhaps the first observation of the effect of low dose rate continued exposures [2]. But this did not cause particular alarm.

Larger tragedies were bound to happen. The first one resulted from the industrial use of radium-226 in the production of luminescent dials for watches and aircraft control panels. Hundreds of young women in the US, the UK, and other countries applied the radium-containing paint with fine brushes. Being paid by the piece, they tipped the brushes in the fastest way, i.e. with their lips. Thus they incorporated large bone-seeking α-ray and γ-ray activities. Many of these young women subsequently died from cancer, especially bone cancer [3]. The tragedy was noted, but was not taken as a warning and certainly not as a warning with regard to possible late effects of small doses. Indeed, the doses to the dial painters had been large.

12.2 The great reversal

When in 1895 W.C. Röntgen announced a mysterious new radiation that could penetrate all kinds of matter, the world took immediate and enthusiastic notice. In the spirit of the nineteenth century, the finding was taken to be the promise of unlimited technical progress and the apotheosis of classical physics. Both beliefs were universal and both, as it would turn out later, were wrong. In the same vein, and long before its nature was understood, radiation and subsequently radioactivity were considered not only a miracle of science, but also a medical panacea: a universally beneficial agent.

For half a century radioactivity continued to be seen as beneficial. Mineral water had to contain radioactivity, even toothpaste contained it. Radium-226 pillows were sold without prescription as a good-for-all agent to those who could afford them.
Other tragedies followed, mostly due to the use or misuse of radioactivity in medicine. One of the major misapplications was the production and use of thorotrast, a contrast medium to be injected into blood vessels for X-ray imaging. As the name indicated, thorotrast contained the α-emitter thorium-232. Being a superb contrast medium it doubtless saved a great number of lives, especially during the second World War. But the price was paid by the long-term survivors who experienced grave health effects. Some 50 years after the applications almost none of the surviving patients have escaped the late effects, the most serious threat being liver cancers with very high lethality [4].

The naïve belief in the healing power of radioactivity persisted in spite of the negative experiences. In Germany after the second World War, when no treatment against tuberculosis was available, patients — many of them children — were given injections of high activities of Ra-224 in the erroneous expectation that this could inactivate the tuberculosis bacilli. The same treatment was administered to patients who suffered from ankylosing spondylitis, a chronic inflammatory process of the skeleton. The Ra-224 did, indeed, reach the bone surfaces, but it damaged them rather than healing the illness. Many of the children suffered growth disturbances, and patients in the entire group incurred other damage, such as opacification of the lens of the eye [5]. The most severe consequence was more than 50 deaths from bone cancer among 900 patients. But other cancers were also increased, and even today the women who were treated as children show an increased rate of breast cancer.

While the treatment was totally inappropriate for bone tuberculosis, it would have made sense for ankylosing spondylitis had it not been for the very high doses that were used. A later form of the therapy with much smaller doses is still found to be effective for a symptomatic, anti-inflammatory treatment.

These and other experiences demonstrated health effects of high radiation doses, but they did not indicate, nor were taken as indication, that small doses of radiation or small dose rates could be detrimental. It was, instead, believed that there was a threshold of dose below which there could be no undue health effects. In fact, small doses of radiation were still seen to be generally beneficial and stimulating to health.

The great reversal happened half a century after the discovery of X-rays and of radioactivity when the atomic bombs destroyed Hiroshima and Nagasaki. About 200,000 people were killed by the heat flash and the blast of the bomb in the two cities, and thousands of those who survived the immediate effects suffered from severe radiation sickness. The horror of the nuclear explosions reversed the perception of radiation and radioactivity and turned the symbol of progress and life into the image of hell. Figure 12.1 is a point diagram of the A-bomb survivors in Hiroshima, colour-coded for dose.

![Figure 12.1: Point diagram of the A-bomb survivors in Hiroshima, colour-coded for dose.](image-url)
While research on radiation-induced delayed health effects was interdicted by the US military administration in the first years after the atomic bombings, Japanese physicians nevertheless noted an increased incidence of leukaemia among the inhabitants of Hiroshima and Nagasaki: a few cases per year, but the first tangible evidence of late radiation effects. The observation, still poorly quantified, changed the perception of the risks of ionizing radiation. It was then realized that radiation may not just cause hereditary damage through mutations of germ cells, but can equally cause leukaemias through mutation of somatic cells. The critical point was that — at least conceptually — the mutation of a single cell could lead to a leukaemia, which suggested that there is no threshold. Even very small doses could — with correspondingly low probability — cause a leukaemia.

Recognition of this changed the philosophy of radiation protection. Earlier it was assumed that detrimental effects could be completely avoided by limiting radiation exposures to a safe level. Now it was realized that all radiation exposures are likely to carry a certain risk and that consequently the aim and ambition of radiation protection could merely be to reduce exposures to a level As Low As Reasonably Achievable (the ALARA principle) [6].

While this is a prudent and by now widely accepted position, it has nevertheless created controversy and has invited misinterpretations that produced — in association with the experience of the atomic bombs and the ensuing nuclear arms race — fear and apprehension. The highly biased perception of radiation risks is perhaps most poignantly expressed in the true but greatly misunderstood statement that even a single charged particle might create a cancer. The force of this statement, but also its fallacy, lies in the fact that it evokes an image without quantifying its probability. A consideration of essential findings in decades of follow-up of the health status and the cancer mortality and incidence of the A-bomb survivors can provide the required numbers.

12.3 Cancer rates among the A-bomb survivors

When the increased frequency of leukaemia among the A-bomb survivors was noted, it confirmed the need for extensive health studies, and even then the question began to be asked whether there might be a similar increase of solid cancers. In 1950 a large study was therefore initiated that included 120,321 survivors from Hiroshima and Nagasaki, the Life-Span Study (LSS) cohort, whose causes of death were to be followed. In addition, tumour registries were established in Hiroshima and Nagasaki, so that the cancer mortality data are supplemented by incidence data. While the detailed results of the study have been described in a number of highly informative reports [7, 8], essential results will be outlined here.

For leukaemia the increased incidence was seen clearly from the beginning of the study. Figure 12.2 gives the number of cases per year in the LSS cohort. No molecular markers or clinical distinctions are known today that could identify a leukaemia or a solid cancer as being radiation induced rather than 'spontaneous'. Even conceptually there is little basis for this distinction: cancer is a complex multi-factorial process and radiation appears to be just one co-factor that tends to increase the incidence. Any excess incidence that is due to the irradiation must, therefore, be determined by statistical comparisons of the subgroups of survivors exposed to different doses. Sophisticated analyses of this type have been performed and the resulting attribution is depicted in Fig. 12.2 by the red area which represents the excess rate, i.e. the cases attributed to the radiation exposure, and by the grey area which represents the expected leukaemia cases, i.e. the incidence that would have occurred even in the unexposed population.

The largest excess leukaemia rate is seen in the first years of the study, subsequently the excess rate declined, and in the most recent observation periods it has largely disappeared. The rise in the expected annual numbers reflects the increase of the spontaneous leukaemia rate as the average age in the LSS cohort increases. Ultimately the annual numbers will, of course,
is only about 4% over the entire period of observation. This corresponds to about 315 out of a total of 7558 cancer deaths during the observation period. While the relative increase is much smaller than for leukaemia, the absolute number of 315 excess deaths is more than four times larger than the number of excess leukaemia cases (75). Since the excess rates persist for solid cancers, but not for leukaemia, the ratio will increase further.

Attribution of 'only' 75 leukaemia cases and 315 deaths from solid cancer to radiation among the A-bomb survivors is at odds with the common perception — a perception shared by the people of Hiroshima and Nagasaki — that most of the many thousands of cancer cases among the A-bomb survivors are due to radiation. While the result is an important qualification, it must not be taken as indication that the observations on the A-bomb survivors are uncertain. In the subgroup of 5489 A-bomb survivors who were exposed to doses in excess of

For quite a number of years after the beginning of the observations, no similar increase was seen in the solid cancer mortality of the A-bomb survivors. When finally ascertained it turned out to be percentage-wise much smaller than the increase in the leukaemia incidence. Figure 12.3 gives the results in analogy to Fig. 12.2 and shows that the attribution to the irradiation

Figure 12.2: Annual number of leukaemia cases (circles) among the A-bomb survivors in Hiroshima and Nagasaki in the different time periods. The grey area represents the annual numbers that would have occurred — according to the detailed statistical analysis — without the radiation exposure. The red area represents the excess incidence that has been caused, according to the computations, by the radiation exposure. The increase in spontaneous cases reflects the rise in incidence with age of the A-bomb survivors. The excess rate was largest in the initial years; no leukaemia registries existed before 1950, but it is known from other radioepidemiological studies that a certain excess rate may have been present already three to four years after the radiation exposure.

Figure 12.3: The annual number of deaths from solid cancers (circles) in the LSS cohort of the A-bomb survivors. The grey area represents the annual number of cases that would have occurred — according to the detailed statistical analysis — without the radiation exposure. The red band represents the excess that has been caused, according to the computations, by the radiation exposure. The increase of the spontaneous cases reflects the rise of the cancer rate with age of the A-bomb survivors. Only a small fraction of the observed cases is attributed to the A-bomb radiation. This is so because the average dose in the LSS cohort was only 0.14 Sv, and the relative increase of the frequency of solid tumours is much smaller than the relative increase of the leukaemia incidence after a radiation exposure.
Figure 12.4: The excess relative risk (ERR) per Sv derived for different types of tumour from the observations on the A-bomb survivors. ERR = 1 per Sv implies that the tumour incidence is doubled after an exposure to 1 Sv. In all but one tumour type, the tumour rate has been found to increase with dose. The estimates are given together with their 95% confidence bands. The total number of cancer cases from 1958 to 1987 appears next to each point.

0.5 Sv the association with radiation exposure is, in fact, firmly established for the majority of cancer types. Figure 12.4 gives the excess relative risk, i.e., the ratio of the excess incidence to the spontaneous incidence, at 1 Sv for various tumour sites.

Similarly, there are reliable conclusions on the age and time dependences of the excess tumour rates. The diagrams in Fig. 12.5 demonstrate some essentials. The upper panel refers to leukaemia. The solid curve depicts the spontaneous incidence and its steep increase with age. The broken curves represent the rates predicted — on the basis of the A-bomb data — for an exposure to 0.2 Sv at the age of five or thirty (see arrows). The excess is wavelike with only a few years latent period and with similar absolute excess for the two ages at exposure. But due to the much smaller spontaneous rates at young ages the excess is much more visible for young ages at exposure. This is why childhood leukaemia is the first indicator of late radiation effects in an exposed population, a point that will be taken up in subsequent considerations of the expected and observed health effects of the Chernobyl reactor accident.

The lower panel in Fig. 12.5 relates to all solid cancer mortality combined. Reference is made here to a higher dose, since the relative excess at specified dose is less for solid cancers than for leukaemia. The latent periods are longer than for leukaemia, and the excess rates persist — unlike those for leukaemia — into old age. The overall excess is somewhat larger for younger ages at exposure, and the relative risks are generally larger at younger ages. The numbers are, of course, subject to some uncertainty, but the essential features and the general magnitude of the excess are reliably represented in the diagram.
12.4 Dependence on dose

12.4.1 Observed dose relations

Most members of the LSS cohort have received doses that were substantially below 0.2 Sv. Their excess risk can not be determined with much precision, since the number of excess cancer or leukaemia cases is very small in comparison to the spontaneous cases. Low-dose risk estimates must therefore be derived by extrapolation, and this has become a matter of heated debate and of continued controversy.

The diagrams in Fig. 12.6 represent the excess relative risk (ERR) for solid cancer mortality and leukaemia mortality of the A-bomb survivors as a function of dose [9]. The values are ‘gliding averages’ computed from the observed data for intervals (±33%) around the specified doses. The shaded bands represent the standard error of the estimated ERR.

The dose dependence for all solid cancers follows a trend that seems to be linear. For leukaemia it appears to be somewhat curved, the data suggesting little or no risk at low doses.

In both instances the statistical uncertainty is too large at low doses, say below 0.2 Sv, to permit meaningful direct estimates. Any such estimates must therefore be based on an extrapolation from the observations at high doses.

12.4.2 ICRP nominal risk coefficient

A conservative approach would base the risk estimates on a simple linear correlation with dose. The International Commission for Radiological Protection (ICRP) has taken a somewhat different point of view [6]. Arguing that the leukaemia data indicate reduced effectiveness at low doses and that animal experiments suggest likewise a reduction for solid cancers it recommended risk estimates at low doses that are only half as large as those obtained on the basis of overall linearity in dose. On this basis a ‘nominal
risk coefficient’ of 0.05/Sv has been derived for lifetime attributable cancer mortality. This is meant to imply that, for example, the exposure of 2000 people (distributed over age and gender) to 0.1 Sv would be expected to cause 10 excess cancer deaths, in addition to the 400 to 500 cancer deaths that would normally occur (in a developed country) in such a group. It is readily seen that the order of magnitude of this risk estimate agrees well with the data given in Fig. 12.6 for the solid cancer mortality which makes up the major part of total cancer mortality among the A-bomb survivors.

The ICRP assumption of a reduction factor for the derivation of the low-dose risk estimates has been disputed. But other assumptions — for example the postulate that the excess relative risk for solid cancers remains constant throughout life — have been conservative. It is also likely that part of the observed effects that are currently ascribed to the γ-radiation have, in fact, been due to neutrons [10]. When all aspects are taken into consideration, the nominal risk coefficient 0.05/Sv appears realistic, regardless of the controversial reduction factor.

While the study of the A-bomb survivors has become the major source of information on the risk of low radiation doses, studies of groups of patients exposed for medical reasons have largely confirmed the results, although they are usually less informative and tend, on average, to suggest somewhat lower risk numbers. Among the low-dose studies, the combined follow-up of several large groups of nuclear workers in Western countries has attracted particular attention. No dose-related statistical excess has been seen for solid cancer mortality in this analysis, but the data are not statistically inconsistent with the current risk estimates, and this is true also for leukaemia.

There has been considerable controversy about the Linear No Threshold (LNT) postulate that underlies the risk estimates, and there is, in fact, no definitive proof for this assumption. Epidemiology can not resolve the issue, since the few postulated excess cancers at low doses exhibit no ‘molecular markers’ that would make them recognizable within the statistical noise of the spontaneous cancer incidence. Certain mechanistic considerations have been invoked to support the idea of linearity in dose. They made use of the traditional ideas of target theory [11] by linking the assumption of linearity at low doses to the interpretation that even individual ionizing particles cause DNA damage, that some DNA damage will be misrepaired, and that certain rare mutations could thus be caused that enable the affected stem cell to initiate — with small but finite probability — a tumour. At very low doses where only few cells are affected by a particle, their number would be proportional to dose, as would the excess cancers.

However, there are radiobiological observations [12]–[14] of complexities — such as ‘adaptive response’, ‘genomic instability’, or the ‘by-stander effect’ — that could modify the cellular or tissue response at low doses. While such observations cast doubt on the, perhaps, too simplistic arguments that have been invoked in favour of linearity in dose, they are still inadequately understood, and it is unclear whether they might be relevant to late radiation effects and, if so, whether they would tend to decrease or enhance the response.

Since there is no direct evidence for the low dose effects, the risk estimates and the ICRP nominal risk coefficient need to be seen as a pragmatic guideline [15]. They are part of a prudent approach to radiation protection that accounts for putative — although statistically unrecognizable — risks by keeping them sufficiently low to be acceptable in comparison to other tolerated risk factors.

12.5 Radiation risks in perspective

Radiation protection is concerned with the small doses of up to 20 mSv per year that may occur in occupational settings, and it is predominantly in this context that the ICRP nominal risk coefficient 0.05/Sv is used as guidance.

The annual limit of occupational exposure is currently 20 mSv. This annual dose is rarely reached, and in one European country (Germany) a lifetime limit for occupational exposure has been set at 0.4 Sv. The average exposure
among the more highly exposed group of nuclear workers or the average dose for members of aircrews that regularly fly on certain long-distance routes is close to 5 mSv per year; if continued over a working life of 40 years this would add up to 0.2 Sv. Space travel is a new condition where even higher doses can occur.

The most straightforward and the most common quantification of the risks of such relatively high doses or of the more common smaller exposures is given in terms of the number of expected excess cancer deaths. Thus, if 100 workers were actually exposed to a lifetime occupational dose of 0.2 Sv — under present conditions in Western countries a rather unlikely assumption —, the collective dose would be 20 personSv, and the ICRP nominal risk coefficient would then predict one excess cancer death in addition to the 20 to 25 cancer deaths normally expected among 100 persons. It is clear that this is a rather substantial risk — a putative 4% to 5% increase against normal cancer mortality — in a small group of persons. It is also clear that this magnitude of risk exceeds the job-related fatality rate in most professions. If smaller exposures, but larger groups of persons are involved, the putative numbers of excess deaths are more difficult to judge, and they can, in fact, be highly misleading if very large populations and small — or even trivial — doses are involved.

To keep the magnitude of radiation risk in perspective, the putative number of excess cancer cases or cancer deaths needs to be quoted in relation to the spontaneous number expected in the exposed population, i.e. it makes far more sense to give the relative increase of cancer frequencies than the absolute number of excess cases.

An added perspective for the magnitude of radiation-induced increases of the cancer rates can be provided by seeing them not only in relation to the overall spontaneous rates, but also to their dependence on age or sex. Figure 12.7 provides such information. It gives as solid lines the age-dependent solid cancer mortality rates for men and women in a European population. Superimposed on the spontaneous rates is the increase that results according to the data of the A-bomb survivors from an assumed maximum occupational exposure to 0.4 Sv during a working life from age 25 to 65. One notes the substantially larger cancer mortality rate for men, and realizes that the increase from the exposure could equally be expressed as an increase of the rates at specified ages or a shift towards increased age with regard to cancer. Either change is percentage-wise larger for women than for men, but the lower overall cancer rate in women accounts for the fact that the absolute excess risk from a specified dose is not greatly dependent on gender.

![Figure 12.7: Solid cancer mortality rates and their increase due to an assumed occupational exposure to annual doses of 10 mSv from age 25 to age 65. The lower solid lines represent the normal age-specific rates for members of a West European population (UK). The red band represents the increase that is caused, according to the current risk model of ICRP [6], by the occupational radiation exposure. Average occupational lifetime exposures are substantially less than 0.4 Sv.](image-url)
Within the European Community, the ‘dose limit’ for the general population is currently set at 1 mSv per year. Very little insight is gained by measuring such small exposures against the nominal risk coefficient 0.05/Sv that has been deduced from exposures hundreds or thousands of times higher. It is far more meaningful to relate them to the magnitude of the ubiquitous ‘natural’ radiation exposure from terrestrial γ-rays, cosmic rays, radioactivity of the human body, and from radon in houses. The overall contribution from this natural radiation exposure is 1 or 2 mSv per year. The regional fluctuations are substantial and the contribution from radon alone can, in radon-prone regions, be far larger than the average total.

The annual limit of 1 mSv for the general public has been based on the principle that any ‘controllable’ exposure of the population — apart from the medical exposures that are justified by individual benefit — must not add appreciably to the natural radiation background. A limit to the public of 1mSv per year ensures that the average added population exposure is considerably smaller than this value and, in fact, the average population exposure from nuclear facilities or from commercial releases of radioactivity amounts to much less than 0.1 mSv per year, which is clearly an insubstantial increase of the natural radiation exposure and lies well within the regional variations of the background level. This comparison to the natural background including its regional variations is a simpler and more reassuring justification of the 1 mSv annual limit to the public than any consideration in terms of numerical risk estimates.

12.6 Other late radiation effects

12.6.1 Hereditary effects

Radiation-induced heritable mutations have been studied extensively in mice, and experiments had earlier been conducted on plants and on drosophila. There can, thus, be no doubt that ionizing radiation produces mutations also in human germ cells. Yet, hereditary damage due to radiation has never been demonstrated in man, and this includes the extensive studies on the children of the A-bomb survivors. Even advanced methods of molecular biology, including sophisticated protein analyses and determinations of the mutation rates of mini-satellites, have failed to show in the children of the A-bomb survivors any association with the parental radiation exposures [16].

The absence of demonstrable hereditary radiation effects is, of course, no evidence against such effects in man. It merely means that any radiation-induced increase of hereditary damage is difficult to detect in the presence of other factors that have much larger impact, such as the increasing age of the parents — in modern society — at the conception of their children.

The studies of the children of the A-bomb survivors can, on the other hand, be taken as proof that the doubling dose for hereditary damage is not much smaller than the dose of about 2 Sv which doubles the total cancer rate in man. Even in the absence of precise numbers it is, therefore, a reliable conclusion that substantial doses would be required to increase the rate of heritable diseases in an exposed population by a few percent. While a detailed discussion would be required of the long-term impact of radiation-induced genetic damage in an exposed population, it is sufficient in the present context to note that quantitative analyses have identified increased cancer rates — rather than hereditary effects — as the dominant detriment from low-level radiation exposure.

12.6.2 Prenatal effects

Prenatal radiation effects are sometimes confused with hereditary damage. In fact, they are quite distinct in not being due to a mutation in a single cell, being instead the result of extensive cell killing in the developing embryo or fetus. Prenatal malformations have resulted from
improper medical application of X-rays to pregnant women in the early days of radiology, and they have also been observed among the children exposed in utero to the A-bomb radiation.

From animal studies it is known that the embryo is sensitive to ionizing radiation and that it dies after comparatively low doses. But it is unlikely to transmit damage if it survives, because its cells tend to be pluripotent, so that they can replace any minor fraction of cells that is inactivated. The major risk of prenatal damage from radiation exposure occurs in the initial foetal period, and particularly in a critical phase between eight and thirteen weeks after conception when highly sensitive processes in the development of the central nervous system take place. Among about 1600 children prenatally exposed to the A-bomb radiation, more than 20 were born with severe mental retardation which was due to radiation exposure in the critical period. The alarming conclusion is that in this period a dose of only 0.5 Sv causes severe mental retardation in two out of ten children. On the other hand, there is still reason to assume a dose threshold for such damage, because it is due to substantial cell killing which requires a certain dose level.

In the same context it needs to be noted that prenatal radiography, a common procedure in the first half of the last century, has been associated with increased leukaemia and solid cancer rates in childhood [17, 18]. X-ray diagnostics of pregnant or potentially pregnant women is now strictly avoided.

12.6.3 Higher non-cancer mortality rates

For a long time it was assumed that the non-cancer mortality rates of the A-bomb survivors were entirely normal. In recent years this has been shown to be fallacious. There is now, in the group of the more highly exposed A-bomb survivors, a recognizable trend of increased general mortality rates. While these observations are not, as yet, of particular concern for radiation protection because the increases are seen only at substantial doses, the issue remains incompletely understood and will require continued study.

12.7 Expected and observed health effects from Chernobyl

The reactor accident in Chernobyl is recognized as the largest and most costly technical catastrophe ever — both in human and in economic terms. It is less clearly perceived that the technical disaster has been amplified by a second disaster of comparable dimension: the continued failure to establish credible communication between administrations, scientific experts and a public that is deeply concerned but fails to find the required guidance in a flood of contradictory information.

If the lasting confusion were confined to the countries of the former Soviet Union, it might be ascribed to years of secrecy and distorted information in a paralysed political system. In actuality the confusion has been equally deep and equally persistent in the much less affected societies of Western Europe.

