CERN-EP/79-35
18 April 1979

CERN LIBRARIES, GENEVA

CM-P00071022

EXPERIMENTS WITH INTENSE SECONDARY BEAMS OF RADIOACTIVE IONS

H.L. Ravn

CERN, Geneva, Switzerland

(Submitted to Physics Reports)

ABSTRACT

Electromagnetic mass separation applied on-line to accelerators and nuclear reactors is now a standard technique for producing preselected isotopic beams (A,Z selection) of nuclear reaction products. The development, performance and anatomy of a large on-line isotope separator facility, the CERN ISOLDE, is discussed. As a result of recent technical developments it is now possible to study individual nuclei of about 40 elements, in many cases out to where the limits of nucleon binding are approached and half-lives become as short as 10 ms. It is shown that the nuclear reaction processes induced with the high-intensity several hundred MeV proton beam can provide secondary radioactive beams in almost all regions of the nuclidic chart with intensities which are not matched by any other method. The intense beams of 10^8 - 10^{11} atoms/s have opened up a number of new experimental possibilities like laser spectroscopy on radioactive atoms, radioactive targets for nuclear reaction spectroscopy, and precision X-ray shift measurements.

CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
2. HISTORICAL DEVELOPMENT	2
2.1 Twenty-five years of on-line isotope separation	2
2.2 The role of radioactivity in nuclear physics	4
2.3 Isotope and element separations in nuclear spectroscopy	7
3. ON-LINE ISOTOPE SEPARATORS	8
3.1 Isotope separators on line to nuclear reactors	10
3.2 Isotope separators on line to heavy-ion accelerators	11
3.3 High-energy proton reactions	12
3.4 The ISOLDE-2 facility	12
4. TARGET AND ION SOURCE	16
4.1 New understanding of the release kinetics	17
4.1.1 The transfer-time distribution	18
4.1.2 Release of reaction products from solid target matrices	19
4.1.3 Release of reaction products from molten metals	22
4.1.4 Special cases of release from molten metals	28
4.2 Selective target and ion-source systems	29
4.2.1 The target design principles	30
4.2.2 Beam heating	30
4.2.3 Production of the alkalis	31
4.2.4 The alkaline earth elements	33
4.2.5 The rare earth elements	34
4.2.6 The halogens	35
4.2.7 The noble gases	36
4.2.8 Mercury, cadmium and zinc	37
4.2.9 Gallium, indium and thallium	38
4.2.10 Chemical separation after the mass separation	38
4.2.11 Notations	39
5. EXPERIMENTS WITH FAR UNSTABLE NUCLEI	
5.1 Yields and purity	42
5.2 Detection methods and limits	43
5.3 The role of proton beam intensity and other particles	44
5.4 Identification of unknown nuclei	46
6. THE USE OF RADIOACTIVE BEAMS OF HIGH INTENSITIES	49
6.1 Atomic beam magnetic-resonance measurements	50
6.2 Optical spectroscopy	50
6.3 Experiments with radioactive targets	52
6.4 Hyperfine effects in solids	52

6.5	Shifts in the energies of K X-rays	52
6.5.1	The crystal spectrometer	54
6.5.2	New effects influencing the energies of K X-rays	55
7.	CONCLUSION AND OUTLOOK	57
	REFERENCES	59
	Tables	68
	Figure captions	70

1. INTRODUCTION

The study of nuclei far from beta stability is one of the newest and most rapidly growing fields of nuclear physics. The experimental programme has broadened, matured, and increased in scope, as a consequence of developments and improvements of techniques associated with on-line electromagnetic isotope separation. The Isotope Separator On-Line (ISOLDE) at the CERN 600 MeV Synchro-cyclotron (SC) has been in operation for the past 11 years. It is the largest facility of its kind, providing a wide range of intense secondary ion beams of short-lived nuclei for experimental investigation. It should be noticed that the progress has not just been on the technological side. The impression is rather that there has been correspondingly great progress in the experimental methods, both in those for preparing the ion beams and in those that make use of the beams. Typical examples of the latter are the experiments based on methods from atomic physics: optical pumping, laser spectroscopy, atomic beam techniques, precision X-ray spectroscopy, and mass spectrometry. Thus, at the moment when on-line isotope separation has passed the 25 year mark and as CERN's ISOLDE has entered its second decade there still is no indication that the progress is levelling off; the methods exploited so far seem to have scraped the surface only. This field still seems to be a young one, where ideas are more important than prestigious investment plans.

It is the purpose of this review to discuss the experimental aspects of on-line mass separation. In order to provide some of the general background information needed to become acquainted with the subject, section 2 contains the historical development.

The new ISOLDE-2 facility, which was taken into use in late 1974, is discussed in section 3 in connection with general problems of on-line isotope separation. Section 4 deals with the most important problem of on-line separation, the target and ion-source technique. Section 4.1 describes off-line studies of target matrices that liberate one or several elements produced in nuclear reactions. By combining these target materials with a suitable ion source, a number of the

elements can be obtained in the form of an ion beam. Target and ion-source systems, which allow a fast and often very selective ionization, are treated in detail in section 4.2. The performance of these target assemblies is discussed in section 5 with special emphasis on the isotopic yields, purity, and cross-sections. The role of detector sensitivity, proton beam intensity, and other particles beams is summarized together with the status of the experiments with the nuclei furthest away from stability, close to the proton and neutron drip lines.