When complex technologies and their inherent risks become unintelligible to the majority of the people, they are bound to lose viability. Nuclear technology and the perception of its potential risk are perhaps the most visible example. The fear of radiation is merely one aspect of the general concern, but it has become a focus. The presentation of scientific facts — even if they are well established — will not resolve a problem that has deeper roots than lack of technical information. But it is still a necessary component of any attempt to arrive at a realistic judgement.

12.7.1 Evacuees and liquidators

Some aspects of the Chernobyl accident are fairly well understood and are subject to little controversy. Within days or a few weeks of the accident, 28 reactor employees and firemen died from acute radiation sickness. Others survived but continue to suffer lasting health damage. The accident caused the evacuation of about 120,000 people from the near zone around the reactor in 1986, and the later relocation of more than 200,000 people. Large territories of what
is now Belarus, the Russian Federation, and the Ukraine were contaminated, and some level of contamination occurred in all countries of the northern hemisphere. About 240,000 emergency workers (‘liquidators’) were sent to the reactors or the evacuated 30 km zone in 1986 and 1987. Many of them — especially among those who worked on the reactors — must have been highly exposed, close to or even in excess of the 0.3 Sv limit that was officially adopted but was unlikely to be reliably enforced.

A systematic follow-up of the liquidators and of the persons who had been evacuated from the 30 km zone with delays of up to a few days would be desirable, but there is little hope for more than limited studies because the people in question have been dispersed and their dose records are poor. According to the report of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [19] none of the existing epidemiological reports can, as yet, be seen as firm evidence for radiation-related cancer or leukaemia increases or the increase of other radiation related morbidity or mortality rates among the liquidators or in the evacuated populations.

12.7.2 Thyroid cancers

The one dramatic radiation effect among the population in Belarus, the Ukraine, and Russia is the large excess of thyroid tumours. When very large activities of the short-lived radio-iodine were released during the accident, no measures were taken to reduce the consumption of fresh milk and vegetables, and no stable iodine was provided to the population to suppress the accumulation of radio-iodine in the thyroid. Very high thyroid doses were thus caused in children in the affected regions. The exposures occurred during the first days and weeks of the accident; thereafter when the radio-iodine had decayed, it was too late for remedial action. In the intervening years about 1800 childhood thyroid cancers — an extremely rare disease under normal conditions — occurred [19]. In adults an excess is less readily quantified, but it is likely to be present. The excess, especially in those exposed at young ages, will continue, with thou-

sands of further cases expected. Although thyroid cancer is rarely lethal, it has major impact on the victim who requires permanent thyroid hormone substitution after successful therapy. There will also be a severe and lasting impact on the health services in the three affected republics: since the prognosis for the thyroid cancer patients depends critically on early diagnosis, extensive and costly mass screening for thyroid tumours will be required for many decades.

12.7.3 Continued radioactive contamination and its impact

The dramatic increase of thyroid cancers has been due to the high thyroid doses that were caused by the short-lived radio-iodine in the first phase after the reactor accident. The lasting concerns of the people in contaminated regions, however, and the discussions on the need for restrictions and for remedial actions are directed at a different issue, namely the continued contamination and the resulting elevated radiation levels. The exposure is predominantly due to caesium-137. It results from external exposure and, to a somewhat smaller extent, from the uptake of contaminated food. The exposures are much lower than the doses to the thyroid from the initial uptake of radio-iodine. However the elevated radiation persists and exposes all organs of the body. Distrustful of official information, the population perceives it as a deadly threat widely taken to be responsible for assumed increases not only of cancer rates, but also of a multitude of other illnesses. Large numbers of persons in the affected regions believe that they are bound to perish from the surrounding danger.

In such a situation, the assessment of radiation risks is not an academic exercise. One needs to ask for: the doses the population receives and one needs to relate these doses to earlier experience, in particular to the findings from the follow-up of the A-bomb survivors. In addition to this assessment — and regardless of its conclusions — one needs to observe the actual health statistics of the population.

UNSCEAR [19] reports population doses — from external and internal exposure to the long-
lived activity — that have been accumulated in the first ten years after the Chernobyl accident in the contaminated regions. These total doses are given in Table 12.1 — together with the number of inhabitants — for Belarus, the Russian Federation and the Ukraine, as well as some of the subregions.

Table 12.1: Mean cumulated doses (excluding thyroid dose) from the Chernobyl accident in the contaminated areas during the period 1986–1995. Contaminated areas are taken to be the regions with initial radio-caesium concentration in excess of 37 kBq m\(^{-2}\).

<table>
<thead>
<tr>
<th>Region</th>
<th>Population (thousands)</th>
<th>Mean dose (mSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Belarus</td>
<td>1,881</td>
<td>8</td>
</tr>
<tr>
<td>Brest</td>
<td>167</td>
<td>6</td>
</tr>
<tr>
<td>Gomel</td>
<td>1,465</td>
<td>7</td>
</tr>
<tr>
<td>Gomel (≥ 555 kBq m(^{-2}))</td>
<td>78</td>
<td>40</td>
</tr>
<tr>
<td>Grodno</td>
<td>28</td>
<td>5</td>
</tr>
<tr>
<td>Minsk</td>
<td>25</td>
<td>6</td>
</tr>
<tr>
<td>Mogilev</td>
<td>195</td>
<td>18</td>
</tr>
<tr>
<td>Mogilev (≥ 555 kBq m(^{-2}))</td>
<td>20</td>
<td>72</td>
</tr>
<tr>
<td>Russia</td>
<td>1,983</td>
<td>7</td>
</tr>
<tr>
<td>Bryansk</td>
<td>451</td>
<td>17</td>
</tr>
<tr>
<td>Bryansk (≥ 555 kBq m(^{-2}))</td>
<td>95</td>
<td>30</td>
</tr>
<tr>
<td>Tula</td>
<td>724</td>
<td>4</td>
</tr>
<tr>
<td>Ukraine</td>
<td>1,296</td>
<td>11</td>
</tr>
<tr>
<td>Zhytomyr</td>
<td>313</td>
<td>14</td>
</tr>
</tbody>
</table>

The major conclusion from Table 12.1 is that the average doses from Chernobyl in the contaminated regions — while being highly undesirable from a protection point of view — are nevertheless smaller than or comparable to the natural radiation exposure during this period, which amounts to about 15 mSv. These doses are therefore not very likely — even for leukaemia as the most conspicuous indicator of whole-body exposures — to cause an observable excess.

Figure 12.6 shows that in Hiroshima there has been no leukaemia excess up to doses of 200 mSv. Even if one were to assume that this was a statistical fluke and that the underlying dose dependence is, in fact, linear with a doubling dose of 200 mSv, a mean dose of 10 mSv would cause only a 5% increase over the normal leukaemia rate for the lifetime of the exposed population, which would be difficult to detect. The excess could be substantially larger in the initial years after the exposure (see Fig. 12.5), but the exposure was spread out over time after the accident and any peak of the excess rate is, therefore, unlikely to be very pronounced.

While it seems, thus, unlikely that an excess of leukaemia will be seen in the overall rates, an increase might most readily be visible among children. Accordingly, special efforts have been made to monitor childhood leukaemia rates, and Fig. 12.8 gives the numbers for Belarus [20].

There is no indication of an increase either in the more highly contaminated regions Mogilev and Gomel in comparison with the less contaminated regions, nor in the period after the reactor accident in comparison with the period before.

If it is accepted that no increases of childhood leukaemia have been seen even in the more highly contaminated parts of Belarus, it is concluded that the lasting radioactive contamination is unlikely to have caused recognizable increases in the rate of other cancers, and it is equally unlikely that it should have been responsible for generally increased morbidity rates from illnesses never before associated with low — or even high — radiation doses.

The contamination from Chernobyl has been much lower in the countries of Western Europe, with cumulated doses due to Chernobyl below 1 mSv in all but a few smaller regions. However, there has been almost as much alarm and apprehension — at least initially — in some Western European countries as in the directly affected regions of the former Soviet Union. The alarm was justified where it was focused on the fact that nuclear accidents can be an immense and far-ranging threat, which demands greatly improved supranational safety conventions. A similar and equally essential alarm had been raised by the increasing number and magnitude of the atmospheric nuclear-weapons tests in the 1950s. In both cases the alarm was also justified by the level of the radioactive releases that had already taken place. The nuclear-weapons tests were in this respect even worse than the Chernobyl accident, since they caused an additional dose of about 1 mSv not just in limited regions but throughout the northern hemisphere. However, the individual health threat due to the comparatively small radiation doses was largely overrated, especially after the Chernobyl acci-
Figure 12.8: Annual childhood leukaemia cases (age < 15 years) for the seven major regions of Belarus. The bars on the points indicate the standard fluctuations that would occur according to Poisson statistics even if the rates were constant. The grey areas indicate the total number of children in the population (right ordinate). The time of the reactor accident is indicated by the vertical red line. No increase of the childhood leukaemia rates after the reactor accident is seen in these data.

dent and especially in some Western countries. The present synopsis of expected and observed health effects from the Chernobyl accident may help to correct some of the misperceptions, but it is equally necessary to understand how such misinformation can arise.

12.7.4 125,000 radiation deaths in the Ukraine?

A multitude of reports have kept alive the perception of increased cancer rates, increased rates of congenital malformations, and increased general morbidity and mortality as a result of the elevated radiation levels in Belarus, the Ukraine, and the Russian Federation. The mere number of such reports, if seen with no possibility to assess their reliability or their degree of documentation, tends to discredit the statements by expert bodies and scientific committees. Even the UNSCEAR may encounter disbelief, when it acknowledges the increase of thyroid tumours due to the initial radio-iodine exposures, but continues to say that otherwise “there is no evidence of a major public health impact attributable to radiation exposure fourteen years after the accident. There is no scientific evidence of increases in overall cancer incidence or mortality or in
non-malignant disorders that could be related to radiation exposure" [19].

To disentangle facts and fallacies will remain a difficult task. But one example — a report that received considerable attention — may elucidate the mechanisms of negligent, if unintentional, misinformation.

A few years ago, on the ninth anniversary of the Chernobyl accident, the Minister of Health of the Ukraine was cited in almost all major international public media with the horrific news that, up to this point, 125,000 people had died, in addition to 6000 liquidators, from radiation effects. These numbers were taken to be the final confirmation that the worst expectations had been exceeded. In reality a statement of the ministry had been misunderstood and had been spread without further examination. The original statement read: “The total number of deaths among the population in the most contaminated regions was more than 125,000 in the years 1988 to 1994”. It was then added that most of these deaths occurred among old people.

The report of the Ukrainian ministry was widely taken to refer to mortality caused by radiation — not a far-fetched assumption for a deeply concerned public. However, the ministry had in fact referred to all deaths in the contaminated regions. Enquiries at the Ministry of Health confirmed this all too evident fact. While no population number was quoted, it was known that the administration generally referred to about 2.2 million people in the most contaminated regions. In Western European countries the mortality rate is somewhat in excess of 1% per year. Assuming the smaller mortality rate of 0.9% per year for the probably somewhat younger population in the Ukraine, one would expect roughly $7 \times 20,000 = 140,000$ deaths in the period 1988 to 1994. The number that was seen as apocalyptic horror in the public media was, in fact, perfectly normal.

12.8 Conclusion

Half a century of naïve confidence in the unlimited power of technical progress had been accompanied by unfounded beliefs in the positive health effects of X-rays and of radioactivity. The atomic bombs on Hiroshima and Nagasaki and the subsequent nuclear arms race have reversed this perception and during another half-century the attitude against radiation and radioactivity has become increasingly critical. Now, fear and apprehension are directed against the peaceful uses of nuclear technology, and — focused on radiation risk — they relate even to basic research and to the medical applications of radiation that help to save or restore countless lives.

It is a tragic aspect of radiation science that the most detailed insights on the late effects of ionizing radiation derive — as has here been outlined — from the use of the atomic bombs. The heritage of the nuclear arms race includes an equally dark source of knowledge which has only recently been uncovered. Along the river Techa in the southern Urals, high radiation exposures occurred a few years after the destruction of Hiroshima and Nagasaki. The exposures resulted from the release into the river of vast amounts of fission products that were generated in the secret plutonium plants of Mayak. Very high radiation exposures of the population along the river resulted, and thousands of workers at Mayak received similar radiation doses [21]. These events were long kept hidden, but the exposures and their health effects are now the object of an international cooperation that is about to parallel and complement the studies on the atomic bomb survivors.

Much is known about the potential effects of low doses of radiation, but the issue will remain a major task for science and a challenge to those who have to convey scientific insights into general knowledge and into political decisions.
Bibliography


PART 3: ATOMIC AND CONDENSED MATTER PHYSICS

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1. Introduction

This part of the report presents selected activities that are related to atomic and condensed matter physics. In the broad field associated with these two domains we only focus on questions whose study requires the use of nuclear techniques and facilities. In some cases the research related to these questions now develops around specific and independent installations, but most of the results presented herein have been obtained in a nuclear physics environment. We have deliberately decided not to present the very broad and productive research achieved with the light sources associated with synchrotron radiation. These facilities are obviously among the most used in physics, chemistry and biology; however, they constitute a whole world that lives its own life independently of nuclear physics.

The work presented here often deals with very basic problems: Concepts, models and techniques in nuclear physics can be applied to the development of fundamental research in other areas. The reverse is also obviously true: the borders between disciplines are porous and cross-fertilization greatly contributes to the development of physics. This is certainly the case in the interplay between nuclear, high-energy and atomic physics that is analysed in Chapters 2 and 3. There, it is shown for instance how spectacular technical breakthroughs have led to orders of magnitude improvements in precision when testing quantum electrodynamics in very strong fields. Much effort goes into the search for physics beyond the standard model: let us mention, for instance, studies of weak interactions through the determination of the full kinematics of nuclear beta-decay in atomic traps. Such traps are also used in mass spectroscopy measurements providing high precision mass determination of stable and radioactive nuclides. This shows how instruments and experimental techniques first developed for atomic physics purposes provide important information on nuclear properties. In this domain, measurements concern determination of half-lives, spins, magnetic and electric moments and charge radii of nuclides. One can also benefit from a solid state physics technique (ion blocking) to study long lifetime components of excited nuclei or of exotic heavy nuclei, in relation to nuclear dissipation.

The following chapters are transitively linked. Chapter 4 is also devoted to atomic physics and specifically describes the impressive progress made in the study of atomic collisions. There is first a definite improvement in precision that now allows tracking of all the collision products and thus provides access to the details of collision dynamics. But there is also an extension of the observations and modelling in more and more complex systems: modification of the target electron density around an ion moving in solids and associated oscillation of its excited state population, cluster ion stability and dissociation, interaction of very slow highly charged ions with surfaces, etc.

Chapter 5 deals with the modification of materials induced by irradiation and associated applications. The connection with the preceding section is clear: the primary events that lead to the observed modifications are the atomic collisions. However, the response of the medium to these primary events involves a higher level of complexity; on this question important progress has been made. This is of prime importance insofar as biological matter and life sciences (particularly therapy) are concerned, and the work presented in Chapter 5 is thus of particular interest for many features of Part 2. The connection with Part 1, devoted to Energy, is also obvious: one has to predict the evolution under irradiation of nuclear fuel and of matrices for nuclear waste transmutation.
Chapter 6 presents major developments recently achieved in the most important techniques of material analysis related to nuclear physics. These developments are connected to the improvement of sources, beams and detectors. They result in spectacular gains in sensitivity as well as in spatial and time resolution for the analysis of the composition, structure, dynamic evolution and physical properties (e.g. magnetism) of all kinds of materials. Among these techniques, those based on the use of ion beams require a precise knowledge of the ion-solid interactions and associated effects that are presented in Chapters 4 and 5.

The technical developments that provide the basis for the work presented in Chapters 2–6, as well as the technical perspectives for the near future, are detailed in Chapter 7. It is apparent that many improvements around nuclear physics facilities are beneficial for various other fields. In the future, it would therefore certainly be important to include the possible use of such facilities as ‘multidisciplinary tools’ in the planning stage, requesting input from members of diverse scientific communities. Of course, these tools should then also be more widely accessible.
2. Fundamental Interactions

2.1 Introduction

The investigation of nuclear properties has played an important role in further developing the understanding of fundamental interactions. This statement is perhaps best illustrated by the original Lamb shift experiment where radio-frequency techniques originally developed for measuring nuclear ground-state properties have been used to determine a small deviation from the Dirac values, indicating the need to reformulate the interaction of light with matter. One could state equally well that the invention and use of accelerators in the field of nuclear physics laid the foundation for the present understanding of nuclear and high-energy physics. Being subject to all kinds of interactions beyond strong interaction, nuclei or nucleons continue to contribute to a deeper understanding of the laws of nature. This extends from strong interaction proper to increasingly precise tests of the structure of electroweak interactions.

Even with the Standard Model (SM) now widely accepted, high-precision studies are still necessary to refine theoretical tools for detailed calculations of higher-order phenomena within the SM. Expressed differently it is not sufficient to know the structure of an interaction in principle, but the theoretical tools should be confronted with experiments at the highest precision. As an illustrative example, the high-precision measurements of QED in bound states of high-Z hydrogen-like atoms (Lamb shift and g-factor measurements) are mentioned here. A second example is the investigation of strong interaction phenomena in elementary hadronic atoms like pionic or pionic as well as kaonic hydrogen. Both experimental programmes have in common that they are conducted with two-body systems, thus avoiding many-body effects which would render high-precision experiments impossible.

As it is also widely accepted that the SM will not be the final theory any effort should be undertaken to find hints for new physics. Again only simple systems with well-controlled properties (both experimentally and theoretically) can be used for a search of physics beyond the SM. Such systems are also available outside high-energy physics, as are bare nucleons or nuclei at rest. Good examples for modern experiments are the search for the electric dipole moment (EDM) of the neutron and correlation experiments with neutrons or trapped atoms. Also the use of nuclear physics techniques for the study of highly ionized atoms or exotic atoms opens many possibilities for high-precision experiments also beyond the SM. Examples are the search for right-handed currents in weak decays and the search for violation of conservation laws like CP or CPT.

2.2 Within the Standard Model

2.2.1 QED

Quantum electrodynamics (QED) is the basis and cornerstone of all present field theories. It is the best-confirmed theory in physics. The more it is important to investigate QED predictions under as many different circumstances as possible and especially at extreme conditions. Atomic systems permit a study of bound-state QED at higher order of the perturbation series expansion and additionally for high values of the electric field. Exotic atoms provide in addition a means to test few-body Coulomb calculations with extremely high accuracy.

Higher-order corrections

Despite the enormous success of QED in predicting the properties of electrons in weak fields, a precise test is still pending for the strong-field limit where new phenomena might show
up. Thus, a primary goal is to explore the behaviour of electrons in the strongest electromagnetic fields accessible to experimental investigation (see Fig. 2.1). Here precision measurements of electron binding energies are best suited to deduce characteristic QED phenomena. Therefore, the comparison of predicted and experimentally determined energy levels of strongly bound electrons provides a critical test of QED in strong fields.

Figure 2.1: Expectation value of the electric field strength for a K-shell electron as a function of the nuclear charge number [1].

Whereas low-Z ions are primarily sensitive to the lowest-order self-energy corrections, the study of the higher-order self-energy and vacuum polarization contributions requires the heaviest ions available. Here, the goal of the experiments is to probe higher-order QED contributions, which correspond to Feynman diagrams such as the two-photon exchange diagrams. For the case of uranium, where the total 1s Lamb shift contributes 465.8 eV [1] to the total ground-state binding energy of 131.814 keV, a stringent test of QED requires an absolute experimental accuracy of about ±1 eV, which represents the accuracy theoreticians claim at present. For such studies the storage and cooler ring ESR at GSI-Darmstadt provides favourable conditions where intense beams of high-Z ions up to bare uranium are available for experiments. This has been demonstrated within the first series of experiments performed at the ESR gas-jet target as well as at the electron cooler device. For the case of the 1s Lamb shift in hydrogen-like uranium, the achieved accuracy of ±13 eV [2] is already a substantial improvement by almost one order of magnitude compared to a former experiment conducted at the BEVALAC accelerator. The result obtained gives the most precise test of QED for a single-electron system in the strong-field region and is now at the threshold of a real test of higher-order QED contributions. A prerequisite for the currently reached precision is the capability of the ESR to decelerate high-Z ions in order to reduce systematic uncertainties associated with the Doppler effect. By using decelerated beams, further progress towards an absolute accuracy of 1 eV can be anticipated. The deceleration mode not only reduces the uncertainties in the Doppler corrections but, in particular, it provides a very efficient production of characteristic projectile radiation (Fig. 2.2).

Figure 2.2: X-ray spectrum measured at the ESR/GSI associated with Lyman transitions in hydrogen-like uranium [2].

Beside the precision measurements of binding energies (Lamb shift), the measurements of the magnetic moment anomaly (g-factor) of the bound electron in highly charged ions provide important tests of bound-state QED calculations. The experimental determination of
the g-factor of the free electron to a few parts in $10^{-12}$, $g_{\text{free}} = 2.002319304377(9)$, performed by Dehmelt and coworkers on a single electron in a Penning trap, is by far the most precise test of the theory of QED for free particles. However, information on the g-factor of the bound electron in hydrogen-like ions was only recently available only for the hydrogen atom and the $^4\text{He}^+$-ion. For the determination of the g-factor of the bound electron in highly charged ions as a test of bound-state QED, a precision Penning trap has been developed in a joint effort between the University of Mainz and GSI-Darmstadt. The g-factor of the bound electron is determined from the Larmor precession frequency of the bound electron and the cyclotron frequency of the hydrogen-like ion stored in the Penning trap. The recent measurements of the g-Factor Collaboration yielded the g-factor of the bound electron in hydrogen-like carbon $^{12}\text{C}^{5+}$ with an accuracy of a few ppb, $g_{\text{be}}^{\text{exp}}(\text{C}^{5+}) = 2.001041596(5)$, in excellent agreement with the theoretical value of $g_{\text{be}}^{\text{th}}(\text{C}^{5+}) = 2.001041591(7)$. This is one of the most stringent tests of bound-state QED. In the future, the g-Factor Collaboration will extend the g-factor measurements up to hydrogen-like uranium $^{91}\text{U}^{41+}$ (HITRAP Project) [3]. This will provide a crucial test of QED calculations in extreme electromagnetic fields.

In addition, the development and understanding of QED in strong fields can be improved significantly for many-body systems by measuring, for example, the $2s_{1/2} - 2p_{1/2}$ ($\approx 280$ eV) or the $2s_{1/2} - 2p_{3/2}$ ($\approx 4.5$ keV) splitting in Li-like uranium to an accuracy of $10^{-2}$ eV. Lithium-like ions are particularly suited since they represent a good compromise between the request for reliable calculations of the electronic factor and low atomic excitation energy. Consequently the most precise experiments were conducted for Li-like high-Z ions at the Super-EBIT and at an accelerator [4, 5]. For the goal of an accuracy of $10^{-2}$ eV, laser-based techniques appear to be most promising. At the ESR, photon energies in the laboratory frame of around 110 eV would be sufficient to induce the $280$ eV $2s_{1/2} - 2p_{1/2}$ transition in uranium when the laser light is counter-propagating with respect to the ions. Such a photon source could be provided by an X-ray laser pumped by multi-terawatt pulses from the 10 J front-end of the laser system PHELIX under construction at GSI [6]. The experimental procedure will be similar to the established scheme used for laser spectroscopy of the ground-state hyperfine splitting in hydrogen-like ions. For low- and medium-Z ions, high-precision spectroscopy experiments can also be anticipated for investigations at EBIT devices.