Finally, section 6 describes a new class of experiments with no relation to far unstable nuclei, which exploits the very high beam intensities (10^6 - 10^{11} atoms/s) only available at ISOLDE.

2. HISTORICAL DEVELOPMENT

2.1 Twenty-five years of on-line isotope separation

The first experiment to make use of the beams of short-lived radioactive nuclei produced by means of an on-line electromagnetic isotope separator was carried out in 1950 by Kofoed-Hansen and Nielsen [1]. Although the importance of this new experimental method was immediately realized in Scandinavia and elsewhere, there was no immediate sequel to this work in spite of the arguments given by Koch [2]. The reasons for this delay were mainly of a technical nature. In the fifties, the methods of nuclear spectroscopy were not good enough to handle the extremely complex decays of far unstable nuclei: NaI(Tl) counters, point-by-point magnetic spectrometers, and slow and unreliable multichannel analysers. The contrast between the techniques of the pioneering experiment in 1950 and those of nuclear physics 24 years later has been underlined in a charming as well as informative exposé by Kofoed-Hansen [3].

Towards the mid-sixties the interest in on-line mass separation began to increase strongly. In part this was due to greatly improved experimental techniques, in particular the advent of the Ge(Li) spectrometer in 1964 and of computers for nuclear spectroscopy experiments, but in addition there was now an understanding of nuclear structure theory that allowed the prediction of a number of effects specific to the far unstable nuclei, such as the new regions of deformation and

magicity. The opening guns in this new development were fired by Rudstam [4] for the ISOLDE collaboration and by Bergström [5], who also was one of the driving forces behind the Lysekil Symposium on "Why and How should we investigate Nuclides Far Off the Stability Line" [6].

A large number of new on-line experiments now began to take shape. The merits of the different kinds of bombarding particles were (and are, maybe) still imperfectly understood, and the target and ion-source techniques were still at a very speculative stage. Thus it is interesting to note that the complex chemical systems that were eagerly discussed in the mid-sixties turned out to be much harder to develop than anyone had thought. The early development of this phase can be followed through the proceedings of the separator conferences in Marburg [7] and Skövde [8], while the early physics results were reviewed at the Leysin Conference [9].

CERN's ISOLDE installation has from the very beginning held a special position. This, to a large extent, is a spin-off of CERN's unique role in European high-energy physics: it has combined strong central support with the efforts of many groups in the member countries. Thus, from the beginning of ISOLDE operation in October 1967, groups from CERN, Denmark, France, Germany, Norway, and Sweden have been involved, and have contributed with ideas, manpower, and money. The original ISOLDE has been described in a report edited by Kjelberg and Rudstam [10]. As new techniques began to develop, it also became clear that ISOLDE had been very fortunate in the choice of bombarding projectile: high-energy protons offer a variety of products in intensities that cannot be matched by other methods. The upgrading of the CERN SC carried out in the year 1973-74 further served to strengthen ISOLDE's position.

Well ahead of the machine shutdown, ISOLDE had undertaken a massive development programme aimed at finding new target and ion-source techniques capable of providing beams of elements not previously available and of improving the yields, especially of the rare short-lived products. The concept of ISOLDE was, already in the early seventies, very much that of a "facility" that delivers beams (of radioactive nuclides) to a number of experiments, and the analogy with other

secondary beams in high-energy physics (muons, pions, antiprotons, etc.) is evident. The effort that was put into the development of new secondary beams amounted over a period of about two years to about 50% of the total machine time available and this turned out to be a good investment.

In May 1973, the old ISOLDE installation was shut down to undergo a major reconstruction concurrently with that of the SC. As will be discussed later (section 3.4), this led to greatly improved conditions, not only with respect to beams and intensities, but also so far as space for experiments was concerned. The range of nuclei accessible to on-line study has been extended along the N axis towards the limits of stability, as well as along the Z axis to make intense radioactive beams of up to 40 elements available. Extreme neutron-deficient and neutron-rich ion beams with intensities down to 1 atom per second have been used to procure data on new nuclear species. The intense (up to 10^{11} atoms per second) beams closer to stability have allowed several high-intensity experiments to go on-line.

2.2 The role of radioactivity in nuclear physics

The first observed natural phenomenon in which the atomic nucleus manifested itself was in the radioactive decay processes. In connection with the discovery of X-rays by W.C. Röntgen in 1895, H.A. Becquerel observed in 1896 the radiation from naturally occurring radioactivity. The study of this phenomenon started the chain of discoveries that in 1934 led to the demonstration of artificial radioactivity. The progress in the study of radioactivity and X-rays became now extremely rapid and these studies have since developed hand in hand. They have called for similar techniques of instrumentation and have played an important role in establishing today's concept of the atom.

The known and newly produced nuclei could now simply be defined in terms of two quantities, the proton number Z and the neutron number N. A grid could be formed with these two axes, where each square represents a possible aggregate of protons and neutrons. This nucleonic chart, as shown in fig. 2.1, not only provides a convenient reference for nuclear properties, it also enables a prediction of various modes of radioactive decay and half-life as one moves away from

the line of beta stability (defined by the squares representing the stable nuclei) towards the regions where the nuclei become particle unbound.

The greater part of our knowledge of the low-energy properties of the nuclei was derived from the radioactivity studies in the years 1934-55. This knowledge formed the basis of the shell model of Mayer and Jensen in 1949, according to which many properties of low-lying nuclear levels can be understood from the motion of a single nucleon moving in a spherical symmetric potential that could be regarded as an average over the interactions with all nucleons in the system. It gave a first firm basis for an explanation of the properties like spin and parity assigned to a number of the measured states. The model was further complemented by Bohr and Mottelson, with the collective model [11,12] which showed that the nuclear motion often involved many nucleons giving rise to shape oscillations and rotations. From radioactivity measurements and Coulomb excitation experiments it was found that the known nuclei largely fell in two different kinds of regions:

- a) where the nuclei have spherical shape with vibrational spectra; these nuclei are found near to the closed shells;
- b) spheroidal nuclei which exhibit rotational spectra and are found in the regions far from closed shells.

An important consequence of this was the realization [to my knowledge first presented by B.R. Mottelson in a lecture (unpublished) at a conference in Copenhagen in 1961] that we must expect "new regions of deformation" away from the beta-stability line. This idea, first taken up by Marshalek et al. [13] has played a large role in the subsequent work on far unstable nuclei.

The single-particle states appropriate to the spheroidal potential have been calculated by Nilsson [14] and the corresponding wave functions allowed a detailed understanding of the level spectra of deformed odd-A nuclei. During the sixties the nuclear level studies shifted towards in-beam techniques like Coulomb excitation and nuclear reaction spectroscopy, whereas the radioactivity studies changed towards the application in Mössbauer spectroscopy and hyperfine studies.

Up to 1965 most radioactivity work was performed with the relatively long-lived nuclei situated close to stability, but the chart of nuclides in fig. 2.1 clearly demonstrates that most particle-stable nuclei still remained undiscovered as exotic species far from stability. With the new experimental techniques in view it was soon recognized that there were a number of specific questions that only could be answered by radioactivity experiments with these nuclei which have extreme proton to neutron ratios. The number of key questions discussed at the Lysekil Conference [6] and in the review by Bergström [5] shows that there was no lack of interesting problems, among which can be mentioned:

- i) Delayed-neutron emission and other properties of neutron-rich nuclei (important for the r-process in astrophysics).
- ii) Delayed-proton emission, and beta decay of proton-rich nuclei ($Z > N$).
- iii) Nuclear-level densities.
- iv) Nuclei along the $N = Z$ line and isospin.
- v) Alpha-particle emission.
- vi) Nuclear radii and moments.
- vii) Systematics of Q-values and nuclear masses.
- viii) New deformed and transitional regions.
- ix) Nuclear-reaction yields.

Over the last decade these studies have advanced significantly and surprising new effects have been found, as often is the case when new techniques are taken into use. The technical development is continuing and while many experiments undoubtedly will benefit from further progress, it is now possible to investigate extensively the above-mentioned items. A large quantity of high-quality data have already been produced so that the theoretical predictions and models can be sensitively tested and developed. A complete overview of the results in this field can be found in the proceedings of the last two conferences on nuclei far from stability, in Leysin [9] and Cargèse [15], and also in a forthcoming review paper by P.G. Hansen [16].

2.3 Isotope and element separations in nuclear spectroscopy

The changes in the nuclear charge due to the radioactive transmutations lead to changes in the chemical properties. Marie Curie was the first to take advantage of this to separate and identify naturally occurring radioactive nuclei. Since then chemical methods have become an important tool in the investigation of the atomic nucleus. Hahn and Strassmann's discovery of the low-energy fission processes, where a complex mixture of hundreds of radioactive nuclei distributed over 40 elements is formed, was followed up by the development of the radiochemical techniques. By application of classic inorganic qualitative separation techniques, methods that allowed a separation of the mixture into its elemental constituents were developed. From 1932, when artificially accelerated particles were taken into use for producing nuclear transmutation, these chemical methods were refined to separate nuclides free of isotopic carrier as needed for high-resolution nuclear spectroscopy.

Many new experimental approaches have been brought to bear on the short-lived nuclei. The classical radiochemical techniques have been further developed to allow discontinuous separations on the time scale of seconds as reviewed by Trautmann [17]. However, most progress of recent years comes from experiments which utilize purely physical or physico-chemical techniques. The use of high-resolution semiconductor radiation detectors either in beam or in connection with various fast transfers of unseparated reaction products like recoil spectrometers, gas flows (helium jet), and mechanical transfers [rotating wheel, pneumatic tube transport systems ("rabbits") and tape transports] are most powerful if the nuclide of interest is formed with a substantial fraction of the compound nucleus cross-section. These techniques have recently been reviewed by Macfarlane [18] and Armbruster [19].