In contrast to electronic atoms where the self-energy graph contributes mostly to the QED correction of binding energies, exotic atoms probe a region at even higher electric field and are mostly sensitive to the vacuum polarization term. The accuracy of about 900 ppm reached today [7] is limited by the fact that the unknown occupation of the electron shell around the exotic atom leads to uncontrollable shifts of the measured energies. Modern experiments can be conducted with a fully ionized electron shell or under circumstances where the assumption on the status of the electron shell can be checked. High-resolution bent crystal Bragg spectroscopy offers many possibilities for a better determination of the vacuum polarization term both in muonic and pionic atoms. This opens the way to test the higher-order QED contributions to third order in the fine structure constant [8].

An important contribution to the test of QED predictions in electronic hydrogen to be mentioned later (see [34] and Section 2.3.4) is the proposed determination of the proton charge radius by a determination of the $2s-2p$ energy difference in muonic hydrogen with a Lamb shift type experiment. An increase in accuracy of a factor of 20 is envisaged for the first step of an experimental series including in future muonic deuterium and helium isotopes [9].

**Experiments with antiprotons: antiprotonic atoms and collision studies**

Present and future experiments with antiprotonic atoms focus on high-resolution laser spectroscopy of antiprotonic helium and hydrogen atoms. The applications in terms of fundamental questions range from the test of conservation
laws or fundamental principles to the calculation of the few-body Coulomb problem.

In the past, the theory of the pure Coulomb few body quantum problem was checked in atomic systems like the helium atom, and further developed in muonic molecular ions during the investigation of muon catalysed fusion. It is extended today to calculations of binding energies of ‘atomcules’ formed by a helium nucleus plus an antiproton and an electron. The corresponding experiments have been very successfully executed at CERN/LEAR and will be exploited further in the antiproton decelerator (AD) period in the ASACUSA experiment [10]. They are based on the discovery of metastable states with lifetimes of the order of \( \mu s \), which allows laser and HF excitation.

At present, a complete picture of the atomcule states is emerging with about 20 different transitions found for the two helium isotopes (three recently at the AD machine). In parallel to the highly developed experimental technique, the three-body calculations have reached a high level of sophistication, which includes relativistic effects and QED corrections like the Lamb shift [11]. Accuracy at the 0.1 ppm level is achieved at present with prospects to increase this still further into the region of several ppb, which requires higher-order QED calculations in a system different from the usual central potential systems, which is of interest in itself.

Future experiments require dedicated laser equipment and the ability to stop enough antiprotons at low pressures in order to minimize pressure broadening. In the ASACUSA experiment a radio-frequency quadrupole (RFQ) will be used to decelerate the antiprotons from the 5.3 MeV delivered by AD to energies in the region of about 100 keV and to collect them in an electromagnetic trap.

In addition the high-precision study of the atomcules delivers information on intrinsic properties of the antiproton like its charge, mass and magnetic moment, which in comparison with the equivalent numbers for protons leads to a test of CPT violation which is also planned for the antihydrogen measurements to be discussed below [12].

With well-developed laser and stopping techniques, it is also planned to perform spectroscopy of high-lying levels (\( n \) between 40 and 50) of the protonium atom. Narrow line widths can be achieved using a two-photon excitation with a subsequent ionization step in order to provide a signal. In case of success, a value for the antiprotonic Rydberg energy of unprecedented accuracy would be achieved.

Based on the RFQ deceleration technique collision experiments will be performed by extending the earlier experiments on ionization cross sections at LEAR [13]. Cooled antiprotons will be extracted again from the electromagnetic trap for e.g. experiments on the energy loss of antiprotons in hydrogen gas. With the stable antiprotons the present knowledge about the ionization processes of negatively charged particles and the formation of exotic hydrogen atoms can be developed much further. With a source of low-energy antiprotons at hand a new version of protonium and antiprotonic deuterium spectroscopy with a fast CCD detector can also be undertaken, resolving the puzzling situation left behind after the LEAR experiments [14].

2.2.2 QCD

Recently much theoretical effort went into the development of perturbation theories of strong interaction in the confinement regime based on the concept of chiral perturbation symmetry, which was also extended to include baryons in the Heavy Baryon Chiral Perturbation Theory (HBChPT). The study of the \( \pi^+\pi^- \) (pionium) system with the DIRAC experiment at CERN, provides a direct insight into the mechanism of chiral symmetry breaking. The high-precision measurement of the ground-state shift and width of piconic hydrogen tests HBChPT to fourth order. Experiments with kaonic hydrogen will extend this kind of experiment to strange quarks.

Pionium

A system consisting of two pions is conceptually the simplest system to study strong interaction effects. In an ongoing experiment at the CERN Proton Synchrotron it is planned to measure the
lifetime of the ground state of the pionium atom with an accuracy of 10% [15]. As the strong interaction width of the ground state is proportional to \( |a_0 - a_2|^2 \) (\( a_0 \) and \( a_2 \) are the isoscalar and isotensor scattering lengths), the measurement directly determines a combination of two isospin-separated scattering lengths with a 5% accuracy. Chiral perturbation theory predictions are at the same level of precision. Further developments are under study to also measure the QCD Lamb shift and extend the experiment to a study of the electromagnetically bound \( \pi K \) system which would allow the mechanism of chiral \( SU(3)_L \times SU(3)_R \) symmetry breaking to be studied.

**Pionic hydrogen**

The linear combinations of the isospin-separated scattering lengths \( a^+ \) and \( a^- \) of the pion–nucleon system can be determined from measurement of the strong interaction shift \( \propto a^+ + a^- \) and width \( \propto (a^-)^2 \) of the ground state of pionic hydrogen. This provides a stringent test of predictions from HBChPT. Moreover, the width \( (a^-) \) value alone allows the determination of the \( \pi \)NN coupling constant via the GMO sum rule. At present, an extraction of both scattering lengths with equivalent precision pionic hydrogen alone is hampered by a relatively bad determination of the width and relies on a separate measurement of the shift in pionic deuterium [16]. A determination from pionic hydrogen alone would not only provide a guideline for the further development of the HBChPT higher than fourth order together with an unprecedented accuracy of the \( \pi \)NN coupling constant, but would also allow the understanding of the pionic deuterium system to be tested [17]. Future experiments will benefit from a much improved peak/background ratio compared to earlier experiments, together with a substantially enhanced statistics.

**Kaonic hydrogen**

The investigation of the kaon–nucleon system aims at the understanding of \( SU(3) \) chiral symmetry breaking. Measurements of the ground-state level shift are planned for kaonic hydrogen and deuterium atoms. First measurements have been performed at KEK where the Lyman series of kaonic hydrogen has been discovered [18]. A dedicated set-up at the storage ring DAΦNE at Frascati will perform a 1% measurement of the isospin-separated scattering lengths in the near future [19]. Such a precision implies a determination of the \( K \)N sigma term, which is also a sensitive and direct measurement of the strangeness content of the proton.

### 2.3 Beyond the Standard Model

#### 2.3.1 Electric dipole moments of particles, nuclei and atoms

The discovery of a particle or nuclear electric dipole moment (EDM) implies a violation of both parity and CP reversal invariance. It would shed some light on the stage in the development of the universe when these symmetries were broken. Depending on time and energy of this event accelerator, experiments would possibly never reach the corresponding energies. High-precision experiments are in this respect complementary. Experiments on the EDM of the neutron have played an especially important role in non-accelerator particle physics and have reached a record in discarding theoretical models. The SM prediction of about \( 10^{-32} \) e cm for a neutron EDM is still very much below the present experimental upper limit (at 90% confidence level) of \( 6.3 \times 10^{-26} \) e cm as published in 1999 [20] and quoted in the compilation of the Particle Data Group. It may be argued at this stage that EDM experiments are therefore not able to answer fundamental questions. The contrary is in fact true, as they are extremely sensitive to possible new physics beyond the SM and because a positive result would shed light on the observed baryon–antibaryon asymmetry [21]. Recent developments in gauge theories with spontaneously broken symmetries have rendered different models for the description of CP violation [22, 23]. Supersymmetric (SUSY) models, two and multi-Higgs doublet models extend the SM and predict neutron EDM to be allowed to first order in weak interaction, which should be at the level of about \( 10^{-26} \) to \( 10^{-28} \) e cm.
Most experiments exploit the Ramsey separated field technique of magnetic resonance with a superimposed electric field. An experimental breakthrough was achieved by using ultracold neutrons (UCN). UCN are easily polarized and may be stored for a long time in a material trap (neutron bottle) which drastically increases the resolution of the experiment. Multi-cell arrangements are considered in order to suppress systematic errors as much as possible. Also the low velocity of the UCN is decisive in reducing systematic errors caused by motional fields. A very delicate part of the technical development is a proper design of magnetometers, which map the magnetic field distribution. New approaches to use solid deuterium to increase the flux density of UCN are being developed with promising results. The technical realization is being considered at Los Alamos and at various European institutions (Munich (Mainz) or PSI).

A completely different experimental method, using superfluid helium with $^3$He as polarizer and analyser with a $^3$He magnetometer, is being investigated at Los Alamos. The goal is to reach a sensitivity of $10^{-29}$ e cm [24].

Experiments (discussed below) concentrating on the high-precision measurement of neutron decay parameters in searching for effects beyond the SM also benefit from the development of high-intensity UCN sources.

Complementary to the experimental efforts with neutrons, a search for electron EDM has also been conducted in atomic physics experiments. Here a drastic enhancement is calculated for the measurement of electron EDM in heavy atoms [25, 26]. Not being subject to strong interaction, electron EDM experiments test theories different from neutron EDM studies. In Table 2.1, a value for the muon EDM is included which originates from the CERN g-2 set-up. At present, there are plans or proposals for a drastic increase in sensitivity for all three particles.

The best EDM limit for an object was obtained for the $^{199}$Hg atom, with an upper limit of $9 \times 10^{-28}$. Interpreted in terms of nuclear EDM this value delivers a result that is still about six orders of magnitude above the SM prediction, similar to the neutron EDM measurements.

<table>
<thead>
<tr>
<th>Particle</th>
<th>Experimental limit [e cm]</th>
<th>SM prediction [e cm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>n</td>
<td>$6.3 \times 10^{-26}$</td>
<td>$10^{-32}$</td>
</tr>
<tr>
<td>e</td>
<td>$4 \times 10^{-27}$</td>
<td>$10^{-40}$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>$1 \times 10^{-18}$</td>
<td>$10^{-38}$</td>
</tr>
</tbody>
</table>

The muon EDM is under consideration for an experiment at the g-2 storage at Brookhaven. Using a remarkable idea to compensate for the precession in the magnetic field by using a suitable radial electric field, a sensitivity of $10^{-24}$ e cm is expected [27]. Based on this idea there are approaches for the measurement of nuclear EDM at cooled storage rings. An EDM should show up in the build-up of a polarization perpendicular to the accelerator plane. Such a polarization could be measured by using $\beta$-decay of radioactive nuclei or by using polarization scattering after extracting the beam.

2.3.2 Weak interaction decays

Weak interaction decays permit a sensitive test of the validity of the SM from the measurement of small effects in different decay parameters. The relevant terms in the formula for the decay rate $W$ for a semileptonic transition from an oriented sample of nuclei or particles with vector polarization $J$, spin $\sigma$ and momentum $p$ of the detected lepton and $q$ being the momentum of the unobserved neutrino are given by

$$W \propto \left[ 1 + \frac{\Lambda(Jp)}{E} + B(Jq) \right] + \frac{D J(p \times q)}{E} E\nu + \frac{R J(p \times \sigma)}{E} + ...$$

where $E$ and $E\nu$ are the total energies of the emitted leptons, $A$ and $B$ are the usual decay asymmetry parameters arising from parity violation, and the $D$ and $R$ parameters are different from zero only if time reversal and/or parity violation is taken into account.

Neutron $\beta$-decay

The SM restricts the weak interaction Hamiltonian to two types of interaction, vector and axial
vector, which form left-handed hadron and lepton currents. Many experiments are carried out inside the framework of the SM in order to determine different parameters with ever-increasing accuracy, such as the lifetime of the free neutron or different correlation parameters from observables in the neutron decay.

A sensitive search for a deviation from the SM can be carried out by measuring for example the $B$-coefficient, providing a new limit for the mass of a right-handed boson MWR > 302 GeV/c$^2$ [28].

One illuminating example for the impact of neutron decay studies is the recently observed evidence for a deviation from unitarity of the first row of the CKM matrix, as obtained at ILL with the PERKEOII experiment [29].

An experiment to search for a T-odd, P-odd observable has been set up at a polarized neutron beam line at PSI [30]. It is sensitive to the $R$-parameter, and will result in vastly improved limits for scalar and tensor contributions to the V-A interaction.

**Nuclear $\beta$-decay**

In the last few years, various facilities in Europe have developed techniques to produce radioactive nuclides which otherwise only exist transiently during astrophysical processes like supernova explosions. Together with atom- and ion-trapping techniques developed in the atomic physics community, this has created new possibilities to study basic laws of nature and to detect possible insufficiencies in today’s theoretical models. A promising means to observe phenomena in disagreement with the SM is a study of processes governed by the weak interaction. As an example, we discuss the $\beta$-decay of a nucleus, when a neutron changes to proton or vice-versa. Measuring the details of the decay reveals the structure of the interaction.

The corresponding ‘ideal’ experiment is conceptually as simple as it is difficult in practice: by optical pumping, selected radioactive isotopes can be polarized while their thermal motion can be minimized. Observing the $\beta$-particle (electron or positron) and the recoiling nucleus in coincidence, the full kinematics of the event, including that of the unobserved neutrino, can be established in detail, as shown schematically in Fig. 2.3. Such ideal measurements give immediate access to observables that test the SM in detail. The correlation $\mathbf{J}(\mathbf{p} \times \mathbf{q})$ is e.g. an observable which is odd under time reversal and, therefore, any *measurable* deviation of its expectation value from zero would be inconsistent with the SM. The energy of the recoiling nucleus (< 100 eV) is extremely small for nuclear physics standards and requires the use of atomic physics detection techniques. This requirement, combined with the possibility to manipulate the spin direction, makes traps the ideal apparatus for this research. To solve the practical problems many technical and scientific innovations will be necessary. For an observation of deviations from the SM, high loading densities of short-lived nuclides for example are necessary, independent of the specific type of trap that will be used. New know-how and skills of scientific and technical staff will be developed.

**Perspectives**

The neutron decay experiments will benefit considerably from the advent of intensity-enhanced ultrasonic neutron beams that will be available in the near future at various reactor and accelerator centres. Experiments using atomic traps are now being prepared at several places in Europe, e.g. the Nuclear Accelerator Institute KVI in Groningen (Netherlands), Leuven (Belgium), and Legnaro (Italy). The joint efforts of atomic and nuclear physicists from various countries — possibly with support from the European Union — can be expected to lead to exciting results in the next few years.

### 2.3.3 Neutrino masses

The recent results at SuperKamiokande [31] claim, for neutrino oscillation, muon neutrinos oscillating to tau neutrinos. The combination of the high-precision elastic scattering data of this experiment with the solar neutrino data from the Sudbury Neutrino Observatory [32] provides strong evidence for neutrino oscillations and therefore indicates the existence of massive
neutrinos. This is at present the strongest hint for physics beyond the minimal SM.

A direct search for the existence of massive electron neutrinos is being conducted with nuclear physics methods. The most stringent upper limits for the electron (anti)neutrino are about 2.5 eV/c² from two experiments using the MAC-E-Filter technique used both in Troitsk and Mainz. [33, 34]. The end-point of the electron spectrum from tritium decay has been chosen as the most promising case. Here the end-point is at a relatively low energy, the atoms involved are the most simple ones and the tritium β-decay is a super-allowed nuclear transition almost free from nuclear matrix element corrections.

The best upper limit for an effective neutrino mass which also implies a determination of the character of the neutrinos (Dirac or Majorana particle) comes from double β-decay experiments. The sensitivity currently reached is an order of magnitude better than in the kinematics experiments [35]. Its interpretation, however, is nuclear-model dependent and may be subject to cancellation.

Both experimental efforts should be continued. In the kinematics experiment a much improved technique will allow an extension of the sensitivity to the sub-eV level. The present second generation of double β-decay experiments with a sensitivity to half-lives of $10^{28}$ years will probe effective Majorana neutrino masses down to $10^{-2}$ eV. In addition these experiments will have huge implications for other fields of physics beyond the SM like the search for dark matter.

2.3.4 Experiments with antihydrogen

The production of antihydrogen atoms ‘at rest’ belongs to the most challenging projects of atomic physics at accelerators. A sufficient quantity of these objects would permit a comparison with the highly developed precision spectroscopy of hydrogen atoms. Two-photon spectroscopy of the energy difference of the
2s–1s states in both the hydrogen and the antihydrogen system benefits from its narrow relative line width. With an anticipated good knowledge of the line shape an extraction of the energy interval with an accuracy of $10^{-18}$ can be envisaged. The accuracy achieved at present is better than $10^{-13}$ and is the result of an impressive development with a gain of about one order of magnitude per year over the last decade [36]. Finally a comparison of the frequencies of the hydrogen and antihydrogen atoms would permit a CPT test at the level of accuracy as is established from the kaon system [37]. Such a determination is planned by two extremely challenging experiments at the CERN AD [38, 39].

Once the frequency is measurable, a test for the validity of the weak equivalence principle (WEP) can be performed for antimatter. It will be tested whether the 2s–1s frequency measured in an antimatter system depends in the same way on the change in the gravitational potential as for matter. The measurement of the transition frequency will be done for values of the gravitational potential on the way of the earth on its elliptic orbit around the sun. With a $10^{-18}$ frequency resolution, a test of a violation of the WEP at the level of the original Eötvös experiment could be performed.
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3. Nuclear Properties Studied with Atomic Physics and Condensed Matter Techniques

3.1 Impact of atomic physics on nuclear physics and vice versa

The very first precise information on properties of nuclei came from atomic spectroscopy and mass spectrometry. These methods were used to investigate the interaction of the nucleus with the electromagnetic field produced by the electrons surrounding the nucleus, or with an electromagnetic field applied externally. Such experiments provided nuclear ground state spins, moments, charge radii, and masses of stable nuclides as well as of many radioactive ones (Fig. 3.1) [1–2]. More recently, condensed matter techniques have been developed allowing one also to determine the spins and moments of excited nuclear states. It should be noted that all these techniques, based on hyperfine structure splitting or isotope shift measurements, yield model-independent information about the nucleus [2]. Therefore it is not surprising that the very precise data stimulated, for example, the development of the nuclear shell model. Certainly, there was and still is a significant impact of atomic physics on nuclear physics.

Also the opposite holds true. Nuclear physics has a strong impact on other branches of science, especially on atomic physics. For example, the elements heavier than uranium up to the ‘superheavy’ elements can only be produced artificially at accelerators or nuclear reactors. These heavy elements are testing grounds for relativistic effects in atomic physics and chemistry. Similarly, as discussed in Chapter 2, quantum electrodynamic (QED) corrections become quite important for heavy highly-charged ions with a single or few electrons or for exotic atoms like muonic atoms which are only provided by accelerators. As a final example it should be noted that the long isotopic chains accessible at radioactive ion beam facilities, on-line isotope separator (ISOL) or in-flight facilities, enabled King plots to be made over a large range of mass numbers and in this way provided a stringent test of the theory of isotope shift consisting of a volume and a mass effect.

Finally, there is a mutual impact and cross-fertilization between nuclear physics on one side and atomic and condensed matter physics on the other: Hyperfine fields in hydrogen-like systems can be calculated very precisely. The comparison of the experimentally observed hyperfine structure splitting in hydrogen-like electronic or muonic ions with that obtained for neutral atoms is a test of the ability of modern many-body perturbation theory to correctly predict the hyperfine field for the complex, many-electron, neutral atom. Still, the uncertainties of these calculations for neutral atoms are rather large. Similarly, the isotope shift in neutral atoms might be compared with the results obtained in muonic atoms or by electron scattering in order to test the calculations of the electron density at the site of the nucleus in many-electron systems. For the benefit of atomic physics such comparisons have only been feasible up to now in the case of stable nuclides. However, once the hyperfine fields produced by the electrons at the site of the nucleus are known, measurements of the isotope shift and hyperfine structure splitting will be possible on unstable isotopes for the benefit of nuclear physics.

Another area of mutual impact is the development of new techniques for the study of ground state properties of short-lived nuclides which, after development to sufficient robustness, can be applied in other fields of science. An example is Collinear Laser Spectroscopy, invented for and tailored to the determination
of nuclear ground state properties of short-lived isotopes at ISOL facilities. Later on, this method was applied to extreme trace analysis in environmental studies.

Excellent overviews and relevant publications on the topics addressed in this chapter are provided by the proceedings of the 1998 ‘Trapped charged particles and fundamental physics’ conference [3] and of the APAC Euroconferences [4]–[6]. Many of the techniques discussed in this chapter were developed for experiments at the on-line isotope separator ISOLDE at CERN, Geneva. Therefore, Ref. [7] is also a valuable source of information. In order to avoid extensive references, no explicit reference is given for information contained in the proceedings cited above.

### 3.2 Optical spectroscopy

#### 3.2.1 Recent achievements

In early nuclear physics, the main source of information on ground-state properties of the nucleus was optical spectroscopy by using spectral lamps and interferometers. Later, radio-frequency methods such as ‘atomic beam magnetic resonance’, ‘double resonance spectroscopy’ and ‘optical pumping’ were introduced; but today it is almost exclusively laser spectroscopic techniques that are used. Owing to the large cross-section for absorption of optical photons (10^{-10} cm^2) and the high available laser intensity with small bandwidth, experiments with a single atom became possible.

Today, it is mainly two laser techniques that are applied: ‘resonance ionization mass spectroscopy’ and ‘collinear laser spectroscopy’. The first technique offers in general the highest sensitivity while the second provides the highest resolution. At ISOL facilities, the present limit for the minimum yield for laser spectroscopy is of the order of 100 ions/s and the shortest-lived isotope investigated is ^{11}\text{Li} with a nuclear half-life of T_{1/2} = 9 ms. In an in-flight production experiment the hyperfine structure splitting and isotope shift of radioactive atoms with a nuclear half life as short as T_{1/2} = 1 ms has been determined at a production rate of about 1 ion/s. In principle, isotopes with shorter ground-state half-lives would still be accessible by laser spectroscopy, which depends only on production
yield and not on nuclear half-life. Condensed-matter techniques like ‘perturbed angular correlation’ are routinely applied for the investigation of spin and moments of very short-lived, excited nuclear states.

A very promising development in recent years is that of the ‘laser ion source’ at ISOL facilities. By replacing the conventional ion source with step-wise resonant photoionization in a hot cavity or tube, a rather high ionization efficiency and a remarkable suppression of isobaric beams can be achieved.