The decisive point is that the product of interest usually is formed in a very low cross-section, while at the same time the main reaction is very complex and forms many different product nuclei in high yields. This is true for high-energy proton reactions, for fission, and -- at high energy -- even for heavy ion

reactions. Figure 2.2 shows a typical example of the mass yields for high-energy proton spallation. The isotopic distributions of the cross-sections for the reaction $\text{La} + \text{p}(600 \text{ MeV})$ are calculated here, according to the semi-empirical formula of Rudstam [20], which agrees well with experiment.

It is clear that the basic difficulty in the study of these nuclei is to achieve sufficiently high selectivity, and that a separation not only in Z , but also in A , is of importance. Consider as an example the observation of ^{114}Cs produced in a cross-section of $\sim 1 \text{ nb}$. With no separation it is clearly an almost impossible task to detect this product, but even after a chemical separation the Cs products are still dominated by the much more intense nuclei close to stability. Only in combination with a mass separation are clean conditions obtained. The application of mass separators in nuclear physics studies is not a new phenomenon. Already in 1934, shortly after the application of artificially accelerated particles to induce nuclear reactions, low-current mass spectrometers were used to provide stable isotopic samples for targets. Their first use for separation of artificial radioactive nuclei was demonstrated by Yamaguchi [21] as early as 1941. In a 5 MeV deuteron irradiation of NaF the well-known 15 h sodium activity was induced. By collecting ions of mass number adjacent to the stable isotope ^{23}Na , the activity was unambiguously assigned to ^{24}Na . For these purposes the mass spectrometers were developed into the high-intensity isotope separators. The large scale ^{235}U separations in the wartime Calutrons produced an enormous accumulation of fundamental data, which was of great importance in the construction of the smaller laboratory machines. The type of separator used in the present work was developed in Scandinavia. A full account of the early history of these machines and their use for nuclear physics purposes can be found in the review articles by Alväger and Uhler [22] and Nielsen [23].

3. ON-LINE ISOTOPE SEPARATORS

The on-line isotope separators which are in operation now are similar to the off-line machines, from which they differ substantially only in the target and ion-source arrangement. As sketched in fig. 3.1, the particle beam intercepts

the production target situated directly inside the ion source or attached to it via a tube. By proper choice of target matrix and ion-source configuration, one or several nuclear-reaction-produced elements are continuously transferred to the ion source. The source is generally optimized for a maximum yield of singly charged ions often containing only one chemical element. By means of an extraction electrode and a lens, the ions are accelerated through a potential of typically 10-60 kV. The beam is separated into its isotopic components in the analysing magnet and the array of well-separated beams is brought to intersect the focal plane from where the beams of interest can be conveniently directed out towards the experimental equipment through a more or less complex beam-handling system. What are now the major problems in achieving this? First one must choose a combination of bombarding particle and target material that allows the produced nuclei of one or several elements to be released in a form acceptable to an ion source. Depending on the chemical and physical properties of the product elements this problem has been solved with varying degrees of success for approximately 44 elements, but only 20 of those can be provided in a chemically pure form. The second problem is the distribution of delay times, i.e. the times between the formation of a nucleus in the target and its detection. Most on-line target and ion-source systems have average delay periods in the range of tens of seconds and only a few systems are fast enough to operate efficiently in the half-life region from 10 to 100 ms. Thus, the elements of fundamental importance in on-line separator work are the nuclear reaction and the target and ion-source arrangement. They determine the range of elements that can be studied as well as their yield and purity. Three types of nuclear reactions are capable of producing the far unstable nuclei: thermal neutron-induced fission, heavy-ion reactions, and reactions with high-energy protons. Since the use of isotope separators on line to nuclear reactors and to accelerators has become widespread, a vigorous development of new experimental techniques continues to broaden the range of elements available for study as well as increasing their intensity. It is not the intention here to give a complete survey of the great variety of techniques in use; for this the

reader is referred to the review articles which may be found in the proceedings of some recent conferences [7-9,15]. However, for the sake of comparison, it may be of interest to briefly discuss the main features of experiments which use other nuclear reactions than those of high-energy protons as used in this work. Given in table 3.1 are the main parameters of modern accelerators and reactors, the data for the three mostly used reaction types, and the present status of the corresponding three kinds of on-line separators.