To name some examples for the impact of optical spectroscopy at accelerators, the following highlights should be mentioned: measurement of spin, magnetic moment and of the quadrupole moment of the halo nucleus $^{11}$Li, the discovery of shape co-existence and shape staggering as critical phenomena in the neutron-deficient mercury region (Fig. 3.2), the determination of the deformation in the second minimum of the fission isomer $^{242m}$Am, the indications for octupole deformation in the radium isotopes, the measurements in hydrogen-like heavy ions of the hyperfine splitting in the ground state by laser and of the 1 s Lamb shift by X-ray spectroscopy.

![Figure 3.2: Changes in the mean-squared charge radii in the mercury region as obtained by isotope shift measurements.](image)

### 3.2.2 Future directions

It can be expected that the nuclear ground-state properties will be determined for nearly all isotopes of the chart of nuclei that are produced with production yields exceeding about 100–1000 ions per second. In special cases, also dedicated laser experiments might be performed on nuclides available only in minute quantities of one ion per second or below. Thus the planned radioactive ion beam facilities of the next generation will enable extension of the knowledge of nuclear ground-state properties in isotopic chains to regions further from the valley of nuclear stability.

Isotope shift measurements have been performed on radioactive isotopes (see Fig. 3.1). Up to now, the lightest element studied is in this way has been sodium. Especially for the light halo nuclei, it would be essential to know the neutron radius. Since the matter radius is known, a measurement of the charge radius by isotope shift determination would allow one to deduce that of the neutron distribution.

In addition, the ground-state properties of isotopes heavier than uranium will become accessible by fusion reactions, subsequent mass separation in gas-filled separators or Wien filter-type separators like SHIP and by ion traps coupled to these facilities. Such new installations, extending the range of present ISOL facilities far off stability or beyond uranium, or intended for other investigations, are in operation, being built or being planned at Argonne National Laboratory, GANIL/Caen, GSI/Darmstadt, Jyväskylä, KVI/Groningen, MAFF/Munich, Michigan State University and TRIUMF/Vancouver. Also the isotopes of refractory elements (not available at ISOL facilities) will become accessible at IGISOL (ion guide isotope separator on-line) and in-flight facilities. Here, the newly developed radio-frequency quadrupole (RFQ) cooler (which is a...
linear, segmented, gas-filled, four-rod structure, see Fig. 3.3) will be used to retard and accumulate the ions of a radioactive beam. After cooling, the ions are ejected as a bunch with much improved emittance. In this way, higher accuracy, sensitivity and signal-to-background ratio will be obtained as recently demonstrated at Jyväskylä for the case of collinear laser spectroscopy of Hf isotopes.

Furthermore, nuclear polarization of radioactive beams will be obtained by optical pumping of neutral atoms, singly-charged or hydrogen-like ions with possible applications, for example in weak interaction studies (see Chapter 2). Measurements of the nuclear g-factor and the A-factor (characterizing the magnetic hyperfine structure splitting) will give model-independent information on the distribution of nuclear magnetism in the nucleus via a determination of the Bohr–Weisskopf effect. A comparison of the nuclear g-factor measured in bare nuclei or hydrogen-like ions with that of a neutral atom will provide the first proof of the calculation of diamagnetic shielding corrections.

### 3.3 Mass spectrometry

#### 3.3.1 Recent achievements

In recent years new techniques have been developed for mass spectrometry, all based on frequency measurements. These are Penning trap mass spectrometry, radio-frequency transmission spectrometry and the use of storage rings or cyclotrons for such purposes. In all cases, high efficiency as well as isobar and even isomer separation can be achieved. The highest resolving power, of up to $R = m/\Delta m_{\text{FWHM}} = 10^9$, is obtained for Penning traps yielding an accuracy of about $\delta m/m = 10^{-10}$ in the case of stable isotopes or the antiproton. For radioactive nuclides, the typical mass accuracy is in the range of $10^{-7}$ to $10^{-8}$. The isotopes with the shortest half-lives investigated so far are $^{33}\text{Ar}$ ($T_{1/2} = 173$ ms) with an accuracy of $\delta m/m = 1 \times 10^{-7}$ and $^{74}\text{Rb}$ ($T_{1/2} = 65$ ms) with an accuracy of $\delta m/m = 3 \times 10^{-7}$.

An older technique newly applied to exotic nuclides is that of a radio-frequency transmission spectrometer (MISTRAL at ISOLDE). By not stopping and cooling the injected ion beam, MISTRAL gives access to ISOLDE’s shortest-lived nuclides. A resolving power superior to $10^8$ and a precision of $10^{-7}$ are obtained. The implementation of a segmented RFQ cooler will improve the sensitivity even further.

At GANIL and Grenoble, mass spectrometry was performed using the cyclotron itself as a mass spectrometer. By use of the second cyclotron CSS2 at GANIL, even the mass of $^{100}\text{Sn}$, the extremely rare nuclide that has been hunted for a long time, could be determined recently.

For storage rings, two techniques were developed: i) Schottky mass spectrometry where the ions are electron-cooled prior to mass spectrometry by detecting the image currents induced by the circulating ions in pick-up electrodes; ii) time-of-flight mass spectrometry where the storage ring is operated in the isochronous mode and the emitted secondary electrons are recorded when the ions pass a thin carbon foil inserted into the storage ring. The direct coupling of the storage ring to a fragment separator, where the radioactive nuclides are produced and mass separated in high charge states (bare, hydrogen and helium-like) makes storage-ring mass spectrometry quite fast and efficient.

In the case of Schottky mass spectrometry, the half-life limit is about 10 s, whereas the time-of-flight technique has the prospect of measuring the mass of isotopes with a half-life as short as some 10 $\mu$s. The isotope with the shortest half-life investigated so far is $^{48}\text{Cr}$ with $T_{1/2} = 50$ ms (Fig. 3.4). With typical resolving powers of $R = 5 \times 10^5$ (Schottky) and $R = 2 \times 10^5$ (time of flight) high-quality mass data are obtained. In addition, the storage ring technique is very efficient since the masses of a large number of isotopes are determined simultaneously.

The achieved mass accuracy in the range of $10^{-6}$ to $10^{-7}$ is already beyond the predictive capability of the state-of-the-art calculations of nuclear binding energies. It therefore allows one to test nuclear models as well as to check and improve the predictive power of mass formulae required to model, for example, the r-process in stellar explosions. The masses of several
Figure 3.3: Schematics and photograph of the linear, segmented, gas-filled Paul mass filter used at ISOLDE to retard and accumulate the 60 keV ion beam. Collisions with helium buffer gas and the radio frequency applied to the four rods serve to cool the ions and to confine them in radial direction. Segmentation of the rods and application of DC potentials provide a potential well in the longitudinal direction. By changing the DC potential applied to the segments (see dashed line in the V(z) graph) the cooled ions can be ejected as a bunch.

Figure 3.4: Time-of-flight spectra of highly charged ions taken at the Experimental Storage Ring (ESR), GSI in the isochronous mode. The number of particles observed is plotted against the revolution frequency of the different ion species in the storage ring.
hundred short-lived isotopes have been measured in recent years at GSI by use of the Experimental Storage Ring (ESR) and the fragment separator (FRS) or at ISOLDE with the triple-trap spectrometer ISOLTRAP.

For testing atomic theory, a higher accuracy is required than in nuclear physics. Here, an accuracy of 1 eV or even better would be desirable. This corresponds to an accuracy of $\delta m/m = 10^{-11}$ for ions with mass number $A = 100$. At present, the best mass measurements on heavier atoms reach $10^{-10}$, performed by the MIT group on singly-charged ions. The mass spectrometer SMILE, which uses highly-charged ions produced by an Electron Beam Ion Source (EBIS) at Stockholm, provides a typical mass accuracy of ppb. The cryogenic trap developed at Mainz for g-factor measurements on hydrogen-like ions (for the HITRAP facility at GSI) with a demonstrated resolving power of $R = 109$ for $^{12}\text{C}^{5+}$, promises an accuracy of the order of $10^{-10}$.

3.3.2 Future directions

Whereas the storage-ring experiments for mass spectrometry at the ESR/GSI and those with ISOLTRAP at ISOLDE/CERN have been going on for several years, the first mass measurement on unstable nuclei has been recently reported from the Canadian Penning Trap (CPT) at Argonne National Laboratory. Many other traps are being installed or planned all around the world, mostly coupled to ion separators (JYL-TRAP at Jyväskylä, SHIPTRAP and HITRAP at GSI/Darmstadt, ISAC-TRAP at TRIUMF/Vancouver, LEPI at Michigan State University (MSU), and MAFF-TRAP at Munich). Storage-ring facilities are planned at RIKEN in Japan and Lanzhou in China.

The primary goal for future mass measurements by storage rings and traps is to obtain higher efficiency or transmission in order to study nuclei further from stability. Penning traps will be applied to perform the first direct mass measurements on superheavy elements produced in fusion reactions. An exact knowledge of the binding energies and shell corrections of these heaviest nuclei will be important for the discovery of further heavy elements. Here, non-destructive single-ion detection techniques are required for making the most efficient use of such rare events.

Also, still higher accuracy of better than $\delta m/m = 10^{-8}$ will be an issue for certain nuclides such as, for example, the $Q$-value of the $0^+ \rightarrow 0^+$ Fermi transitions for verifying the unitarity of the CKM matrix of the Standard Model. Part of this gain in accuracy will be achieved by using carbon clusters as an absolute mass reference, as demonstrated recently at ISOLTRAP. Another option is to increase the cyclotron frequency. The line width of the cyclotron resonance is ultimately determined by the time of observation or by the time of interaction of the ion with the radio-frequency field. This period of time is limited by the nuclear half-life. As a consequence, only an increase of the cyclotron frequency will result in higher resolving power and thus in higher accuracy or enable an extension of Penning-trap mass spectrometry to even shorter-lived radionuclides.

Such higher cyclotron frequencies can be obtained by use of a higher magnetic field for the Penning trap as foreseen at MSU. Another possibility is the use of higher harmonics. Finally, highly charged ions can be used to increase the cyclotron frequency, as demonstrated at SMILE-TRAP and planned for ISAC-TRAP at TRIUMF. Use of highly-charged ions is also foreseen for HITRAP at GSI via a non-destructive cyclotron resonance detection at liquid-helium temperature. This will enable an accuracy of the order of $10^{-10}$ or even better to be achieved for nuclides with a half-life exceeding about one second (limited by the period of time required for deceleration) or stable ions of interest for atomic physics. In the latter case, direct mass measurements of bare or few-electron ions up to uranium will yield information on atomic binding energies and correlation effects in few-electron systems as well as on QED corrections in extremely strong electromagnetic fields approaching $\alpha \cdot Z \rightarrow 1$.

Finally it should be mentioned that a heavy-ion accelerator combined with the single-ion detection efficiency and the mass spectrometric
3. Nuclear Properties Studied with Atomic Physics and Condensed Matter Techniques

capability provided by a storage ring represents a powerful tool for extreme trace analysis. The application of this combination for accelerator mass spectroscopy has not yet been exploited, in particular for heavy particles where the isobaric problem prevents investigation by tandem accelerators.

3.4 Nuclear half-lives

3.4.1 Bound beta-decay

The measurement of nuclear half-lives of ground and isomeric states is, technically, just a by-product of mass measurements in storage rings. It is achieved by observation of the amplitudes of the Schottky signals as a function of time or by observing the number of turns in the storage ring by the time-of-flight detector. In the latter case, such measurements are restricted to half-lives shorter than about 10 ms since the highly charged particles undergo charge exchange in the carbon foil.

Nuclear half-lives of neutral atoms are routinely measured by the various well-established methods of nuclear decay spectroscopy. Ion cooler rings and ion traps are at present the only tools that preserve atoms for a long time in a high charge state and allow the observation of nuclear decays, in particular beta-decay, of highly ionized atoms. The impact of this new possibility for our understanding of the creation of heavy nuclei in stars is obvious: In a hot stellar environment (kT of the order of a few tens of keV) all atoms that are produced become highly ionized. Then a new beta-decay channel, the bound beta-decay, opens, where the emitted electron will be bound in an inner atomic shell (predominantly the 1s shell) of the daughter atom.

Whereas bound beta-decay is a completely negligible decay branch for neutral atoms due to the lack of empty inner-shell atomic states, it can lead to a significant and sometimes even dramatic change of beta-half-lives if the atoms are highly ionized, as evidenced in a pioneering experiment on 163Dy. Bound beta-decay has a crucial but previously overlooked impact for radioactive mother/daughter pairs that serve as ‘clocks’ for the age of our galaxy (and the universe). Together with 238U/232Th, the couple 187Re/187Os has been widely accepted as providing the most reliable limits for the age of our galaxy, supposing only that both the beta-half-life of 187Re and the relative abundance of 187Re and its beta-daughter 187Os are precisely known. An experiment at the storage ring ESR has shown, however, that the half-life of 187Re depends dramatically on its atomic charge state (see Fig. 3.5): the half-life of 42 × 10^9 yr for neutral 187Re is shortened to only 14 yr if all of its electrons are stripped off [8]. As a consequence, all the different charge states of 187Re during its galactic history must be taken properly into account to deduce the age of our galaxy. It can be argued that similar restrictions also hold for the few other galactic clocks.

![Figure 3.5: Principle of bound-beta decay of 187Re.](image)

In the future, mass-resolved Schottky spectroscopy will be performed, providing simultaneously the relative strengths of continuum \( \beta^- \) decay and of bound beta-decay and covering a broad range of nuclei and charge states. Thereby one obtains the ratio of the bound 1s and of the continuum electron wave function, i.e. essentially the Fermi function. For \( \beta^- \) decay, the latter has never before been probed by experiments in contrast to \( \beta^+ \) and orbital electron capture decay.
3.4.2 Internal conversion between bound states

In most cases the nuclear and atomic parts of one atom can be considered as two separate entities. The consequence is that the detailed properties of one part of the system do not affect (or very weakly affect) the properties of the other part. Nevertheless, these two independent systems can exchange energy as is well known from the decay of nuclei by internal conversion. A virtual photon from the nucleus scatters one electron which is sent into the continuum with an energy equal to the difference between the nuclear transition energy and the binding energy of this electron.

However in some cases, the two parts of the atom cannot be considered as separate and isolated from each other. One of these cases arises when the excitation energy of the nucleus and the excitation energy of the electronic system are nearly equal. In this situation an atom can be formed in an excited state in which the excitation energy is independent of its distribution between the nucleus and the atomic core. Therefore the two components of the atom can resonantly exchange their respective energy. The atom can be seen as an oscillating circuit with an extremely large $Q$ value (if this atom was isolated with no other possibility of exchanging energy with the surrounding world).

If the energy flows from the nucleus to the atomic system, the process is called Internal Conversion between Bound states (BIC). It has recently been observed at GANIL in $^{125}$Te [9]. If the energy flows from the atomic system to the nucleus, the process is called Nuclear Excitation by Electronic Transitions (NEET). It has been convincingly observed in $^{197}$Au [10]. The two processes are shown schematically in Fig. 3.6. In the case of $^{125}$Te, the M1 nuclear transition between the first excited state and the ground state was brought into resonance with the electronic transition between the 1s state and the 1s states by removing 45 electrons from the Te atom. In $^{125}$Te$^{45+}$ the difference between atomic and nuclear excitation energies is less than 3 eV. Taking into account the natural width of the 1s hole state of $\Gamma = 5$ eV, one sees that the matching of the two energies is achieved in such a situation.

One of the consequences of BIC is the opening of a new channel for the decay of the nucleus, which adds to the decay rates in the other usual channels such as gamma emission. This therefore affects the half-life $T_{1/2}$ of the nuclear excited state. It is interesting to note that now the lifetime of the nuclear excited state depends directly on the total width of the excited atom state. The excited atom may decay either because the nucleus decays or because the electron core decays by X-ray or Auger emission [11]. Since the energy resonance condition is only achieved for a small number of ionic charge states, the nuclear half-life becomes resonant for some particular charge states. Direct implications of BIC may be found in the variation of nuclear half-lives with the charge state in astrophysical plasmas and nuclear synthesis of the elements. Furthermore one can think about the possibility of using this resonance condition for triggering nuclear decay in the storage of nuclear energy and the development of a gamma-ray laser.

![Figure 3.6: Schematics of Internal Conversion between Bound states (BIC, left) and Nuclear Excitation by Electronic Transitions (NEET, right) recently seen at GANIL.](image)

3.4.3 Effect of nuclear dissipation on fission times: a study by ion blocking

Ion blocking in single crystals, a solid-state and atomic physics technique, coupled with a nuclear calorimetry technique has recently been used in nuclear physics to probe the fission time distributions at high excitation energies up to very
long times [12]. These cross-disciplinary experiments give access to quite important information about a basic quantity, the nuclear dissipation involved in the fission process and its evolution with temperature. Nuclear dissipation arises from the viscosity of nuclear matter; it tends to oppose the deformation of a hot nucleus and to maintain the stored energy in its thermal form. As a consequence, the probability of the nucleus cooling by particle emission increases during (or instead of) the fission process. Then, the magnitude of nuclear dissipation affects in a crucial way the fission probabilities and the mean fission times of excited nuclei. Knowledge of this fission time is important in considering the possibility of studying (or accelerating) very exotic fission fragments, or of synthesizing superheavy nuclei from hot fusion reactions.

The interpretation of standard experimental studies performed to investigate nuclear dissipation (analysis of fission rates [13], or analysis of pre- and post-scission multiplicities [14]) depends on various assumptions and leads to contradictory results. The blocking technique in single crystals is model-independent and permits the fission time distributions to be probed for up to very long times in a very simple and direct way. Here, fission is the result of the nuclear excitation produced by the nuclear collision between a swift ion and an atom of a crystal lattice. There is a one-to-one relationship between the fission time and the distance from lattice site at which fission occurs. If this distance is small at the scale of lattice parameters, i.e. if the fission time is short, fragments emitted along major crystallographic axes or planes will be deflected by a set of correlated atomic collisions. As a consequence, there will be a lack of fragments leaving the crystal parallel to these directions (blocking effect). Fitting the angular distribution of the emerging fragments by Monte Carlo simulations then provides the mean fission time and some information on the fission time distribution. This technique has been known for quite some time and has already produced many interesting nuclear lifetime measurements.

The main novelty of the experiment reported in Ref. [12] is that the fission times are directly obtained as a function of the excitation energy of the nuclei that undergo fission. Practically, swift uranium ions were sent in a silicon thin crystal. The angular distribution of the emitted fragments was measured together with the neutron emission yield of each nucleus submitted to fragmentation, providing the excitation energy. The amplitude of the blocking effect (related to the mean fission time) is shown in Fig. 3.7 [12] as a function of excitation energy. Low values of the reported ‘blocking ratios’ correspond to strong blocking effects. The corresponding mean fission times are also shown in the figure. For low excitation energies, the mean fission time becomes very long. These results demonstrate that the nuclear physics approaches yielded fission times much too short, and this could be the origin of the inconsistencies previously reported. In the future, more refined experiments coupling the blocking technique with nuclear physics techniques should permit a rather precise determination of the nuclear dissipation and its possible evolution with temperature and mass.

![Figure 3.7: Blocking effect and fission time as a function of excitation energy.](image-url)
Bibliography


4. Interaction of Atomic, Molecular and Cluster Ions with Matter

4.1 Introduction

Research in this field in the last few years has resulted in astonishing progress in the understanding of the dynamics of particle–matter interaction. Key to these advances were the availability of high-performance ion storage rings, of swift ion and cluster accelerators producing intense beams with excellent optical quality and of ion sources providing highly charged slow ions. Moreover, novel developments in target preparation techniques (atomic traps [1], supersonic jets) along with the recent breakthrough in atomic many-particle imaging techniques (such as recoil-ion and low-energy electron momentum spectroscopy [2]) have opened up the opportunity to study the collision dynamics in unprecedented detail. The fundamental goal is to probe the response of matter under the influence of strong and short pulses of electromagnetic radiation. Concepts related to the non-linear response of complex systems can be tested. Examples include the study of slow highly charged ions and clusters, of sub-attosecond pulses, and of strongly coupled non-ideal plasmas. Targets of increasing complexity ranging from individual atoms in dilute media and clusters to surfaces and bulk matter can be investigated. The latter play a crucial role for the understanding of the first stages of material modifications discussed in Chapter 5. Spectacular improvement has also been achieved in the high level of accuracy reached to test fundamental aspects in atomic structure (QED), using other methods than those described in Chapter 3. There is also a strong impact on many other scientific areas, including astrophysics and astrochemistry, atmospheric physics, radiobiology and radiation chemistry. We limit ourselves to a brief discussion of a few examples of important studies performed recently.

4.2 Interaction with dilute media

4.2.1 Electronic excitation and charge exchange processes

At heavy-ion accelerators (GANIL, UNILAC/SIS) and at the ESR storage ring facility the interaction of highly charged ions with matter can be explored in a unique regime that is governed by virtual as well as by real photon fields, advancing our basic knowledge about the physics of strong fields. At high velocities it has been demonstrated recently that the sub-attosecond ($t < 10^{-18}$ s), super-intense ($10^{20}$ W/cm$^2$) electromagnetic field, generated by the moving ion, can be interpreted as a pulse of virtual photons (multiply) exciting or ionizing the target [3]. In this limit, two-electron processes such as simultaneous excitation of two K-shell electrons in lithium forming hollow atoms [4] or double and multiple ionization reactions at minimum momentum transfer [5,6] provide a unique tool to investigate dynamic electron correlations on ultra-short times-scales that have not previously been accessible.

At relativistic ion energies, the electromagnetic interaction becomes strongly affected by the magnetic field component significantly influencing the population of excited states [7]. Real photon fields are involved when a target electron is captured into a bound state of a fast highly charged ion with the simultaneous emission of a photon. Angle-resolved photon detection during this time-reversed photoionization process is a novel and, at present, the only approach to investigate photoionization of highly charged ions at hundred keV photon energies [8]. Here, spin-flip contributions mediated by the magnetic component of the electromagnetic field have been unambiguously identified for the first time (see Fig. 4.1).
Figure 4.1: Angular distributions for the time-reversed photoeffect, i.e. radiative electron capture, measured for bare uranium ions at 88 MeV/u in collision with N\textsubscript{2} (full line: complete relativistic calculations, dashed line: sin\textsuperscript{2}θ distribution, shaded area: spin-flip contributions) [8].

In the last few years, C\textsubscript{60} fullerene molecules have become fashionable targets for collisions with highly charged ions at low velocities because they bridge the gap between atoms and surfaces (for a review see Refs. [9, 10] and references therein). As in the case of ion–surface interaction (see Section 4.4.3), the formation of hollow atoms is observed. Also, an image charge (induced by the polarizability of C\textsubscript{60} molecules) causes an acceleration of the collision partners during the approach. Unfragmented fullerenes with charge states up to q = 10+ have been observed after collision with slow Xe\textsuperscript{25+} [10]. A recent experiment has shown that the number of electrons active during the collision is even much higher (up to six times) than the final fullerene charge. For close collisions where the ion penetrates the cluster, the fullerene cage is destroyed through ionization and excitation. The small-fragment yield presents the same characteristic oscillatory behaviour with the projectile atomic number as the energy transfer. This demonstrates that energy deposition into the electronic degree of freedom is responsible for the destruction of the C\textsubscript{60} structure. Detailed studies of the fragmentation mechanisms and decay times have been undertaken to determine the relative importance of evaporation and fission at various excitation energies.

4.2.2 Dissociation and fragmentation of molecules and clusters

Collisionally excited states of the transient molecular ion, populated before the dissociation, have been determined for the CO molecule, shedding light on limitations of the Coulomb explosion model to reproduce the fragmentation dynamics [11]. Geometrical modifications during the break-up, time-sequencing of different processes and many-body Coulomb interactions have been explored for CO\textsubscript{2} [12] and H\textsubscript{2} [13], respectively. Of special interest for radiobiology, fragmentation of H\textsubscript{2}O induced by highly charged ion collisions has been investigated [14]. Striking results have been obtained in the investigation of cluster–atom collisions. There, the use of clusters as projectiles offers many technical advantages. For instance, the fragmentation of hydrogen cluster ions of high kinetic energy colliding on helium atoms provides information on phase transitions in finite systems [15]. Evidence is also obtained on the sensitivity of the multi-ionization cross-sections to the shape of the clusters [16] as well as on the unexpectedly strong size dependence of non-dissociative rates in electron capture collisions [17]. Future studies will concentrate on the role of the cluster–electron polarizability and the internal vibrational energy.

4.2.3 Collisions with trapped atoms

Atoms trapped by means of electric, magnetic and optical fields offer novel and exciting possibilities for atomic collision physics for various applications as well as for nuclear physics research. Recently, state-selective charge transfer measurements have been achieved down to 5 eV/amu where experimental cross-sections are found to be orders of magnitude higher than theoretical predictions [18]. At present, a
magneto-optical trap is implemented in the Heidelberg test storage ring TSR and experiments are in preparation to use this ultra-cold, μK target as a beam profile monitor and for recoil-ion spectroscopy. First encouraging results, obtained at Groningen, Aarhus and at KSU, demonstrate that an unprecedented recoil-ion momentum resolution will be achievable with these instruments. Fascinating perspectives are envisaged using highly charged recoil ions, emerging from collisions with fast ions, as ultra-low-energy secondary projectiles for collision processes at astrophysically relevant collision energies in the sub-meV range.

4.2.4 Perspectives and technical developments

A major effort is dedicated to precisely preparing, controlling and, in many cases, efficiently cooling highly charged atomic or clusters ions as well as neutral atoms and molecules. The aim is, on one hand, to reach ultimate precision in the investigation of atomic structure and collision dynamics. On the other hand, reliable data as well as new information will be provided for other fields of physics where the knowledge of fundamental collision processes is needed (nuclear physics, plasma physics, astrophysics). At the upcoming TESLA–FEL (Tera-electronvolt Energy Superconducting Linear Accelerator–Free Electron Laser) in Hamburg, basic non-linear atomic processes in the interaction of this super-intense, short-pulse radiation with surfaces, biomolecules, magnetic materials and condensed matter will be studied. First differential measurements on multiple ionization of atoms and molecules in intense femtosecond laser fields open the door to such future experiments [19].

4.3 Interactions with free electrons and plasmas

Since Danared [20] first employed adiabatic expansion to lower the transverse temperature of electrons in coolers, collisions of atomic and molecular ions with free electrons of temperature around 1–10 meV are extensively explored in storage rings. The quality (density and energy spread) of the electron source is essential. New states of atoms, molecules and clusters (positively or negatively charged) are formed. Their study provides sensitive tests of refined theoretical predictions. These studies are related to important issues in astrophysics, chemistry and atmospheric physics. Free-electron–ion collisions are also of prime importance in plasmas with potential applications related to heavy-ion inertial fusion.

4.3.1 Studies with positive atomic ions

The increased resolution, obtained in recent years through improved cooling devices, has made dielectronic recombination an excellent tool for spectroscopic studies to improve our understanding of the formation of atoms and the binding of their electrons. Our knowledge about atomic structure and quantum electrodynamic (QED) corrections to energy levels is almost exclusively obtained through photon spectroscopy (see Chapter 3). Although it would hardly have been anticipated a few years ago, resonances in collision cross-sections provide today an alternative access to highly accurate information on atomic energy levels. Several studies have been performed with highly charged Li-like ions and an energy splitting in Cu-like Pb, carrying a large QED correction, was determined with relative accuracy of \( \sim 10^{-5} \) [21]. Spectroscopic studies with accuracy in the measurements of energy splittings of the order of \( \sim 10^{-6} \) are envisaged for the future.

Ion–electron recombination studies also provide information on dynamic aspects of the collision process, when varying the electron energy. As an example, we show in Fig. 4.2 the recombination spectrum of F\(^{6+}\) [22,23].

This work addresses in particular the longstanding question of enhanced radiative recombination (RR) at low energies [23, 24, 25]. Possible explanations include an enhancement of the electron density in the ion vicinity originating both from the external magnetic field and from the Coulomb field induced by the ions [23].

In the scenario of heavy-ion inertial fusion,
the energy deposited in a target by swift heavy ions is mainly converted into X-radiation. The efficiency of this X-ray converter depends on the spatial and time evolution of the beam energy deposition profile. When increasing the target temperature of low-Z materials, one observes a large increase of the average ionization state of the ions. The stopping power and the energy straggling also undergo a dramatic increase. This feature is related to the fact that atoms of low-Z targets are efficiently ionized at increasing temperature and thus form a plasma. The charge-state evolution and the energy loss of heavy ions passing through a plasma show pronounced differences when compared to the passage through cold gases and solid-state matter [26]. These results demonstrate that charge and energy fluctuations must be considered to predict correctly the profile of energy deposition.

Intense ion beams open new opportunities to investigate the interaction with dense plasmas and to study the hydrodynamic and radiative properties of beam heated matter. The first experiments made use of externally created plasmas. Today, dense plasmas in the temperature range of up to several 10 eV can be created directly by intense swift heavy-ion beams (at GSI for instance) irradiating a light target (hydrogen). In this temperature and density regime, the potential energy of the particles, constituting the plasma, is of the order of their thermal energy. In these strongly coupled non-ideal plasmas, ion beam induced effects, such as hydrodynamic instabilities or ‘metallization’ of hydrogen, are evident [27]. Plasma temperatures of up to 300 eV and higher, where highly charged species prevail, can be explored in beam-plasma interaction experiments with laser-driven plasma targets [28]. Future developments will involve a petawatt high-energy laser for ion experiments (PHELIX) which is currently under construction at the accelerator facilities of GSI.

4.3.2 Studies of positive molecular ions

Significant progress has been made in studies of molecular cations in storage rings. During the long storage time (many seconds), these molecules may cool down to the lowest vibrational state by slow infrared radiative transitions. In this way, well-defined molecular states can be studied [29].

When a positive molecular ion captures a free electron by exciting one of its bound electrons, it may respond by emitting an electron by autoionization. However, the resulting electronic change normally yields a neutral molecule in an unstable, dissociating state which may lead to the formation of a number of neutral atomic and molecular fragments. In this case, the process is termed dissociative recombination: a process of great importance but very hard to treat theoretically.

Three important issues should be mentioned here (see Ref. [29]). One concerns H$_3^+$, a species that plays an important role in interstellar chemistry. The rate coefficient for dissociative recombination of this ion was measured at the CRYRING storage ring where the branching ratios (H versus H$_2$ production) were also determined. At the ASTRID storage ring, an imaging technique was applied to measure the kinetic energy release of the atomic fragments.
in the dissociative recombination of \( \text{O}_2^+ \). The dissociative recombination leads to a substantial population of oxygen atoms in the metastable \( ^1\text{S} \) state. The resulting \( ^1\text{S} \rightarrow ^1\text{D} \) transition is responsible for the green light emission of the Earth’s ionosphere. In a recent work at the TSR storage ring, Coulomb explosion together with the fragment imaging technique was used to obtain rate coefficients for individual initial vibrational levels \( (\nu = 0, 1, 2, \ldots) \) of the \( \text{HD}^+(\nu) \) ion. This work provides a very critical test for the understanding of the dissociation dynamics of small molecules. Recently, the first studies of dissociative recombination of twofold positively charged molecular ions, where the dissociation follows repulsive curves of the mono-cation, have also been carried out [30].

### 4.3.3 Collisions involving negative ions

Most atoms and molecules form stable negative ions and many of these play an important role in man-made plasmas and in nature (e.g. in the interstellar space and in planetary atmospheres). The electron-impact detachment cross-section of negative ions has been investigated at the ASTRID storage ring. The aim is to understand the dynamics of the detachment process and to study the possible formation of small doubly charged negative ions (dianions), a new form of ions in the gas phase where electronic correlation is expected to be extremely important. It appears that atomic dianions do not exist. However, many small molecular ions form highly unstable dianions. This formation is revealed by the presence of resonances in the scattering cross-sections. Figure 4.3 shows an example with \( \text{NO}_2^- \) where two resonances are visible [31]. \textit{Ab initio} calculations show that the lower one corresponds to the dianion ground state.

### 4.3.4 Perspectives

The magnetically confining heavy-ion storage rings are widely used for atomic and molecular ions where the mass-to-charge ratio is not too big. However, very heavy molecular and cluster ions cannot be stored at sufficiently high energy to match the speed of the electrons of the electron cooler in the merged beams configuration of the rings. Crossed-beam set-ups are being prepared for studies of electron interactions with gas-phase biomolecular ions and cluster ions in new electrostatic storage rings [32]. Newly developed electrostatic traps have also been used to investigate molecular or cluster ions [33].

### 4.4 Interactions with condensed matter: bulk and surfaces

Apart from quantities of immediate practical relevance such as mean charge states and stopping power of the projectile, an improved microscopic understanding of the complex array of interaction processes of ions with solids has been the focus of recent investigations. In addition to their own fundamental interest, these studies have an impact on many different subfields in physics. For example, knowledge of the projectile electronic state in matter permits optimization of the injection of high-intensity beams for radioactive beam production or for spallation neutron sources and better control of beam-induced material modification. The latter includes track formation and flux pinning in high-temperature superconductors, selective sputtering, and fundamental concepts of the non-linear response of matter to strong fields (Chapter 5).
4.4.1 High-velocity ions inside solids

As a swift ion penetrates a solid, it undergoes a large number of collisions with ionic cores as well as valence and conduction band electrons (‘electron gas’), leaving behind a track of electronic excitation while simultaneously changing its charge state and state of excitation.

While the basic properties of the stopping power at high energy are well described by the perturbative theories of Bethe and Bohr, recent studies with highly charged ions — at GANIL (Caen) and at the Van de Graaff CN accelerator of Legnaro National Laboratories (Catania) — have added new features. Measurements of absolute electron emission cross-sections have been performed with large detector arrays (ARGOS), initially designed for nuclear physics studies. One of them is the production of ultrahot electrons [34] in the target with energies $E_e$ beyond the classical binary encounter limit $E_c$, $E_e > E_c$ in both forward and backward directions. They have been identified as a signature of the so-called shuttle acceleration, originally proposed by Fermi to explain high-energy cosmic rays and later invoked for the acceleration of ionized electrons and atoms in particle–solid collisions [35].

The production and transport of excited projectile states is also a powerful tool for the study of ion–solid interactions. Only recently have detailed measurements and theoretical description of the approach towards equilibrium in the excited states of highly charged projectiles become possible. The measurements were achieved via high-resolution X-ray spectroscopy [36]. The complex array of interaction processes between projectile electrons and the solid (‘the environment’) can be described as an open classical or quantum system employing master equations [36, 37] and Monte Carlo wave function techniques [38]. The resulting excitation dynamics bears signatures of the wake field due to the collective response of the solid [39]. The modelling of this complex process has reached a state at which detailed quantitative predictions of the nl distributions of the excited ions have become feasible.

One important area where these advances are useful is the control and optimization of ion beam production. High intensity beams are used as injection for radioactive beam production (SPIRAL-Caen, for instance) or for spallation neutron sources. To optimize the injection intensity, the projectile excited state population in matter must be known with high accuracy. For example, in the injection design for accumulator rings for spallation neutron sources, high-intensity $H^-$ beams are stripped to protons by transmission through carbon foils. Even small fractions of contaminant excited neutrals $H(n)$ in low $n$ states ($n = 2, 3, 4$), that can be ionized by motional electric fields, lead to an ill-defined magnetic rigidity of the beam and to unacceptably high levels of radioactivity. Accurate determination of the nl distribution of the emergent beam is therefore of crucial importance for the design future megawatt beam injectors. The ETACIIA code [36], using binary atomic collision cross-sections as input, and a recently developed quantum transport theory [38] can successfully predict the evolution of charge-state distributions of high energy ions at the exit of solid targets.

Exciting developments are also likely to emerge in the area of channelling: One topic is coherent multiple scattering in ion–solid interactions, specifically employing the Okorokov effect. A projectile, propagating along a low-indexed channelling direction of a crystal, experiences a time-periodic potential due to the spatial periodic crystal potential. If its frequency is tuned to a resonant transition in the projectile, resonant coherent excitation (RCE) can ensue. Recent work at the HIMAC facility in Japan has reached a spectral resolution of this coherent source of virtual X-rays of $\Delta E/E \sim 10^{-5}$. Such high resolution is within striking distance for probes of QED effects in highly charged ions [40]. An even more advanced goal for the future is to drive coherently $\gamma$ excitations in nuclei. Another topic of current interest is recombination of channeled ions. These ions explore regions in which the electron density arises mostly from valence and conduction electrons. Cross-sections associated with electron impact ionization have been measured with precision; detailed studies of the impact parameter dependence of stopping
power have been conducted [41]. Radiative electron capture and resonant transfer and excitation are used to yield information about the momentum distribution of the electrons captured far away from atomic strings. One can also search for new processes with very low cross-section like trielectronic recombination.

4.4.2 Cluster and molecular ions inside solids

New features emerge when more complex projectiles such as molecules and clusters are used. When such projectiles penetrate a solid, there is a transient regime during which the distances among the atomic species of the cluster are comparable to the interatomic distances in the target. Therefore, these species interact in a coherent way with each other. As the penetration depth increases, the combined effect of Coulomb explosion and multiple scattering enlarges the distance between the components, and the latter interact with the target in an increasingly independent way. The processes studied are energy loss [42], secondary electron and induced ion emission, charge state of the cluster components and dynamics of cluster dissociation. The physical parameters involved are the cluster components, the cluster size, and the impact velocity.

At very high velocities (i.e. when the equilibrium charge state of the cluster components corresponds to bare ions), measurements with hydrogen clusters show that the stopping power and secondary electron emission efficiency of the cluster components are increased relative to those of individual ions. At lower velocities, however, the efficiency of the cluster components is smaller than that of individual ions [43]. Even more dramatic effects are observed for sputtering yields discussed in Chapter 5.

Recent theoretical studies [44] demonstrate the role of the screening in charge exchange processes for cluster projectiles formed of heavy atoms. Experimentally, a strong charge reduction is observed when MeV/atom atomic fragments of C_n clusters exit carbon foils [45]. This screening effect increases strongly with the cluster size. The dependence on cluster size demonstrates that the Coulomb repulsion between cluster constituents is strongly shielded with a screening length smaller than 2.5 Å.

4.4.3 Interaction of ions, molecules and clusters with surfaces

The advent of intense sources for slow highly charged ions opened up the possibility of exploiting the huge potential energy stored in these projectiles for highly localized surface excitation and modification. Applications ranging from nano-fabrication to information storage have been envisaged. Unlike kinetic sputtering which causes radiation damage in deep layers, potential sputtering of slow highly charged ions holds great promise for gentle nano-structuring [46] of insulator surfaces (for details see Chapter 5, Section 5.3.4). A profound understanding of the microscopic mechanisms responsible for the deposition and dissipation of potential energy is of great importance and currently pursued in many laboratories. In the case of metal surfaces, an understanding of the charge exchange processes has been reached based on the classical over barrier model [47]. However, materials accessible to surface modification are semiconducting or insulating surfaces, for which the neutralization and relaxation dynamics is still poorly understood. An artist’s view of the most relevant processes is shown in Fig. 4.4 [9]. Among them is the formation of ‘hollow atoms’ [47, 48] by multiple resonant charge transfer.

These transient multiply excited configurations feature empty or only sparsely populated inner shells. Because of the extremely short lifetime of this species limited by the time of impact at the surface, direct photon and electron spectroscopy of hollow atoms has been largely elusive.

This difficulty has recently been overcome with the advent of new target geometry, the interaction of multiply charged ions with the internal surface of microcapillaries [49]. Hollow atoms and ions can be extracted from the capillary before they undergo close encounters with internal walls. Observation of electron and photon emission from multiply excited atoms in flight downstream from the target has become
Figure 4.4: An artist’s view of processes induced by the approach of a highly charged ion to a surface [9].

possible. New decay mechanisms that have been proposed, such as ‘internal dielectronic excitation’ (IDE) [50], could be probed. Other features of distant interactions remain to be explored. Most notably, the highly charged ion experiences a drag force, equivalent to stopping power, when passing by a surface at distances tens of Å from the surface. First predictions of this type of dissipative polarization force, unexplored up to now, have become available [51].

Scattering at grazing incidence permits local probes of the surface layer. They include the transient trapping of the projectile in a skipping motion due to the image attraction [52, 53] and the production of surface excitons of LiF single crystals. Another avenue is surface magnetism. Spin polarization may serve both as a labelling technique for delineating the pathway of charge exchange and a source for the formation of long-lived, highly magnetized hollow atoms. Resonant electron capture predicts transfer from the conduction band mostly from near the Fermi edge, while ‘sidefeeding’ proposals emphasize the role of inner shells of the target. The spin polarization of a magnetized target ‘tags’ the magnetized conduction band. The first data [54] point, indeed, to a strong spin polarization of the hollow atom and, hence, to a quasi-resonant charge transfer mechanism.

4.4.4 Perspectives

Work in the future will most likely be dedicated to highly charged ions and low energies. One promising proposal to extend the range of high charged states and low kinetic energies involves the ion deceleration at GSI. In the past, highly charged ions have been produced by electron impact ionization in ion sources of the ECR, EBIS or EBIT type. With these sources bare ions of high-Z elements can only be produced in extremely low quantities, often not sufficient for performing collision experiments. By stripping in a foil at high energies, bare ions up to U$^{92+}$ can be produced. The possibility to subsequently decelerate the projectiles to produce extremely highly charged ions at very low kinetic energies is being explored at GSI. This will open up new horizons for the study of ion–surface interactions at well-defined energies with unprecedented high perturbation parameters $q/v_p > 10^3$. Highly charged ions approaching the surface in this parameter regime expose the surface to a field strength that is comparable to lasers with very high intensities in excess of $10^{17}$ W/cm² over periods of several femtoseconds. As this is a d.c. rather than an a.c. field, the nonlinear response of the surface should be dramatic.
Bibliography


5. Material Modifications

5.1 Introduction

This chapter is obviously linked to the preceding one. There, the basic processes governing atomic collisions were analysed; here the interest is focused on the subsequent modifications induced in condensed matter. Radiation is a means, sometimes unique, to induce non-equilibrium states of matter. In this sense, it is an interesting subject of physics by itself. To understand and to control such effects, one clearly needs to take proper account of the primary events studied in Chapter 4. But this is far from sufficient.

The response to these primary events and the permanent modifications that can be induced depend on the nature of the materials. This is already true in the ‘low-velocity’ regime where elastic collisions dominate and the energy is directly transferred to target atoms. The residual modifications induced in this situation depend drastically on the diffusion properties of the implanted species and of the defect formed, on their probability to annihilate or to agglomerate, and on their ability to induce stable or metastable phase transitions. The nature of the materials is even more decisive with regard to the modifications induced in the ‘high-velocity’ regime where inelastic collisions dominate. In this case the ability to induce material modifications is mainly determined by the efficiency and the rapidity of the energy transfer from the target electrons to the target lattice. Very spectacular effects are observed when extremely high densities of energy deposition are reached. In the last few years, impressive progress has been made in the understanding and control of material modifications. Numerical simulations are now performed and are becoming able to test the validity of more empirical but rather general approaches (thermal spikes, Coulomb explosion). Nonlinear and threshold effects as a function of deposited energy density are observed and understood.

As radiation induces non-equilibrium states of matter, new materials can be created with novel properties. In industry, many applications of material irradiation have been developed for the production of micro- and nanomaterials of high technological interest. Moreover, the materials modified by irradiation can be used to study basic problems in many domains of physics; for instance, in solid-state physics important information is obtained on phase transitions of vortex lattices in high $T_c$ superconductors or on interactions between magnetic nanoparticles.

Irradiation concerns all classes of materials, ranging from metals to living cells. The present chapter is thus closely related to Parts 1 and 2 of this report, which are devoted to the impact of nuclear science on energy and on biology (and therapy), respectively. Within the field of energy, it is of prime importance for nuclear industry to know the evolution of nuclear fuel and of matrices for nuclear waste transmutation which are submitted to fission fragments. It is also of interest to use ion accelerators to simulate the radiation damage induced by different radiation fields. In another domain, it is important to simulate the wide spectrum of irradiation to which electronic devices are submitted in space and to predict their behaviour. To conclude, it is necessary to understand irradiation-induced effects both on basic and technological grounds.

Studies in the radiation-damage field started fifty years ago with the advent of nuclear energy. Most of the interest in this early period was devoted to the modification of metallic compounds under neutron irradiation. Some time after, the use of implantation for doping semiconductors extended massively. Finally, the formation of new phases by ion irradiation, ion implantation,
atomic mixing of multilayers and by dynamic mixing during implantation or deposition, constituted an active research area. Implantation induces a correlated damage and consequently the development of such materials studies, using low-energy ion beams, necessarily involves the build-up of a basic understanding of radiation damage processes, particularly those due to the elastic collision effects. On the other hand, the conjunction with significant developments in neighbouring fields (such as the production of metastable alloys by, e.g., laser annealing or mechanical techniques) has led not only to new ideas for tailoring new materials, but also to the introduction of concepts derived from non-equilibrium thermodynamics [1]. The enhanced ability to model a non-equilibrium phase diagram produced by ion irradiation or implantation (the latter adding a source term to the former) is providing predictive power to ion-based techniques. Recent examples have shown how one can, in this way, go so far as to design systems with interesting physical properties that may lead to applications in microelectronics or magnetism [2], [3]. All the research and applications are now mature. Although at present their evolution is largely independent of the nuclear physics research facilities (and will thus not be detailed here). It is only fair to emphasize the crucial role that the latter have played in a recent past.

In the last 15 years, a sizeable scientific community has used facilities shared with nuclear physicists or originally devoted to nuclear science studies. The interest of this community is on the highly excited states of matter induced by swift heavy ion (SHI) high- or low-energy cluster beams and very low velocity highly charged ions (HCl). In this chapter we focus on these specific ion beams.

5.2 Energy deposition

5.2.1 Radiation-induced excited states of matter

The interaction of charged particles with a target can be analysed by considering independently inelastic interactions with target electrons and elastic interactions with screened target nuclei. The former interaction is responsible for the ‘electron stopping’ (dE/dx)e that dominates at high velocities, the latter for the ‘nuclear or atomic stopping’ (dE/dx)n that dominates at low velocities. Ion irradiation can deposit quasi-instantaneously very high energy densities in matter, a feature that is certainly not accessible with other irradiation modes such as, for instance, high-power fast lasers.