3.1 Isotope separators on line to nuclear reactors

The up to 20 b high cross-sections for the formation of individual fission products in thermal neutron-induced fission of ^{235}U make this reaction very attractive for the production and study of the heavy neutron-rich nuclei. Today the majority of the experiments are being performed with isotope separators on line to nuclear reactors; in a recent summary Wohn [24] lists 11 experimental programmes. Most of these arrangements work with external thermal neutron beams, where the flux has dropped by orders of magnitude. In this case the neutron beam can be conveniently directed towards the target positioned outside or directly inside an ion source, but the obtained yields are generally very low. In order to study the very neutron rich nuclides produced in fission, the high neutron fluxes available in a reactor channel close to the core have to be used. The transport of the radioactive atoms should be carried out as a well-focused ion beam to the analysing magnet outside the reactor. This technologically difficult problem of positioning an ion source near to the reactor core was first solved at the OSIRIS isotope separator on line to the R2-0 thermal reactor at Studsvik in Sweden [25]. It is probably not too much to say that this is the most important separator for large-scale production and study of thermal fission products. The target consists of 3 g of ^{235}U distributed as finely divided U_3O_8 particles onto the fibres of a graphite-cloth cylinder, situated inside a typical plasma ion source. All the fission product elements except the low volatility elements Y to Pd are separated simultaneously with ion-source yields which vary between 30% and 0.1%. It is clear that this system has no appreciable separation from the isobaric precursors. The delay time is reported to be only 0.5 s and the

production rate for a neutron flux of 4×10^{11} n/cm² s, brings the most abundant radioactive beams up to an intensity of 10^8 atoms/s.

3.2 Isotope separators on line to heavy-ion accelerators

The principal heavy-ion reactions for production of nuclei far from stability are:

- i) complex multinucleon transfer reactions which permit the formation of neutron-rich isotopes of the lightest elements;
- ii) heavy-ion xn and (xn,yp) reactions, which are confined to the production of nuclei on the neutron-deficient side of stability.

It is the reactions mentioned under point (ii) that have been of main interest in the on-line separator work. The production cross-sections fall appreciably below the compound nuclear cross-section (typically 200 mb) as one moves away from stability, where the low proton-binding energy causes proton evaporation to compete favourably with neutron evaporation. Owing to the short range of the heavy ions in matter, only very thin targets can be used, typically a few mg/cm² or less. This limits the maximum production yields to about 10^6 s⁻¹ for an incident particle beam of 10^{12} s⁻¹ as available today. However, at the extreme neutron deficient wing of the yield distribution, the heavy-ion reactions appear to give the highest possible yields. This is clearly demonstrated in the work of Karnaukhov's group [26] at the BEMS-2 separator on line to the U-300 heavy-ion cyclotron at Dubna. In their system the large linear momentum transfer is exploited to permit the reaction products to recoil through a 1 μ m thick tungsten window into a high-temperature surface ionizer. Most of the recoils are stopped in a catcher in the front plate of the ion source, from where they diffuse out and are ionized with average delays of 5-10 s and in yields of 15-40%. This system has mainly shown its strength in extending the region of known nuclei of the elements Ba-Eu. However, the surface ionization in the form used here does not provide any element selection. At the heavy-ion accelerator UNILAC in Darmstadt, Roedel's group uses a similar recoil system combined with a plasma ion source [27,28]. By means of this system very high yields of the neutron-deficient alpha emitters in the region above tin have been obtained. Since the efficiency of a plasma source only varies

little with Z (in this case 1-10%), the system yields isobarically pure sources exclusively.

Common for the heavy-ion on-line arrangements are the limitations in target and ion-source lifetime, due to the extreme strains in the thin targets and windows subjected to the beam heating and sputtering.

3.3 High-energy proton reactions

The main mechanisms in reactions with protons in the GeV range are spallation, fragmentation, and fission, for which the independent reaction cross-sections rise, respectively, to 20 mb, 10 mb, and 3 mb, near to stability, and rapidly become very small far from stability. These reactions are therefore not especially favoured in terms of product formation cross-section. Nevertheless it is becoming increasingly clear that high-energy protons, as discussed in detail in section 5.3, are the most powerful particles for a large-scale production of nuclei far from stability in almost all regions of the nuclidic chart. The reasons for this are the high penetrating power of the protons combined with the high beam intensities available from modern proton accelerators. Separators on line to high-energy proton beams can use thick targets typically 20-200 g/cm²; this gives potentially much higher yields (up to 10¹⁰ atoms/s for a proton beam of 6×10^{12} p/s or 1 μ A) of the nuclides of interest and the deleterious effects of the beam are weakened, being spread out in a volume of about 20 cm³. The maximum intensity of the proton beam available at ISOLDE is 2-5 μ A, but much higher ones are available at several other accelerators and there seems to be no technological reason prohibiting the use of beams between 10-100 μ A.

3.4 The ISOLDE-2 facility

Situated as seen in fig. 3.2, next to the SC [29], the ISOLDE installation restarted its operation in December 1974 after 1½ years of reconstruction in conjunction with the SC improvement programme, which aimed at an external proton beam intensity increase to 4 A. A number of newly developed experimental techniques together with numerous other improvements, based on the experience gained during the previous 5½ years of ISOLDE experiments, were introduced. The facility has

now been working successfully for four years, during which time further improvements have been introduced and some shortcomings have become apparent.

The major points are seen from the layout in fig. 3.3. Most of the installation was for space and shielding reasons placed 8 m below the ground level, which has the disadvantage of making the now needed extensions of the experimental area extremely cumbersome and expensive. The changes undertaken to cope with the greatly increased radiation levels were to make additional shielding in the form of a double wall with two movable doors between the target region, the collector chamber, and the experimental area, as described in detail in another paper [30]. After the first years of operation, experience showed that further radiation protection measures were needed around the target and the last quadrupole lenses in the proton beam. An additional 50 cm marble wall was erected around the last quadrupole lenses with the double purpose of screening the hall UR-2 against the radiation from the active target and from the activity induced in the lenses by the beam losses. Since some of the recently developed target matrices react with moist air and possibly could liberate radioactive dust, a remote-controlled system permits the used target units to be sealed off and kept under vacuum. For the same reasons, the target area UR-6 is hermetically enclosed within a reinforced glass wall and kept at an underpressure with respect to the surroundings by means of a ventilation system provided with absolute filters.

The rapid target-changing mechanism [31] shown in fig. 3.4, that allows a remote-controlled change between four different targets, has been of decisive importance for the extensive use of the facility. The activation and radiation damage of this equipment and the acceleration chamber caused by the 10^8 rad received during 10^4 μ Ah of operation calls now for a complete exchange. Any future increase in beam time and intensity necessitates a substantial improvement in the remote-handling equipment. The location in UR-7 of the electronics rack at 60 kV, which contains the power supplies for the target, ion source, and the extraction lens, was dictated by space considerations. It has the advantage of a short distance to the target, but the radiation restricted access and the space limitations

in this room are clearly a bottleneck in the efficient utilization of the facility. An elaborate external beam-handling system, which allows the parallel operation of four mass beams and a rapid changeover between 10-15 experiments, is based on a switchyard [32] suggested by Andersson [33]. Its four electrostatic deflectors shown in fig. 3.5, which move mechanically along the direction of the external beams, cover altogether a mass range of $\pm 15\%$ of the central mass.

The increased space gained in the experimental room UR-8 by rearrangement of the separator and the introduction of an elaborate system of external beams soon proved to be insufficient to cover the demands from the experiments. In order to gain more space and also to reduce the background problems caused by radio-activity accumulation in UR-8, a continuation of beam line 2 through a vertical branch to a new 140 m^2 experimental hall situated 8 m above and on the ground level has been constructed. From fig. 3.3 it is seen that the vertical deflection is made electrostatically with an additional experimental station in the intermediate room UR-9.

With the present construction of a similar extension of beam line 3 the possibilities for further expansion in the existing buildings are exhausted. Since the general tendency is for the present and the planned experiments to be increasingly space-demanding, the limitation in floor space remains an unsolved problem. The general technique of the isotope separator itself is not much different from the conventional off-line machine, but it may be worth while to mention a few points specific to the on-line mode. The oil-free vacuum obtained by the consequent use of turbomolecular pumps, and the low-capacitance motor-generator system used to transfer the power from the mains to the supplies situated at 60 kV, has greatly contributed to the stability and simplification of the machine. The original high-tension screen solution, adopted to cope with the load variations on the acceleration supply due to the ionization of the air caused by the proton beam and secondary radiations as described in ref. 29, proved to be insufficient. The leak current from the high-voltage terminal, which has a peak value of 3 mA and an average value of 1.5 mA, with the SC working in the fast burst mode at an intensity of $1 \mu\text{A}$, caused a high-voltage ripple with an amplitude of 100 V. The stability

was restored by reintroducing a series regulator acting on the high-current screen supply using the low-current stabilized supply as a reference. The stability is now better than 2×10^{-5} , which assures a beam-ripple amplitude of less than 0.3 mm. Another perhaps essential feature, which has added considerably to the long-term stability and simplification of operation, comes from the change in total beam current. Originally the machine was designed as a so-called low-current ($I = 500 \mu\text{A}$) separator, where additional gas and mass-marker elements had to be introduced in the ion source. With the introduction of surface-ionization sources and low-pressure plasma-type ion sources as discussed in section 4, combined with more sensitive beam scanners, the systems for mass-markers and supporting gas could be abandoned. Mass calibration is assured by means of the most intense radioactive beams and weak impurity beams which both can be observed by means of the oscillating needle scanner, as illustrated in fig. 3.6. The total beam current is now of the order of 300 nA and the operation resembles more that of a mass spectrometer.

The high intensities and the wide range of nuclides available at ISOLDE-2, in combination with the flexible beam-handling system, have considerably increased the number of experiments that can be pursued on line. In fig. 3.7 it is seen that 10 major experimental stations can be accommodated in the experimental area. There has been a steady increase in the beam time used for experiments at ISOLDE-2, so that the design aim of 1972 (180 shifts) has been exceeded. At present (1979) the operation is at 30 shifts per month. The utilization of the beam time is illustrated in tables 3.2 and 3.3. Up to 13 groups have taken data, including off-line measurements and collection of long-lived activities, during a typical ISOLDE beam-time period of 10 shifts. The remote-controlled target-changing mechanism that allows changing from beams of one element to another within a few hours, has permitted experimental periods of up to 26 shifts in which 1-4 different targets have been in operation. During that time the stability of the ion beams is such that they can be kept within an area of 0.5 mm^2 , with a focus that allows apertures a few tenths of a millimetre wide. The transmission from the collector chamber to the experiments varies from 95% for the shortest beam tubes to 80% for the longest.