Inelastic collisions: electronic excitation

Swift heavy ions lose their energy by electronic excitation and the corresponding stopping can reach very high values: some tens of keV/nm. Half of this energy loss is deposited in a range of a few nanometres around the ion path; the remainder is transported far away by the energetic electrons produced. Monte Carlo simulations have been used to describe this energy deposition but there are no direct measurements in the solid state to verify their predictions. The only approach attempted to validate the numerical predictions was to compare the measured electron emission from thin carbon foils to the electron transport calculations based on a master equation in phase space [4] (see Chapter 4, Section 4.4.1).

High-energy polyatomic beams offer the possibility to reach extreme values of (dE/dx)e. Therefore, they have allowed unknown regions of electronic excitation to be explored. Before fragmentation in matter, a C60 beam, at only 10 MeV, experiences the maximum (dE/dx)e reached with a monoatomic beam; at 20 MeV, (dE/dx)e is 50% higher. Varying the energy and the nature of the cluster, it is possible to explore a large range of (dE/dx)e and in particular the same stopping power values that are accessible with monoatomic beams but in a much lower velocity regime. This high excitation regime is only obtained in the near surface region of the solid since, mainly due to multiple scattering, the trajectories of the individual constituents of the cluster spread out (see Chapter 4, Section 4.4.2).

Low-energy very highly charged ions provided by the new generation Electronic
Cyclotron Resonance sources (ECR) or Electron Beam Ion Sources (EBIS) can extract a large number of electrons from surfaces. As the ion approaches the surface, target electrons are captured in high n levels of the projectile and re-emitted by Auger effect. Consequently, the number of electrons extracted can be higher than the charge state of the projectile [5], [6]. This induces a so-called potential track. In the vicinity of the ion impacts, holes are now created in the electronic band structure of the solid instead of electron-hole pairs generated in the case of high velocity projectiles (see Chapter 4, Section 4.4.3).

Elastic collisions

Low energy ions develop displacement cascades in solids. Within these cascades atoms are expelled from their stable site in the target with a kinetic energy ranging from a few tens of eV to a few hundred eV. Recently [7], an experimental study of the slowing down in crystals of ions in this energy range has been done at the Laue Langevin Institute in Grenoble via a gamma-ray-induced Doppler broadening technique. In this technique one uses two features: i) neutrons penetrate deeply into matter and excite nuclei if they are captured, and ii) this capture leads to a newly formed isotope, which de-excites down to the ground state by a sequence of gamma-ray emissions. The first emitted gamma ray imparts a recoil to the nucleus and the entire atom will start to move inside the crystal. While the atom is moving through the bulk, the nucleus remains excited for some time (the nuclear state lifetime) and then emits a second gamma ray. A high-precision measurement of the second gamma ray’s line-shape provides the Doppler broadening and thus the velocity distribution of the moving isotopes. This distribution is influenced by blocking and channeling effects (see Chapters 3 and 4). Such effects can be simulated and the comparison with measured line-shapes thus provides information on the interatomic potential governing the ion trajectories.

Nonlinear cascades are induced with heavy monoatomic projectiles when the atoms set in motion start to hit other moving atoms. Low energy cluster beams provide new perspectives for studying highly nonlinear dense cascades. The effect induced by a cluster exceeds the sum of the effects produced by the atoms bombarding individually the same targets.

5.3 Relaxation of deposited energy

The structural modifications that follow the energy deposition last typically some picoseconds. Only in organic materials where secondary chemical reactions occur, can the time-scale of the damage process be much longer. In this last case, the dynamics of the relaxation of the deposited energy could be experimentally followed. In the other materials, there are no direct experimental techniques to observe the dynamics of the structural modifications. Only numerical simulation or the use of the ejected particles as messengers of the primary steps of the energy deposition gives some insight on short time processes. This situation contrasts with laser irradiation where pump-probe methods allow short time observations.

The relaxation of the highly excited states of matter can induce permanent structural changes. High (dE/dx)\textsubscript{e} projectiles can create, above a (dE/dx)\textsubscript{e} threshold, a damaged zone all along the particle path generally called latent track. High (dE/dx)\textsubscript{n} projectiles develop displacement cascades collapsing in a small damaged region.

5.3.1 Time-resolved measurements

At high (dE/dx)\textsubscript{e}, the time-resolved measurements are at present restricted to water radiolysis [8], [9]. Electrons are very rapidly (< ps) solvated in polar liquids. The solvated electron reacts with the counterpart cations on a nanosecond to microsecond time-scale. The kinetics greatly depends on the heterogeneity of the energy deposition. The decay of the solvated electron is a severe test of the heterogeneous chemical reactions supposed to occur in the ion tracks [10]. On the nanosecond scale, the energy per pulse delivered by heavy ion accelerators is far below the value offered by electron machines. The highest possible inten-
sity is highly desirable in this domain. The increase of the intensity of the GANIL beams, coupled with an enhancement of the detection sensitivity, recently allowed the decay of the solvated electron to be measured with a time resolution of 1 ns. Testing heterogeneous kinetics in model systems, e.g. water, is crucial because secondary chemical reactions probably determine the high \((dE/dx)_e\) behaviour of the organic matter including biological samples.

### 5.3.2 Numerical simulation

Molecular dynamics (MD) has been used for studying nonlinear displacement cascades. A plastic flow of hot liquid towards the surface is predicted when a cascade is centred 'inside' the sample while, for a cascade developing nearer to the surface, microexplosions occur \[11\].

In the electronic deposition regime, a realistic model must explicitly include the target electron system (\textit{ab initio} simulations). At present, this is only feasible for small atomic clusters in vacuum \[12\]. Bypassing the electron-lattice energy transfer, classical MD calculations are urgently needed to better understand the atomic relaxation of the high-temperature high-pressure tracks induced by SHI or clusters and slow HCI beams. In solid rare gases a comparison of the sputtering predictions — given by MD, classical thermal spikes without mass transport and by solving the Navier–Stokes equations — has been done \[13\]. These studies point out the strong coupling between pressure and temperature and reveal the limits of the often-used classical thermal calculations \[14\].

### 5.3.3 Electron emission

A few femtoseconds after the passage of the fast ion, the energy thermalizes in the electronic system. Measuring the shift and width of Auger lines and convey electron yields, it has been possible to determine the nuclear track potential of polypropylene and the electron temperature of amorphous carbon in the track centre on a time scale of about 10 fs. These data provide the basic input for thermal spike models and illuminate the relevance of Coulomb explosion in metals and polymers \[15\].

#### 5.3.4 Electronic and nuclear sputtering in organic and inorganic materials

Ionic and neutral emissions from surfaces represent a signature of the atomic motion short times after the energy deposition \[16\]. Because it is easier to detect, ionic emission was mostly studied, allowing measurements of angular and energy distributions as well as identification of cluster emission. But the ionic fraction represents a very minor part \((< 1\%)\) of the total emission. Most of the measurements on neutrals are on total yields. Data on angular distributions and energy distributions are limited. The detection of the neutral clusters is still a challenge. Particle emission (electron, ions and neutrals) is very sensitive to the surface quality (roughness and adsorbed impurities). At present, ultrahigh vacuum chambers fitted with surface characterization set-ups and installed on SHI, slow HCI and cluster beam lines are too scarce.

In polymers exposed to SHI, the angular and velocity distributions of ions coming from extensive fragmentation-rearrangement of the original molecular structure or of ions closer to the original molecular structure are markedly different \[17\]. These observations nicely show how sputtering can reveal the core and peripheral chemical reactions and the related atom movements occurring in a heavy ion track.

Sputtering was the first evidence that very low HCI causes severe damage to insulating surfaces (see Chapter 4, Section 4.4.3). Figure 5.1 shows that high yields are observed and, as expected, that the yield is clearly connected to the ion charge state \[18\].

High-energy cluster beams can have extremely high \((dE/dx)_e\) and consequently induce dramatic effects \[19\]. For organic films, giant craters corresponding to an emission of \(10^7\) mass units have been observed \[20\]. These beams also produce a significant number of large clusters. In particular, for targets formed of large organic molecules the emission of intact molecules is highly enhanced, with respect to what is obtained with swift monoatomic projectiles.

For gold samples, the nonlinear sputtering effects of low energy cluster beams have
been analysed [21]. These particles deposit their energy in elastic collisions. Contrary to monoatomic beams, the sputtering yield with these cluster beams is maximum at a velocity substantially lower than the one corresponding to the maximum of nuclear stopping power. A large nonlinear sputtering enhancement is observed between cluster and atomic projectiles. When the cluster projectiles have more than three atoms, a square dependence of the sputtering yield is found as a function of the number of constituents.

5.4 Permanent effects induced in solids by strong electronic or atomic perturbations

The application of sophisticated characterization methods, like high-resolution transmission microscopy, near-field microscopy, defect spectroscopy, and X-ray diffraction (sometimes online) resulted in a significant progress in collecting experimental data about track formation and track structures in solids. The mechanisms by which the electronic excitation energy is converted into atomic motion and, finally, into stable structural changes are still under debate. There is now growing evidence that in the late phase (\(\geq 0.5\ \text{ps}\)) of track formation the matter around the trajectory of a fast heavy ion can be conceived as a high-pressure, high-temperature spike.

The attention paid to the effects of SHI on organic materials is rising. Probably, the interest in biological effects and especially in heavy-ion radiotherapy partially explains this trend.

5.4.1 Plastic instability of amorphous materials

The hammering effect of track-generating ions in amorphous materials was one of the most unexpected and remarkable effects of high \((dE/dx)_e\) irradiation. This effect is now much better understood. In these solids, a particle track represents essentially a thermo-elastic inclusion [22], [23]. The consideration of mechanical equilibrium [23] of a fluid track in a solid matrix results in a constitutive equation, which describes the mechanical behaviour of amorphous solids during heavy ion bombardment. Surface deformations and ripples appear at the sample surfaces as a consequence of the bulk deformations [24].

5.4.2 Phase stability under radiation

The investigation of non-equilibrium phases produced by track-generating ions is far from systematic. In the past, most attention was devoted to amorphization, which has often been interpreted as melting and freezing of the hot track matter [14].

More recently, the effects of extreme \((dE/dx)_e\) induced by high-energy \(C_{60}\) cluster beams have been investigated. For the first time, the amorphization of some extremely resistant materials such as sapphire [25] and silicon [26], [27] has been induced. The damage does not only depend on \((dE/dx)_e\); at given \((dE/dx)_e\), it decreases when increasing the projectile velocity. A very complete analysis of this ‘velocity effect’ has been performed on YIG oxide [28].

Investigations of phase transformations between two or more crystalline phases are rare [29], [30], [31]. In yttria the \((dE/dx)_e\) cubic to monoclinic phase transformation becomes easier when the mean crystallite size of the target decreases [32]. The design of high-power fuels, for research reactors, benefits directly from a development of criteria for phase stability under irradiation.
With bilayer or multilayer targets, studies of phase stability and phase formation benefit from usually planar interfaces and, additionally, allow the investigation of diffusional processes (mixing). Some attractive experimental approaches such as multilayers with $^{57}$Fe isotope enriched layers associated with a detection of the intermixing by Mössbauer spectroscopy have great potential [33]. To progress in the understanding of the relaxation paths of highly excited materials, tailored nanostructured material should play a crucial role in the near future.

In addition to high-resolution electron transmission microscopy, small- and wide-angle X-ray scattering should be applied more for the characterization of the phases formed and their interfaces. An on-line X-ray facility with a large position-sensitive counter is in operation at GANIL (Caen). A four-circle diffractometer will be installed at the cyclotron of the Ionenstrahlabor of the HMI (Berlin) this year. Neither on-line, small-angle scattering apparatus nor grazing-angle X-ray diffraction equipment for multilayer studies exists at any of the big European ion accelerators. Cluster beams have largely contributed to the understanding of the track generation. Larger cluster ion accelerators than available at present are considered necessary.

5.4.3 Surface modifications

Surface modifications are related to sputtering and to track formation. Near-field microscopes now allow the imaging of the topographic effects of single ion impacts [34]. Depending on the materials and irradiation conditions, craters or hillocks are observed. Protuberances of various shapes, located in the vicinity of the craters, are also seen. Recently transmission electron microscopy in the topographic contrasting mode has been successfully used for analysing the surface irradiation effects [35].

Surface modifications of mica were extensively studied by atomic force microscopy [36]. Figure 5.2 shows the radii of the surface tracks versus the restricted energy loss of monoatomic and polyatomic projectile. In the bulk, the restricted energy loss re-scales the results by taking into account the ‘velocity effect’. This rescaling can be obtained for monoatomic ions of different velocities but not for cluster irradiations that give smaller radii. This latter discrepancy could be due to an overestimation of the cluster energy loss arising from the difference in the incident charge state of atoms and clusters. This result stresses the importance of charge equilibration (cf. ETACHA-Code, see Chapter 4, Section 4.4.1) when looking at near surface effects.

Figure 5.2: Track diameter measured by atomic force microscopy [36] as a function of the reduced electronic stopping power calculated with a cut-off energy of $\delta$ electrons of 200 eV. The open symbols correspond to irradiations with monoatomic ions of different velocities and the black symbols to the cluster irradiations (squares: aluminium clusters, circles: carbon clusters).

5.4.4 Organic materials

Polymers are one of the simplest organic solids. Besides their use in the applied field, they have been much studied as model systems of organic matter. Tracks in polymers are complex. Small-angle X-rays and neutron scattering measurements visualize the ion tracks as a cylinder of density smaller than the bulk [37]. Infrared spectroscopy provides information on the chemical modifications induced by radiation [37], [38]. This technique has revealed that high $(dE/dx)_e$ irradiation induces specific defects. The gas release greatly contributes to the track formation. The gas-release analysis shows that
the stability of some specific molecules in the chemically reactive core of the tracks largely influences the production yields and consequently the track formation [39].

Closer to life sciences, analysis of the degradation products at high \((dE/dx)_e\) of the elemental constituents of DNA has been done by nuclear magnetic resonance [40]. As for simple polymers, high \((dE/dx)_e\) irradiation of nucleosides and dinucleosides induces specific modifications. Irradiation of plasmid DNA allows the observation of the \((dE/dx)_e\) incidence on the single- and double-strand breaks which are supposed to be determinant in radiobiological effects. In addition to standard electrophoresis measurements, transmission electron microscopy was recently used and has allowed the first detection of locally multiply-damaged sites formed in DNA after exposure to a heavy-ion beam [41].

5.5 Use of induced effects

5.5.1 Novel radiation-induced properties of materials

Radiation-induced structural changes are used to obtain materials with interesting new properties. Slow HCl could be of interest for surface etching on a nanometre scale. Cluster beams are envisaged for the emission, in the gas phase, of intact large organic molecules. However, at present, the latent tracks induced by SHI represent certainly the most important modification in terms of applications.

The production of nanopores by chemical etching of ion tracks has turned over to industry. Nanopores are used in many fields, e.g. in medicine for a well-defined drug release, in electronic-circuit housings in motor cars for water-repellent venting holes, and as semi-permeable membranes in miniature fuel cells. At present, these applications are limited to nanopores in polymers but materials withstanding higher temperatures should be developed. The preparation of asymmetric membranes by electro-stopping [42] and of responsive membranes by grafting open new perspectives [43].

Replica or template techniques allow the production of cylindrical objects of micrometre–nanometre size on a wide variety of materials [44], [45]. Utilizing the field effect, electrically conducting objects could serve as electron emitters for displays [46]. Recently developed semiconductor growth modes can be exploited to fabricate nanometre-sized semiconductor devices in chemically etched ion tracks. Another possibility is track doping for tailoring the properties of the track [47]. Clearly, the trend is to decrease, as much as possible, the diameter of the cylindrical objects (Fig. 5.3) and to identify the novel properties related to the nanoscopic dimensions of the materials. Nowadays, pore sizes of 15 nm are realistic [45].

Very recently, it has been shown [48] that above a rather low threshold in deposited inelastic energy density (about 5 keV/nm), ion irradiation of glasses could induce the controlled nucleation of metallic clusters. One then obtains well-adapted materials for studying individual optical and magnetic properties of nanoclusters with interesting applications in optical switching or filtering.

5.5.2 Basic studies with radiation-induced structured materials

The radiation-induced material modification is a tool for basic studies in other branches of physics. Once more, the latent tracks induced by SHI are the defects that have found a marked interest for basic studies.
Columnar defects induced by fast heavy ions are ideal pinning centres for flux lines in high-\(T_c\) superconductors (HTSC). There, latent tracks have the same geometry as the flux-vortices and same radius as the core of a vortex. This fact has prompted a considerable number of studies and hundreds of publications. Sophisticated theories on pinning and melting of the flux line lattice could be tested by the generation of splayed particle tracks \cite{49,50}. Induced latent tracks in metallic compounds produce magnetic nanostructures and have allowed unique studies on nanomagnetism \cite{51}.

Some of these experiments would greatly benefit from regular track lattices. The feasibility of the latter is still a time-consuming task. An ion microprobe steering individual ions to pre-set locations is only available at the UNILAC of GSI (Darmstadt) \cite{52}.

5.6 Materials under irradiation

Materials are subjected to radiation. It is therefore necessary to understand and predict the induced effects. Very contemporary and important problems exist with regard to radioactive waste management and to present and future reactors. In order to deal with these problems, it is now crucial to reinforce the community of physicists expert in problems related to irradiation of materials and thus to train young scientists in this domain.

The experimental study of the materials submitted to irradiation is performed in two ways. One is to reproduce the ‘real’ irradiation conditions in the laboratory; the other, which offers substantial practical advantages, is to perform a simulation with beams different from the real exposure.

5.6.1 ‘Real’ irradiation conditions

The irradiation conditions, in terms of ionic species and energies, can be relatively close to the real exposure conditions. This is the case for the study of the fission fragment damage and for the study of SHI effects in electronic components. The flux effect is probably small and acceleration of the irradiation conditions is not very problematic.

Today the problem of radiation-induced malfunctions in integrated systems is very real \cite{52,53}. It is not strictly limited to exposures in space, but malfunctions caused in satellite electronics by a single ion hit are probably the major issue. These bit-flips were named single event upsets (SEU). Cosmic radiation presents a very large spectrum of ions and energies. It is a real challenge to predict the in-flight behaviour of a component starting from a necessary limited number of measurements with accelerator-delivered beams. An in-depth understanding of the effects of \((dE/dx)\), and of the velocity effect is necessary. Microbeams give a valuable contribution to the electronic component studies \cite{54}. They permit the spatial localization of the ion impacts that induce a SEU (Fig. 5.4) \cite{52} and subsequently the sensitive zones of the circuit (see Chapter 6, Section 6.4).

![Figure 5.4: Upper part: electron microscopy image of an array of 24 elementary storage cells. Lower part: radiation sensitive sites within a chip area similar to the one of the upper part of the figure. Every point is due to an ion hit leading to an upset. The hits themselves are uniformly distributed \cite{52}.](image-url)
actinides will also be submitted to fission fragments. Track generation by fission fragments could create unaffordable morphological changes of the target. In spite of the numerous studies of latent track formation (cf. Section 5.4.2), it is still difficult to foresee the behaviour of a given material. Moreover, all the basic studies have been done at room temperature and low fluence, i.e. very far from the working conditions in a nuclear reactor. Realistic irradiation must be performed. For that, a new beam line allowing high flux irradiation with heavy ions in the 0.5–1 MeV/A range is under construction at GANIL.

5.6.2 Radiation-damage simulation

Shortening the irradiation duration and lowering the irradiation cost are the main reasons for departing from the actual irradiation conditions.

When, in the real situation, the irradiation times are very long, an artificial acceleration of the irradiation is unavoidable. In several cases, the material stability is not only determined by the fluence or the absorbed dose, but also depends on the flux or dose rate. Consequently, the understanding of the flux or dose rate effects is one of the challenges for the prediction of the stability of materials under irradiation.

The use of small ion accelerators instead of neutron irradiation or very high energy proton irradiation allows a greatly reduced irradiation time, generates cost benefits and permits a strong decrease of the radioactivity of the samples. The decrease of the activity of the irradiated samples permits a much better characterization of the damage at a much lower cost.

The simulation by different particles will never be sufficient to ensure the correct behaviour of a material submitted to a very different radiation field. But it is of great help in selecting promising materials and in understanding the effect of the different parameters such as flux, fluence and temperature.

Ion simulation of fast neutron damage

Ion irradiation is a well-known way to simulate fast neutron damage to materials. The irradiation conditions must fit the primary knock-on spectrum generated by the neutrons of the reactor. One problem is that the thickness of the irradiated layer is generally much smaller than that of the materials irradiated under real conditions. This is a true limitation since intergranular processes can, to a great extent, control the macroscopic mechanical behaviour. The irradiation over a thickness greater than the grain size implies the use of relatively high-energy accelerators. The other difficulty is that nuclear reactions induce significant doping of the target by H and He atoms. The simultaneous implantation of these species is often considered necessary. This leads to the use of dual or triple ion beam facilities, the availability of which is one of the crucial issues of radiation-damage simulation.

Ion simulation of material damage in ADS

The new subcritical reactor concepts (ADS for accelerator driven systems) raise new and severe radiation damage problems. Very high-energy protons and neutrons irradiate the structural materials. The atomic displacements are indirectly generated by the spallation products and the nuclear reactions produce a large spectrum of foreigner atoms with very significant concentrations. Several European programmes are dedicated to the material problems of subcritical reactors. This emphasizes the importance of the effort needed in this direction.

Simulation of radiation-induced corrosion

The contact of metals with water in classical reactors and with liquid metals in ADS induces a detrimental corrosion. The irradiation of a liquid–solid interface requires a large penetration depth and the use of high-energy accelerators seems essential.
5. Material Modifications

Bibliography


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6. Material Analysis

6.1 Introduction

Materials manufacture and characterization are today top branches of activity inherent to the
development of any new technology. The development of modern characterization using nuclear
techniques is linked to the study and understanding of basic phenomena, often undertaken around large-scale facilities. The number of applications of such know-how to material science has been continuously growing. This is particularly evident in the case of neutron scattering and synchrotron radiation applications, which have developed to the status of ‘standard techniques’, providing, among others, information on magnetism, crystallographic and molecular structures. Today, the number of subjects and user requests of beam time for material science purposes justifies alone the development of new facilities with increasing power and improved beam quality, i.e. bunching, polarization and microbeams.

On the other hand, the present needs for increased integration in electronics, photonics, nanostructured materials and microrobotics provide fertile ground for the application and development of nuclear techniques, which provide local information, scaling from the neighbourhood of atoms to micrometres. Phenomena and technologies which are highly dependent on the submicron nature of doping, thin films, micro- and nanostructures have found in techniques like muon spin resonance (μSR), ion beam microprobes and new radioactive ion beams the ideal source of microscopic information.

Four subsections are developed which emphasize the perspectives opened by recent technical developments, new methods and applications [1]: neutron scattering study of condensed matter; exotic beams – positron and μSR methods–; ion beam techniques; radioactive ion beams and techniques for solid-state research. Among the four techniques presented in this chapter, the last two, which make use of ion beams, benefit particularly from the knowledge on atomic collisions and material-induced modifications that are the subjects of Chapters 4 and 5.