The running of the facility is done without the continuous presence of operating personnel. A start-up procedure lasting from 3 to 6 hours, during which time the target ion-source and proton-beam parameters are optimized to maximum radioactivity output, is normally sufficient. After this it is left to the user to adjust the transmission of the ion beam through the switchyard and the external beam line to the experiment, to choose the wanted mass, and to do minor adjustments of the ion-source during the experiment. This procedure, which for the longest beam lines involves the somewhat elaborate setting of up to 25 different power supplies, is performed manually, but it has often been discussed whether to computerize it. Since the adjustments are strongly dependent on the not very reproducible ion-optical properties of the different ion-source and target assemblies it is still considered to be a too difficult and especially time-consuming task to change to computer-controlled operation. However, there are a few parameters which lend themselves to automatization. The remote-controlled stepping of the magnet field between four programmed masses has been extensively used in mass-measuring experiments. Automatic change of beam-line settings, as needed by different experiments, is possible owing to remote-controlled deflector and lens supplies [34]. Furthermore, remote-controlled variation of the acceleration voltage together with the rest of the ion-optical system as needed in collinear laser experiments (see section 6.1) are being studied.

4. TARGET AND ION SOURCE

The function of an on-line target and ion-source unit is to provide, over prolonged periods, a continuous release and subsequent ionization of the element of interest. The product nuclei should leave the unit on the shortest possible time scale in order to minimize the decay losses of short-lived nuclei. To assure the proper operation of the ion source, this process has to be performed in a vacuum of 10^{-4} Torr, which sets the upper limit to the evaporation of the target matrix and other vapours accompanying the radioactive nuclei at 10^{-8} mol/s. In other words, the composition of the target has to withstand the heating and radiation damage induced by the proton beam. Owing to the requirements of high yield

and selectivity, no universal target or ion source can be found. For each product element a separate unit has to be developed and techniques for all elements have not yet been found. The crucial points in achieving this are the kinetics of the release from the target matrix and the combination of the target with an ion source to form a selective system. In the following these two points are discussed in detail.

4.1 New understanding of the release kinetics

The extraction of one specific element from the complex mixture of an irradiated target is not a new problem. Over the last 60 years radiochemical separation methods for all the known elements have been developed. Many of these methods have in the last few years been further refined to become fast and continuous, so that they constitute a useful tool for the study of short-lived nuclei. Recent reviews of this field have been given by Herrmann and Denschlag [35] and Trautmann [17].

The use of these classical radiochemical procedures in connection with an ion source of an on-line isotope separator is generally not possible, since the final isolated product most often is presented as a solution or wet precipitate, i.e. accompanied with large amounts of foreign matter which are incompatible with the vacuum requirement of an ion source. These conditions are most easily met by volatile species. Techniques which involve what is often called physical separation methods, like recoil momentum, evaporation from low vapour pressure target matrices like powders, foils, molten salts or metals, appear for the moment to be the only useful methods. As shown in the review by Herrmann and Denschlag [35], such methods were rarely used in the past. A noticeable exception is the release of noble gases from solid emanating compounds as discovered by Hahn and Strassmann [36]. The early uses of this effect for the study of short-lived nuclei have been reported by Bjerge [37] for 0.8 s ^6He and 38 s ^{23}Ne . Later ISOLDE started by using noble-gas emanating targets consisting of hydroxide powders [38]. The rapid escape of these and other elements from solid or molten targets is generally governed by one or more of the following three steps:

- i) The transfer of the reaction products from the target interior to the surface by means of recoil, diffusion, and convection mechanisms.
- ii) Release from the target surface by desorption or chemical reaction.
- iii) Transport to and through the ion source may play an equally important role because of wall adsorption and desorption steps encountered by the diffusing atoms.

The final yield is determined by the decay losses during these mass-transfer processes and the efficiency of the ion source.

In spite of the great abundance of diffusion data in the literature, it has been difficult to predict the properties of new on-line target and ion-source systems, because release-kinetic studies of target and product combinations of interest for the nuclear studies seldom were available. Even in cases where seemingly relevant data exist, they may not be applicable, since the trace amounts present in an actual target assembly often behave differently from the macro amounts used in the experiments for obtaining the data. In addition, radiolysis and reactions with crucible materials may cause further complications. Therefore suitable target matrices and conditions for a rapid escape as well as ionization and containment have to be sought experimentally. The vapour pressures of the elements may serve as a guideline for the choice of target-product combinations. As a rule of thumb one may assume that a fast target is possible if the pure product has a vapour pressure > 1 atm at the temperature where the target-matrix vapour pressure is $< 10^{-6}$ atm; still there is in general no simple correlation between these properties and the escaping fraction. For example, as shown in ref. 31, Te does not evaporate from molten La at 1400°C despite the fact that its vapour pressure at this temperature is ~ 10 atm. A lanthanum telluride with low vapour pressure is formed, which keeps the Te strongly bound in the liquid phase. It is clear that the chemical properties have to be taken into account, and not only the vapour pressure.