6.2 Neutron scattering study of condensed matter

A very broad community of some 7000 people worldwide (5000 in Europe) uses neutron scattering for studies ranging from mechanical engineering to life sciences. A number of unique properties of slow neutron radiation make it an indispensable tool in the study of materials. These include particularly neutron sensitivity to light elements such as hydrogen and the direct interaction with magnetic samples via its magnetic moment. In addition, due to the value of the neutron mass, the wavelength of thermal neutrons — with energy in the 5–80 meV range comparable to the energies associated with atomic motion in condensed matter — happens to coincide with the length scale of interatomic distances (a few Å). Therefore, neutrons provide the unique capability of exploring both atomic structure and atomic motion.

Relevant examples of applications can be presented in the fast-growing field of soft matter research. In such materials the macroscopic properties are strongly determined by the microscopic dynamic behaviour on the molecular level. For example, in polymers the intermingled chain molecules mutually limit each other’s motional freedom, leading to the reptation process that has been the subject of fundamental theoretical studies by de Gennes. Recent Neutron Spin Echo experiments have unquestionably evidenced and characterized this type of motion [2], after all other experimental attempts remained inconclusive.
Neutrons have constantly played a major role in the study of magnetism in condensed matter. The last decade has seen strong activity and important findings concerning high-$T_c$ superconductors and superconductivity. In this field a recent breakthrough was the observation of vortex lattice melting in superconducting Nb [3]. In this type of superconductor, the magnetic field penetrates in channels called vortices whose core has a normal conductance. At low temperature, the interaction between these vortices results in the formation of a periodic ordered lattice. Neutron small-angle scattering diffraction studies, performed as a function of temperature, evidence supercooling and superheating effects and thus demonstrate that the vortex lattice undergoes a first-order phase transition.

The sensitivity of neutrons to light elements, such as oxygen in high-$T_c$ superconductors or hydrogen in biological matter, is in stark contrast to X-rays where the lattice of the heaviest elements of a compound mainly determines the diffraction pattern, and little or no information can be found on the position of the lightest elements. This gives an important complementary role to neutron diffraction, particularly useful in the study of biological samples. After the backbone of the structure formed by the heavier elements, the position of hydrogen atoms associated, for instance, with hydration water, is elucidated by neutron scattering. This position information is of particular importance for studying biological functions [4].

Traditionally, the preferred neutron sources used for producing beams of slow neutrons for neutron scattering instruments are research reactors using enriched uranium fuel for fission. Most reactors have switched, or are in the process of switching, to low enriched uranium, in the spirit of the nuclear nonproliferation treaty, but those with the highest neutron flux still use high enriched uranium. Higher density uranium–molybdenum fuel should be available in a decade or so, which will open up the possibility for the highest flux research reactors to also switch to low enriched uranium fuel without substantial loss in their neutron flux.

![Figure 6.1](image.png)

**Figure 6.1:** The figure represents a sequence of monochromatic pulses shaped by mechanical chopper devices (1...6, illustrated by horizontal lines for ‘closed’ positions, interrupted by ‘open’ periods). In this distance–time diagram the straight lines, whose slope corresponds to the neutron velocity, illustrate neutron trajectories. On a pulsed source, neutrons originating from a pulse will arrive at the sample over the whole period between pulses.

The ISIS (Didcot, UK) and SNQ (Villingen, Switzerland) neutron centres use spallation for neutron production. In this more recent technique fast neutrons in the 2–10 MeV energy range are produced as the result of a number of secondary nuclear processes following the irradiation of a heavy-metal target by protons in the 800 MeV energy range. The fast neutrons are moderated to the typically 1–500 meV energies required, with very few exceptions, for the study of condensed matter by scattering experiments. Spallation offers two principal advantages over fission: the heat deposited per extractable neutron is about 30 MeV as opposed to 190 MeV, and a spallation source can easily operate in pulses, which enhances the efficiency of using the available neutrons produced in the source. The principle is illustrated in Fig. 6.1 [5]. In time-of-flight neutron scattering spectroscopy, pulses of neutrons arrive at the sample at a frequency determined by the time needed to analyse the velocity distribution of the scattered neutrons, typically 2–20 ms. In order to deliver the series of pulses illustrated in the figure a continuous reactor source will produce neutrons all the time, most of which will be eliminated by the neutron-absorbing mechanical chopper system. In contrast, on a pulsed spallation source,
neutrons will emerge from the moderators only for a fraction of the total time, and neutrons eminating from the same pulse with different velocities (different slopes in the time-distance diagram in the figure) will arrive at the sample at different times, with a repetition rate as required by the spectrometer. As the desirable duty factor is around 1–5%, the neutron flux released by the source during the short time can be by orders of magnitude larger than the time average flux, relevant for the average power. For example, at 5 MW proton beam power the time average cold neutron flux of coupled liquid-H$_2$ moderators achieves the steady flux of the most powerful reactor in existence. Its instantaneous ‘peak’ flux during the pulses, which matters mainly for most experiments, can achieve 30–60 times this time-average flux value [6]. In the next 5–10 years, different projects in Europe, USA and Japan aim to construct very powerful spallation neutron sources, with effective neutron fluxes much greater than those of the most powerful reactor.

Progress in neutron scattering experimental techniques accounts for most of the spectacular gain in performance and sensitivity in neutron scattering work over the past decades. Most recently the use of ‘supermirror’ coatings in neutron guides is in the process of revolutionizing neutron beam extraction and delivery, producing five- to seven-fold beam intensity gains on the sample. The combined effect of improved source performance, improved neutron optical beam delivery and improved detector solid-angle coverage will enhance data collection rates in the next decade by a very substantial factor, between 10 and 1000 times, in all types of neutron scattering applications. Such a tremendous gain in sensitivity and the fact that spallation sources in the range of several MW proton beam power have now become technically feasible will open up completely new fields of application in neutron scattering research.

6.3 Exotic beams: positron and muon spin resonance methods

Positron spectroscopy techniques are powerful tools with a large spectrum of applications, providing microscopic information on the electronic, crystallographic and chemical properties of materials. Recently, progress has been made mainly in positron beam techniques. The availability of a MeV positron beam at the Max Planck Institute für Metallforschung (Stuttgart, Germany) allowed new experiments with the age–momentum correlation method [7, 8], which correlates positron and positronium lifetime, and Doppler-broadening spectroscopy. This allows in particular, the characterization of transient processes in matter such as thermalization, bubble formation in liquids, and slow trapping of positronium in polymers [9]. Low-energy positron beams, typically up to 30 keV are used for studying depth profiles of positron trapping defects such as vacancies, dislocations and voids, as well as layered structures. The most recent development is the pulsed low-energy positron beam which was developed at the Universität des Bundeswehr (Munich, Germany) and which allows depth profiling of positron lifetimes. At least three more pulsed low-energy positron beams are under construction in Europe: in Ghent, Gothenburg and Helsinki. An overall review of today’s research and development on new techniques can be found in Refs. [10, 11]

Muons and muonium spin resonance ($\mu$SR) methods are an outcome of particle physics, and are now well established as universally applicable microscopic magnetic probes in solid-state physics, chemistry and materials science. Nowadays, muons can be implanted in very low number and at very low energy in any material without producing radiation damage, under a large variety of external conditions (temperature, pressure, magnetic or electrical fields, etc.). Polarized muons are an important probe of static and dynamic magnetic properties of materials. Their large magnetic moment (three times that of protons) and mean lifetime of 2.2 $\mu$s make local magnetic fields and field distributions accessible in the range of microtesla to several tesla, the time-scales for dynamic properties ranging from pico- to milliseconds. As a ‘light isotope’ of the proton, the positive muon can form the hydrogen-like atom muonium, which may replace hydrogen in insulators, semiconductors and organic materials, thus
providing a very sensitive spin label.

Research applications of muon-spin rotation, relaxation and resonance (μSR) include the study of magnetic, superconducting, metallic, semiconducting, ion-conducting, amorphous and molecular materials. The advent of low-energy muon beams extends the applications to depth-resolved studies of magnetic properties (on a nanometre scale) of thin films, multilayers, nanometre-sized clusters, and near-surface regions.

The advantage of continuous-beam μSR is its high (sub-nanosecond) timing resolution allowing the study of fast dynamics and high magnetic fields, whereas a pulsed beam offers the ability to determine very weak atomic magnetism and is well suited for examining materials in pulsed environments such as magnetic and electric fields, laser or radio-frequency radiation. Two unique special facilities have been developed at the continuous-beam μSR facility of the Paul Scherrer Institute (Villigen, Switzerland):

- A low-energy muon beam (LEMB) allowing the implantation of muons at very small and controllable depths, a few to a few hundred nanometres below the surface of a sample. A wealth of new applications become possible by extending the studies to very thin films, multilayered structures, surface regions and by measuring material properties as a function of the implantation depth on a nanometre scale. An example application of LEMB is shown in Fig. 6.2 where the penetration of a magnetic field into a superconductor in the Meissner state is studied [12]. The measurements were performed on a 700 nm thick, c-axis oriented and epitaxial YBa$_2$Cu$_3$O$_7$ film. A vertical magnetic field of 10 mT was applied after cooling the sample to 20 K so that no flux lines were trapped inside and the field below the sample surface was due to the finite value of the penetration depth. The depth of the muon implantation was controlled between 20 and 150 nm by tuning the incoming muon energy from 3 keV to 30 keV. The depth-dependent field profile $B(z)$ within such a semi-infinite superconducting slab follows the well-known London exponential decay law.

![Figure 6.2: Magnetic field, $B(z)$, measured by muons as a function of implantation depth in an YBa$_2$Cu$_3$O$_7$ film, for different sample temperatures. The solid lines represent exponential fits, with the penetration depth $\lambda$ as the only free parameter [12].](image)

- The extraction of one muon at a time from a continuous beam (Muons on Request) reduces background to the level of a pulsed muon source while keeping nanosecond time resolution. This provides unique sensitivity to small magnetic field differences and extends the measurable characteristic times into the millisecond range.

Despite the increasing user demand, there is at present no dedicated facility for μSR methods alone. Production facilities optimized for μSR would provide at least one order of magnitude improvement in data collection time, flexibility in beam arrangements and availability to simultaneous users that would allow routine materials characterization.

6.4 Ion beam techniques: high depth and lateral resolution for materials analysis and microfabrication

The tremendous progress in the computer, communications, imaging and micro-machining fields can largely be linked to the art of creating ever-finer structures in a variety of materials. After reaching the boundaries of photon and electron lithography, finely focused ion beams are expected to play an increasingly important role in the next generations of integrated circuits, where geometric features in the
range of 10–100 nm are required to satisfy the needs of gigascale integration. In mesoscopic systems, i.e. structures between the single atom with discretely spaced electronic energy levels and bulk material with a delocalized electronic energy band structure, the band gap may be tuned by the geometrical design of nanostructures rather than by impurity doping. High depth and spatial resolution ion beam analysis (IBA) techniques are contributing to this evolution, with hundreds of IBA laboratories around the world closely involved with industrial and basic research.

Important recent developments have also been made in IBA data analysis. The development of new fitting procedures using computer techniques such as simulated annealing [13], Bayesian inference with the Markov chain Monte Carlo algorithm [14] and the application of neural networks to Rutherford backscattering spectrometry (RBS) data [15], has drastically reduced analysis time through automation. This opens the way to implement on-line RBS data analysis, particularly relevant for industrial applications.

Very high depth resolution IBA is finding increasing use in investigations of ultra-thin films, with particular interest for thin (1 to 5 nm) dielectrics on semiconductors for MOSFET technology. Several methods are used — narrow nuclear resonance profiling, RBS with time of flight, magnetic or electrostatic detectors — to provide high energy resolution. Another method is high energy resolution elastic recoil detection, using heavy incident ions such as iodine at energies of some tens of MeV [16]. In contrast to sputter-based profiling methods such as secondary ion mass spectrometry, IBA can have nanometre depth resolution from the surface, without being destructive. IBA is a quantitative technique and in addition to elemental sensitivity also offers isotopic discrimination, which is exploited in stable isotopic tracing studies. An example of such sub-nanometre resolution near the surface is illustrated in Fig. 6.3 where narrow nuclear resonance profiling with $^{18}\text{O}(p,\alpha)^{15}\text{N}$ (full width at half-maximum ~100 eV) is used to perform isotopic exchange studies of $^{18}\text{O}$ in the outer few nanometres of a $\text{SiO}_2$ film [17]. The incident beam energy is scanned around the resonance energy to obtain the excitation curve, which is an image of the $^{18}\text{O}$ concentration profile, yielding information on growth mechanisms of ultra-thin $\text{SiO}_2$ MOSFET gate dielectrics. The addition of small amounts of nitrogen at the $\text{SiO}_2$/Si interface leads to substantial improved electrical properties of ultra-thin $\text{SiO}_2$ films. Such systems are also studied, with very high depth resolution, by medium energy ion scattering [18] in which ~100 keV protons are elastically scattered from the sample and their energy spectra recorded with high-energy resolution. Concentration profiling of ultra-thin oxyxnitride films on silicon with sub-nanometre depth resolution has also been obtained by high energy-resolution RBS and elastic recoil [19]. In fundamental condensed matter physics studies, electrostatic energy analysis of 100 keV protons, in channelling–blocking geometry, has been applied to achieve sub-nanometre resolution and to study the surface relaxation (variation of the interplanar spacing) and thermal vibrations of each of the outer few atomic layers of a copper single crystal [23]. Atomic monolayer resolution has also been demonstrated for RBS on single-

Figure 6.3: Excitation curves of the nuclear reaction $^{18}\text{O}(p, \alpha)^{15}\text{N}$ around the resonance energy $E_R = 151$ keV for a silicon wafer oxidized (a) in $^{18}\text{O}_2$ for 3 h at 1000 °C, followed by $^{18}\text{O}_2$ at 930 °C for 5 h; (b) as in (a) followed by oxidation in $^{18}\text{O}_2$ at 930 °C for 2 h. The insets shown the related $^{18}\text{O}$ concentration profile [17].
crystal PbTe(001) with 300 keV $^4$He incident beam and a purpose-built magnetic spectrometer [21] at a grazing exit angle of 2.6°. It is shown that the first and second Pb layers may be distinguished in this case.

The technology underlying the ability to focus MeV ion beams to spot sizes between 100 nm and 1 μm, using nuclear microprobes, has also continually evolved, opening up new applications for the characterization of crystallographic and electronic properties of materials and increasing understanding of ion–solid interactions [22]. The nuclear microprobe operates similarly to a scanning electron microscope: a focused beam of charged particles is swept over the sample surface to produce spatially resolved images using an analytical signal produced by the particle–solid interactions. While it is relatively difficult to focus MeV ions to small spots, the beneficial side-effect is that the beam tends to stay focused through tens of microns in the sample owing to the large ion mass. Strongly-focusing magnetic quadrupole lens systems can now focus MeV ions with a focal length of 10–20 cm, giving a demagnification of 10–100, and hence a small focused beam spot size. The best resolutions of 100–200 nm with a small current, and 300–400 nm with higher currents for conventional IBA techniques, have been achieved by the Singapore group [23] and, recently, in Europe by the Leipzig group [24]. There are now some 50 to 60 microprobe groups in the world, of which about half are in Europe. A review of IBA microprobe applications in biomedicine, geology and archaeology, and the microprobe hardware may be found in [25, 26, 27, 28].

The most promising recent developments relate to experiments with low-current ion beams (in the ~1 fA range), in which one exploits the penetration in matter of each single ion. For material analysis, such experiments are not affected by beam-induced damage, which is sometimes the case when studying, as in conventional IBA, events with relatively low cross sections. Also, working with low currents allows one to limit the beam emittance and thus to produce parallel microbeams that can be used for the study of sample defects and crystallographic structure, using channelling. Low-current nuclear microprobes are used in materials science for a wide range of applications. Many of such recent applications are reviewed in Ref. [29]. In the text below, we refer to some of the most promising.

Applications of ion beam induced charge (IBIC) microscopy

IBIC microscopy has become a well-used microprobe technique in recent years because it gives a method of analysing working integrated circuits, radiation detectors, solar cells, etc., through any thick surface layers. The resultant IBIC images can be interpreted to give information on the physical and electronic properties, such as layer thickness, the distribution of the underlying pn junctions, mask misalignment errors, radiation hardness and upset sensitivity. One example from Ref. [30] is shown in Fig. 6.4 where IBIC and ion beam induced luminescence (IBIL) images of polycrystalline diamond detectors are shown. These detectors, elaborated by chemical vapour deposition, are designed as alpha-particle and UV sensors. They were constructed with a co-planar electrode structure, fabricated only on the growth side of the diamond film. IBIC results show the different efficiencies of individual 10 to 20 μm wide grains in facilitating charge transport at different locations away from the surface metallization. IBIL results show that some of the large grains, which exhibit poor charge collection, give a high luminescence yield, whereas other regions exhibit both low IBIC and IBIL signals. These results directly show that the charge collection efficiency in the inter-electrode region is limited both by the size of the diamond crystallites and by the high luminescence yield of certain grains.

Measurement of small lattice strains using ‘beam rocking’

Measurements that use the ion channelling process in conjunction with a microprobe, have recently been developed for the analysis of small strains from micron-size areas of a crystal. A focused MeV ion beam is rocked in angle about a stationary point on the crystal surface. When the lattice strain varies along the surface, the direction associated with the best channelling
effect is not the same at each surface point. The measurement of these small angular shifts permits us to quantify small lattice strains across uniform composition and graded silicon–germanium layers.

Imaging crystallographic defects using transmission channelling

Defects within the crystal lattice disrupt the regular atomic arrangement and so affect the ion channelling process. This allows the production of images of defects using a scanned, focused MeV ion beam, which is transmitted through a crystal 50 μm thick.

Maskless patterning and fabrication of high aspect ratio features

The use of focused MeV ions using a microprobe to produce microstructures in resist materials such as polymethylmethacrylate, mica or glass is an important step in the production of microelectromechanical components and other microstructures. MeV ions have the ability to produce features such as cogwheels, gears and turbines which are tens of microns in diameter in resist thickness up to one hundred microns, due to the high penetrating power and low lateral scattering. Curved surfaces, buried, angled features and multilevel structures can also be produced.

At the time writing this report a superconducting multipole lens has gone into operation at the Munich tandem accelerator [31], which focuses ions of three times greater rigidity than standard microprobes to a submicron beam spot at large beam currents (> 10 pA). A prominent improvement is gained in 3D-hydrogen microscopy utilizing proton–proton scattering of 10–20 MeV protons, which can now be applied to any micro-structured sample with simple preparation schemes. Ten times better position resolution and many orders of magnitude improved sensitivity are obtained compared to any other nondestructive technique for hydrogen microscopy.

6.5 Radioactive ion beams and techniques for solid-state research

Sophisticated nuclear techniques, which were developed for detecting particle radiation and to study the interaction of nuclear moments with external electromagnetic fields, were introduced in solid-state research in the late 1960s. However, the limited number of adequate probe isotopes, particularly in off-line experiments, has restricted their acceptance by the solid-state research community and their applications to a small number of subjects. The on-line production of radioactive isotopes is the technical approach that best fills this gap. Nowadays, mainly due to the needs of nuclear physics research, several radioactive facilities have been built around the world [32] that can be used for nuclear solid-state physics research. Europe has strong traditions in the field, with radioactive isotope facilities running daily which offer specific complementary features [33]. So far, due to the high diversity, elemental-isotopic purity and high yields of the radioactive beams, the world’s best laboratory is the ISOLDE facility at CERN in Geneva [34]. Currently, the scope of studies there covers a vast range of techniques and subjects in metals, semiconductors, oxides with electronic correlations (superconductors, manganites, etc.) and surfaces and interfaces.

Solid-state nuclear techniques can be ordered in three groups.

‘Hyperfine interaction’ techniques

These rely on specific properties of excited nuclei, and are extremely sensitive at the nanometre scale to the material’s local fields via their interactions with the nuclear moments [1]. Applications of Mössbauer effect, perturbed angular correlations, nuclear orientation and the β-nuclear magnetic resonance have been the subject of many proposals and publications within the last few years which are reviewed in Ref. [35].

‘Emission channelling’ techniques

These make use of the fact that charged particles (α, β⁺⁻, c.e.) emitted from a very low concentration of radioactive isotopes in single
crystals experience channelling or blocking effects along low-index crystal directions [36]. This leads to an anisotropic particle emission yield from the crystal surface that depends in a characteristic way on the lattice sites occupied by the emitter atoms. Recently, a new 2D Si pad detector system has been developed at CERN within the scope of high-energy physics experiments [37], and has been adapted for use in electron emission channelling experiments [38]. Since then, the number of available probe nuclei has increased tremendously, being extended to all relevant element groups in the periodic table. Figure 6.5a–c shows normalized emission yields of the integral β-intensity in the vicinity of low index principal directions following room-temperature implantation of about \(10^{12} \text{ cm}^{-2}\) of \(^{67}\text{Cu}\) into \(\text{n}^+ - \text{Si : As}\) and post-annealing at 600 °C. Figure 6.5d–f shows best fits of simulated patterns to the experimental yields, with roughly 85% of \(^{67}\text{Cu}\) on ideal substitutional (S) and 15% on displaced S sites [39].
Figure 6.6: Left: photoluminescence spectra of $^{111}$Ag-doped GaN recorded at 4 K at different times as a function of luminescence wavelength or energy. Right: normalized photoluminescence intensities of the Cd band (circles) and the Ag band (squares) in GaN as a function of time [47].

‘Tracer’ techniques

In tracer techniques, element transmutation due to the radioactive decay changes the concentration of impurity atoms in the sample according to a well-known time-scale that is defined by the nuclear half-life of the isotope used. This transmutation process represents an extremely useful analytical tool for understanding the opto-electronic properties, particularly in semiconductors, since these properties are sensitive to the presence of small amounts of impurities. Currently, deep level transient spectroscopy [40], Hall effect [41], capacitance–voltage measurements [42] and photoluminescence spectroscopy [43] use radioactive isotopes to overcome the chemical ‘blindness’ of the non-radioactive versions in studies of the electrical and optical properties of impurities in elementary [44], III–V [45] and II–VI [46] compound semiconductors. Figure 6.6 (left) shows photoluminescence spectra of $^{111}$Ag-doped GaN recorded at 4 K at different times between 1 d and 68 d after the implantation and annealing step. All spectra are normalized to the intensity of the yellow luminescence at 1.97 eV. The right-hand plot shows normalized photoluminescence intensities of the Cd band (circles) and the Ag band (squares) in GaN as a function of time. The solid lines correspond to exponential fits to the experimental data resulting in time constants in perfect agreement with the decay constant of $^{111}$Ag. From these observations, it is clear that the photoluminescence transitions that exhibit a decreasing or increasing intensity as a function of time have to be correlated with defect centres containing exactly one Ag or Cd atom, respectively [47].

The success of nuclear radioactive solid-state physics research depends essentially on the availability of a broad range of appropriate radioactive isotopes and on adequate sample preparation. This requires increased access to beam time in large-scale radioactive facilities and depends on good interaction between the solid-state community and the physicists who have the expertise in the continuously developing field of nuclear science and nuclear experimental techniques.
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7. Facilities and Technical Developments

7.1 Introduction

Development of new facilities and experimental opportunities is essential for research in all natural sciences. In recent decades the atomic physics and material science communities, which use particle beams, have often exploited nuclear physics facilities. Accelerators, detectors, and target developments which were motivated by nuclear physics needs found useful applications here. This situation has changed somewhat: one now finds devices which were developed within the atomic physics and material science communities being increasingly applied in nuclear physics. They include synchrotron radiation facilities, free-electron lasers, high-power optical lasers, traps for cooling atoms and ions, highly charged ions and selective ionization sources. These developments demonstrate the effectiveness of a healthy cross-fertilization between the disciplines.

A few of these developments, selected from those involved in the experiments previously presented in Part 3, will be briefly described in this chapter.

7.2 Ion beam facilities

7.2.1 Ion sources

New ion source developments involve both the production of very Highly Charged Ions (HCI) and the selective production of singly charged ions.

Highly charged ion sources

Sources for highly charged ions are based on electron impact ionization of gas atoms, plasma clouds, or injected low-charged ions. The well-known Electron Cyclotron Resonance (ECR) ion source, Electron Beam Ion Source (EBIS), and laser-plasma ion source have been developed further to a high degree of performance and reliability. They have the following typical characteristics.

- ECR: high-intensity d.c. beams ($\mu$A) of medium-charged ions.
- EBIS: medium-intensity pulsed beams ($10^7$ particles/pulse) of highly charged ions.
- Laser-plasma ion source: high-intensity pulsed beams ($10^9$ particles/pulse) of medium-charged ions.

An interesting possibility is also the further development of Electron Beam Ion sources and Traps (EBIT) with electron beams reaching 300 keV to completely ionize uranium atoms effectively.

For nuclear physics, a main application of highly charged ions is still improving the effectiveness of acceleration and deceleration in accelerators. EBIS and ECR sources are used as ionization boosters in radioactive beam facilities. Externally produced isotopically pure or radioactive singly charged ions are injected into an ECR or EBIT for breeding the charge state to higher values for further acceleration or for trapping. The charge breeding efficiency is rather high, in the order of 10–30%. This type of beam production is already being studied at GANIL with the SIRA (Source d’Ions RAdioactifs) facility, which is the test-bench of the target-ion source system of SPIRAL (for Système de Production d’Ions Radioactifs et d’Accélération en Ligne). Charge breeders for accelerating radioactive beams in compact radio-frequency quadrupole accelerators are going into operation (e.g. at REX-ISOLDE). These will probably be combined with laser ionization to enhance the selectivity for radioactive isotopes. Highly
charged ions are exploited for high-precision mass measurements in traps (SMILETRAP in Stockholm), and for mass determinations of radioactive nuclei in storage rings (ESR at GSI). In both cases the systems rely on the high charge state and on the fact that the systematic errors can be reduced with charge-dependent measurements. A strong activity is atomic spectroscopy in few-electron atomic systems, which relies on the availability of highly charged ions at almost any velocity and in any abundance. Very heavy highly charged ions in large abundance are needed for studies of nuclear-atomic decay modes, e.g., bound-state beta decay and hyperfine transitions.

Unique features of ultra-slow HCI are exploited in fragmentation of atoms, molecules, and clusters with fragment-momentum imaging in recoil-ion spectrometers. HCI are employed as probes for solid-surface investigations.

A previously applied technique for obtaining slow HCI in complex accelerator structures is undergoing a revival: the accel–decel technique. At present, GSI is exploring the possibility of efficiently producing extremely highly charged ions up to bare \( \text{U}^{92+} \) ions at very low kinetic energies. In this so-called HITRAP project, relativistic ions are stripped in a foil, and after injection into the storage ring ESR they are cooled and decelerated to 3 MeV/u. The ions are then extracted and decelerated further in a linear decelerator and brought into a Penning trap for cooling further. It is expected that about \( 10^4 \) ions per second of, for example, \( \text{U}^{91+} \) can be obtained basically at rest in this way. A fair number of such ions are expected to be cooled to 4 K.

Special sources for low-charged ions

Isobar-selective ion sources should deliver isobarically clean beams with a short pulse length at low charge state. High-intensity ion sources for negative ions will be needed in the future, mainly \( \text{H}^- \) sources for producing mA currents of \( \text{H}^- \) for applications in neutron spallation sources and beam plasma heating. There is, of course, still a need for special negative ions for tandem-accelerator operation.

Laser ion sources that are wavelength tunable for selective ionization should be further improved in sensitivity and effectiveness. More sophisticated techniques than are applied today, such as Doppler-free techniques and continuous wavelength lasers instead of broadband-pulsed lasers are to be exploited to improve the selectivity. With these new laser ion sources one could obtain pure beams of any isotopes of Cu, Fe, Ag, etc., at, for example, ISOLDE.

Ion sources for cluster beams are in use now and are being developed further to produce even more highly charged cluster ions. This will allow the currently available range of velocities to be extended upward for cluster beams by efficient acceleration, which is important for several applications.

7.2.2 Acceleration devices: recent developments and projects

The availability of high-performance ion storage rings, swift ion and cluster accelerators producing intense beams with high optical quality and ion sources providing highly charged slow ions has made possible a series of benchmark experiments on particle–matter collisions. Besides important technical improvements in ion beam production, two main types of device open the way to new results in atomic and condensed matter physics.

Cluster accelerators

Apart from the standard sputter-ion sources, which deliver high-energy cluster beams, Liquid Metal Ion Sources (LMIS) such as those operating in Orsay and Lyon have significantly increased the available beam current for metallic clusters like \( \text{Au}_n \). Cold LMIS allow the acceleration of very heavy gold cluster beams (\( \text{Au}_{20} \) to \( \text{Au}_{400} \)) to a total energy of 10 MeV/q, extending considerably the range of projectile size for cluster–solid interaction studies. A new type of molecular ion source has also been developed at IPN-Orsay. It will deliver multiply charged polyatomic ions of high mass (10,000 to 100,000 u) at 10–50 keV/q. The multiple ionization of large molecules or clusters is achieved with an ECR source combined with various injectors.
of neutral or low-charged molecular ions (gas, oven, LMIS, etc.). To reach higher energies, this type of source should be installed on a high-voltage platform.

Storage rings

Heavy-ion storage rings with magnetic confinement are now widely used for atomic and molecular ions, both negatively or positively charged. In recent years, the electron coolers of these rings were pushed to the limit of energy resolution by the very effective beam-expansion method. Having the coldest beams in the merged-beams configuration, they are expected to play an important future role also in electron–ion collision experiments.

A new ‘electrostatic’ type of storage rings and traps has recently appeared, especially suited to very heavy molecular and cluster ions where the mass-to-charge ratio is too large for a magnetically confining heavy-ion storage ring. In the electrostatic devices, however, the species cannot be stored at sufficiently high energy to match the speed of the electrons of the electron cooler in the merged-beams configuration of the rings. Crossed-beam set-ups are being prepared for studies of electron interactions with gas-phase biomolecular ions and cluster ions in electrostatic storage rings. In the future, we can expect to see systems of double rings as well as rings completely cooled to liquid-helium temperature for cooling internal degrees of freedom of clusters and molecular ions.

7.3 Neutron beam facilities

7.3.1 Experimental reactors

Five neutron centres have been built or refurbished in Europe in the past 10 years, and are likely to remain operational without major investment, at least for another 15 years. They are the Institute Laue Langevin (ILL) Grenoble, France; the ISIS Facility, Didcot, UK; the BER-II at the Hahn-Meitner-Institut (HMI), Berlin, Germany; the SNQ at Paul Scherrer Institut, Villigen, Switzerland; and the BRR at KFKI, Budapest, Hungary. In addition, a new research reactor is being built in Munich, Germany. Further lower-flux reactors, which run on less tight time schedules, are still currently requested for student training and isotope production in medical and materials science applications. Most recently the use of ‘supermirror’ coatings in neutron guides is revolutionizing neutron beam extraction and delivery, producing five- to seven-fold beam intensity gains on the sample.

7.3.2 Ultra-cold neutron sources

Ultra-cold neutrons (UCN) are neutrons with energies low enough to be totally reflected from many material surfaces under all angles of incidence. Typically, they have velocities below 7 m/s. They can be trapped in materials or in magnetic and gravitational traps, and they serve as tools for fundamental physics experiments. The precise measurement of the neutron decay lifetime and the search for an electric dipole moment (EDM) of the neutron are the flagship experiments of UCN physics. The EDM experiment, in particular, is considered one of the most important low-energy particle physics experiments.

Over the last 15 years, ILL has provided the most intense source of UCN. At present, it is the only relevant UCN source in the world. However, there are strong new efforts to build next-generation UCN sources in the US, in Japan, and in Europe. There are two major source projects in Europe, one at the new Munich reactor FRMII in Germany and one at PSI in Switzerland. Both projects are based on solid deuterium converters to produce UCN from cold neutrons. The FRMII source will use the continuous cold flux from the reactor, while the PSI source will use pulsed neutron production on a dedicated spallation target. Both projects have the potential to become the world-leading facilities within the next few years. Both aim at increasing the available UCN density by about two orders of magnitude and the intensity by one or two orders of magnitude. Great improvements for fundamental particle physics experiments can be expected. Moreover, the increased UCN density and intensity might open up new fields of applied research.
7.3.3 Spallation source projects

For a long time Europe was the world leader in neutron-scattering science. The US is attempting to catch up by a spallation source project, which is currently aimed at building a facility very similar to ISIS in the UK but with a strongly enhanced power, up to 2 MW compared to 160 kW. This neutron source, SNS, should go on-line by 2007. In December 2000, the Japanese government approved a similar project to be realized by about 2005 in Tokai, Ibaraki province. The basic difference between the US and Japanese projects is that the first calls for a facility essentially exclusively devoted to neutron-scattering research, while it is envisaged in Tokai that the power of a large proton linac, which will ultimately exceed 10 MW after several stages of enhancement, will be shared for a variety of uses, including neutron scattering, nuclear waste transmutation development and nuclear physics.

Since 1992, a consortium of European research centres and universities has been studying the construction of a powerful new spallation source for neutron-scattering research: the European Spallation Source (ESS). A feasibility study completed in 1997 concluded that a 5 MW facility could be built for about 1000 million euros. A central ESS project team was installed in 2000 at FZ Jülich to co-ordinate the decentralized research, development and design efforts of the consortium members. The goal is to present a detailed updated proposal by the end of 2003 to the European governments that calls for the completion of ESS in 2010. Based on the French proposal CONCERT, an alternative is also under study to realize ESS following the Japanese model as one of the facilities housed at an up-to-25-MW proton linac. In order to realize this concept, a liquid-metal target is under consideration, for which recently the MEGAPIE (MEGAWatt PIlot target Experiment) initiative was launched by Commissariat à l’Énergie Atomique (France) and Forschungszentrum Karlsruhe (Germany) in collaboration with PSI (Switzerland) to demonstrate the feasibility of a liquid lead–bismuth target for spallation facilities at MW beam power levels. The realization of ESS will preserve the leadership role of Europe in neutron-scattering studies of condensed matter, in the face of US and Japanese advances which are in the process of being realized. Without an ESS-class facility, this leading position would fade away in the next decade.

7.4 Exotic beam facilities

Two widely complementary muon spin resonance ($\mu$SR) user facilities are operational in Europe: the continuous-beam facilities at the Laboratory for Muon-spin spectroscopy (LMU) at PSI in Villigen, Switzerland and the Pulsed Muon Facility (PMF) at ISIS (CLRC) in Oxfordshire, UK.

Other facilities exist in the world, which are currently providing muons for materials and chemical analysis. Besides these centres, the Joint Institute for Nuclear Research in Dubna, Russia, and CERN in Geneva, Switzerland have developing programmes for implementation of muon factories. Important technical achievements in recent years are the development of low-energy muon beams and the extraction of one muon at a time from a continuous beam at PSI.

Recently, the availability of MeV positron beams at the Stuttgart MPI für Metallforschung has allowed new experiments leading to the characterization of transient processes involving positrons and positronium.

7.5 Radioactive ion beam facilities

7.5.1 Developments

There is a need for technical developments concerning beams produced by ISOL facilities having increased intensity, energy and elemental/isotopic variety.

ISOLDE at CERN is the best example, and it is continually evolving. However, for some specific cases, other institutes with different production methods could be complementary, provided that adequate investment for targets and beam intensity is given.
SPIRAL, based on the ISOL method, will provide production and separation of radioactive beams at GANIL. The radioactive atoms are ionized by an ECR source and subsequently accelerated by a $K = 265$ cyclotron to energies of $1.7-25$ A MeV. The charge states of the accelerated ions are typically 9+ for Ar and 15+ for Kr beams. The ejection energy of the ion source is between 12 q keV and 34 q keV. The installation of a dedicated beam line at this energy should permit the implantation of radioactive ions for solid-state studies. These studies are already possible (but with low beam intensities) at GANIL in the SIRA facility.

Energetic radioactive ion beams with controlled energy ranging from a few eV up to MeV are needed. Particularly in solid-state physics, a tunable beam energy opens new possibilities, e.g. to reach a specific layer in a multilayer system, to reduce the local concentration of implanted ions by performing a series of implantations with different energies, and to reach the active depletion layers in semiconductors, outside the surface defects and space-charge region.

### 7.5.2 Radioactive nuclei accumulators

As in the case of mass spectrometry in storage rings, fast cooling of hot fragments should be achieved by stochastic cooling in a dedicated accumulator and cooler ring for measurements of very short half-lives by the time-of-flight technique. For measurements of long half-lives by the Schottky technique, an optimized storage ring should provide efficient electron cooling and detection. In the latter case resonant pick-ups for the Schottky signal should be further developed in order to increase the signal-to-noise ratio. Clearly, higher intensities of radioactive ion beams as planned for the second-generation RIB facilities are crucial.

Beam cooling and bunching techniques are prerequisites for efficient use of RIB. The segmented ion cooler (SIC) devices will be essential elements for beam improvement with respect to time structure and emittance. The techniques should be further developed to reach nearly 100% transmission efficiency in the cooling as well as in the bunching mode with short residence time in the SIC. Stopping of relativistic ion beams in gas cells and its combination with a SIC will enable ISOL-type experiments to be performed with ions at rest or at an energy of some 10 keV also at in-flight facilities where the reaction products have an energy of several 100 MeV/u. Here and in the case of the SIC, systems have to be developed which provide a closed cycle for the noble gas of extreme purity used to stop the relativistic ion beam and to cool it.

### 7.6 Development of microbeams

Nuclear microprobes have well-established applications in materials analysis and microfabrication. There they provide a rich area of research for small groups having relatively modest budgets. Microprobes are highly versatile, and there is an ever-increasing number of material subjects to study. Consequently, there is a multitude of laboratories in Europe equipped or being equipped with ion microbeam facilities. The obviously desirable technical developments are improved energy stability and 3D spatial resolution.

With the advent of radioactive beams, there will be requests to obtain radioactive microbeams for material research. Such beams will soon be available at European radioactive beam facilities: the ISOLDE facility at CERN, Geneva; the ISL at the Hahn-Meitner Institute, Berlin; the cyclotron and isotope separator of the ISKP, Univ. Bonn; and the SPIRAL facility at GANIL, Caen.

### 7.7 Laser beam facilities

#### 7.7.1 Free-electron lasers

Novel electron accelerator techniques can be used to create free-electron lasers (FEL) in a wide wavelength interval extending to the X-ray region. Generally it is the case that the shorter the wavelength, the higher the electron energy necessary. For an infrared FEL one uses today typically 50 MeV electrons whereas for an X-ray laser 1 GeV electrons are needed. X-ray FELs
are therefore developed at large accelerator facilities (DESY Hamburg, Stanford, Brookhaven). The wavelength ranges from millimetres to microns and from VUV to X-rays are of special interest. Due to the coherent emission via the self-amplified spontaneous emission (SASE) process, very large intensities can be obtained, with peak brilliance ten orders of magnitude higher than in conventional synchrotron radiation. This is accompanied by very short pulse structures (in the femtosecond regime). The pulse structure is a direct consequence of the electron-beam time structure; it can therefore be varied over a large range, taking advantage of modern accelerator technology. A unique example is the TESLA facility of DESY, Hamburg. This, in contrast with competing facilities (SLAC, USA and KEK, Japan), is based on highly advanced superconducting technology. TESLA, a 33 km long e⁻e⁺ collider integrates the SASE X-ray laser complex. These machines therefore open completely new prospects for sciences that study material and molecular structures as well as their dynamics, using the time resolution, wavelength and brightness. Three-dimensional imaging, with phase-sensitive holography and very high space-time resolution will be possible. These studies will elucidate the structures of atoms, molecules, crystals and materials in femtosecond snapshots.

### 7.7.2 High-power laser systems

The tremendous advances in producing intense short-time laser pulses in the visible wavelength regime at several facilities can be expected to reveal completely new phenomena in atomic physics, accelerator physics, and material sciences. In Europe, particularly in France and Germany, a number of excellent high-intensity lasers exist. In the short-pulse high-intensity mode, they will be able to focus hundreds of terawatts power on a spot of a few microns on the target. A petawatt laser system has so far only been realized with NIF at LLNL, USA. At GSI the PHELIX project provides a kJ high-power laser that will be combined with a heavy-ion beam simultaneously focused on the same spot, in order to reach even higher power densities or to study time evolutions in the plasma. The main motivation for these intense lasers is the study of high-temperature highly unstable plasma physics and developments of drivers for inertial-confined fusion devices.

These laser facilities are beginning to bear fruit in the spectroscopy of few-electron highly-charged ions, in a new access to study atomic and molecular fragmentation dynamics, and in providing high-intensity short-time X-ray pulses or in driving an X-ray laser in the future. This again is due to the large abundance of highly charged ions produced in the plasma. The use of plasma heating is being considered for new types of particle accelerator. In material science, there exists the exciting option to produce and to study for the first time metallic hydrogen in a laboratory. This is expected to form when the very high-density compression is reached with the short-pulse high-intensity mode of these facilities.

### 7.8 Instrumentation

#### 7.8.1 Ion traps

Traps for ion mass spectrometry should be further developed along two branches. One is extremely high precision mass determinations. This requires cooling of highly charged ions in a Penning trap, either resistively or by electrons or positrons. A development in this direction is followed at SMILETRAP and HITRAP. The other branch will be non-destructive single-ion identification and single-ion mass measurements by the Fourier-Transform Ion Cyclotron Resonance (FT-ICR) technique. This will only be possible by operating the trap — as well as the electronics for detection of image currents induced by the orbiting ion — at liquid-helium temperature. This development is essential in the case of rare events such as found in the region above $Z = 100$.

Superconducting magnets with fields considerably higher than available today, but still very homogeneous, will lead to higher accuracy or enable access to shorter-lived nuclei. Along these lines, the use of higher harmonics to excite cyclotron motion should be investigated. Furthermore, the efficient transfer of the ions under investigation into Penning traps is a central
issue for further studies. Stopping and cooling techniques (gas cells and segmented ion coolers) should be further explored with respect to achieving the highest efficiencies and shortest periods of time for cooling and bunching the ions. This is a special challenge in the case of gas cells for stopping relativistic fragment beams.

Another type of trap is also of primary interest for the study of fundamental interactions: the Laboratoire de Physique Corpusculaire of Caen has built a transparent Paul radioactive ion trap (PIF, for Piège-Interactions-Fondamentales). Here, the goal is to study the beta-neutrino angular correlation in a beta-decay process for singly charged ions. This trap will be installed on a future low-energy line of SPIRAL.

### 7.8.2 Target developments

Rapid developments in target preparation techniques occurred recently by utilizing atomic traps and cold supersonic jets. The development of atom traps by the atomic physics community has led to various applications within the field of nuclear physics. First of all the ensembles of cold atoms can be used — similarly to those from supersonic jets — as thin targets for collision experiments, also suitable for storage rings. For example, at TSR in Heidelberg a target of cold Li atoms is being developed for monitoring the spatial profile of the circulating ion beams. Moreover, the detection of collisionally induced reaction fragments with high spatial, momentum and energy resolution (as in recoil-ion momentum spectroscopy) allows the study of collision processes in much detail. Perhaps most promising is the possibility to investigate decay processes of accelerator-produced radioactive atoms in a trap with unprecedented completeness and accuracy: With spin-polarized nuclei practically at rest the complete kinematics of the decay process — including the neutrino — can be obtained by measuring the electron or positron in coincidence with the recoil nucleus. Corresponding experiments are now being prepared at various places in the world and will look for physics beyond the Standard Model.

### 7.8.3 Detector developments

*Particles and photon detectors*

There are two areas in detector developments urgently needed by the community where some progress can be reported.

- The first one concerns the development of two-dimensional position-sensitive detectors for electrons, ions, neutral particles, photons, etc. They should be equipped with fast readouts, cover a large detection area, and be able to handle high count rates, with multi-hit capabilities. Such detector systems would need to be tailor-made around high-power lasers or intense ion beams. They would also be very useful for experiments involving low intensities of radioactive beams or trapped ultra-cold highly charged ions. Finally, they could detect the emission of radioactive samples with short half-lives. Such a detector made of a 256-anode array coupled to standard channel plates has been developed at Orsay to study the interaction of fast and large clusters with solids. This device, associated with dedicated time encoding electronics, allows the simultaneous measurement of 256 time-of-flight spectra on an event-by-event basis, triggered by the same start signal. It provides the mass identification and the localization of several hundred ions emitted under a single impact with repetition rates of several kHz.

- The second area is dedicated to effective high-resolution detectors, which have a wide spectral coverage. There is a growing need for this type of detection device for X-rays, charged, or neutral particles. Promising developments have begun based on the bolometric or calorimetric principle. For example, in these detectors a photon is converted in an absorber to phonons, which lead to an accurately measurable temperature rise. The temperature rise is amplified by the low heat capacity of the absorber at a very low temperature, and by the high sensitivity of the thermistor used to measure the temperature rise. So far a resolution of 8 eV at
6 keV X-ray energy has been reached. The size of the absorption chip is, however, only a few μm square, so that an array of such chips is required for efficient photon detection. The resolution is mainly governed by thermodynamic fluctuations in the absorber and the Johnson noise in the thermistor. At present, it is still difficult to control these parameters for a large array of such detector chips. That problem should be solved in the near future. One would then have a powerful detection system for many applications in heavy-ion research, such as Lamb-shift spectroscopy, nuclear reactions with radioactive beams with fast mass spectroscopy, and energy analysis of heavy-ion channelling experiments.

*Neutron detectors*

An important factor determining the neutron data collection rates is the solid angle around the scattering sample covered by detectors. Here the advances with cost-effective, position-sensitive, pixel detectors present a real breakthrough. The recently commissioned neutron spectrometer at ISIS has a total of 16 m² detector area covered with 2.5 cm resolution detectors position sensitive in both dimensions on a quasi-spherical surface segment of 6 m radius. This amounts to detector solid-angle coverage of 0.44 sr. On modern neutron diffractometers up to 4–8 sr detector coverage has also been achieved. A particularly spectacular breakthrough has been made at ILL by adapting the cheap image plate technology to Laue-type single-crystal neutron diffraction work, primarily for small biological samples. The LADI detector covers close to 4π solid angle with a resolution of about 10⁶ pixels.