4.1.1 The transfer-time distribution

The bases for the target and ion-source systems to be described later are the studies of the release and transfer of the radioactive products formed in

different target materials. These studies are usually pursued off line in connection with a search for suitable containment materials as shown in refs. 31 and 39. The emphasis has been on the rate at which the release takes place. By means of test ovens, which simulate the on-line experiments, irradiated samples of prospective target materials are taken through successive heating and gamma-spectrometric measurement cycles at different temperatures. The result for a given element is presented as the fractional activity as a function of sample heating time. This fractional activity is defined as the ratio between the activity of a characteristic isotope in the sample after the heating period and the activity before any heat treatment. Later on these delay curves may be studied in more detail in on-line experiments by observing how the yield of a given isotope falls off with time after the proton beam is switched off, as described elsewhere [31,39-41]. These data allow the determination of the delay-distribution function, that is the probability $p(t) dt$ that an atom formed at time zero is deposited on the collector in the time interval t and $t + dt$. This important distribution function is found by differentiation of the measured delay curves. As shown in the next section, it allows the calculation of the delay-time losses of short-lived nuclides and the prediction of the yields. In order to analyse the influence of the various release mechanisms on the function $p(t)$, the discussion below is given separately for solid targets, liquid targets, and special cases. A complete formulation of the rate equation for the over-all rate of release embodying the rate of all known steps is not excluded, but would probably be too complex to offer much insight. Fortunately, the experiments usually point to a single rate-determining step.

4.1.2 Release of reaction products from solid target matrices

Already in 1965 it was pointed out by Andersen and Scharff [42] that Ta would constitute a possible target material for spallation production of the neutron-deficient rare-earth isotopes, which are of particular physics interest owing to their alpha-decay mode. Later on Hansen et al. [43] showed that refractory oxide powders kept at high temperature formed a fast target owing to the short solid-state diffusion path combined with the fast diffusion at high temperature.

Likewise, a variety of fission products were quickly released from composites of uranium oxide and thin foils of graphite or graphite fibres kept at high temperature, as shown by the groups of Bernas [44] and Rudstam [45].

In ref. 39 the investigation of the release properties of a variety of refractory metals and chemical compounds as a function of temperature, particle size, and time was described. A typical result, given as fractional activity as a function of sample heating time, is shown in fig. 4.1. In all investigated cases of release from solids the measured delay curves showed a complex behaviour. It could be shown in good agreement with known diffusion coefficients and activation energies for surface desorption that the following two different mechanisms could be rate-determining.

- i) The diffusion of the products through the bulk to the surface of the powder particle.
- ii) The desorption from the particle surfaces, which in the case of porous targets may lead to a slow pore diffusion.

In the case where the diffusion in the solid state was found to be time-controlling, the time-dependence is described by Fick's second diffusion law, written for a sphere in terms of spherical polar coordinates and assuming purely radial diffusion:

$$\frac{\partial c}{\partial t} = D \left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r} \frac{\partial c}{\partial r} \right), \quad (4.1)$$

where D is the product diffusion coefficient, c the concentration, and r the particle radius. By solving this equation for uniform spherical particles with an initial homogeneous activity distribution as boundary condition the following expression for the fractional activity can be derived [46]:

$$F(t) = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2 \pi^2 D t / r^2} \quad (4.2)$$

if

$$p(t) = - \frac{\partial F(t)}{\partial t} \quad (4.3)$$

$$p(t) = \frac{6D}{r^2} \sum_{n=1}^{\infty} e^{-n^2 \pi^2 D t / r^2} . \quad (4.4)$$

By fitting the experimental data to eq. (4.2), shown by the full drawn line in fig. 4.1, the diffusion coefficient D could be determined. Consequently, for cases where literature values for D exist the assumption of grain diffusion as the rate-determining step could be checked. The present experiments are not suited to give a precise value for the material property D ; only a diffusion parameter μ_s characteristic of the system can be determined with sufficient accuracy owing to the uncertainty in the particle shape and size distribution

$$\mu_s = \frac{\pi^2 D}{r^2} . \quad (4.5)$$

Like the diffusion coefficient, the temperature dependence of μ_s can be described by the Arrhenius relation:

$$\mu_s = \mu_s^0 e^{-Q_s/RT} , \quad (4.6)$$

where Q_s is the activation energy for the diffusion process, R the gas constant, and T the absolute temperature. The activation energies can now be determined from eq. (4.6) [39,47].

Depending on the forces between the product atoms and those of the surface of the powder particles or any other surface the product atom encounters on its way through the target and ion-source system, it will take a certain time before it acquires sufficient energy to desorb from the surface. This average lingering time t is given by the Frankel equation [48]

$$t = t^0 e^{Q_p/RT} , \quad (4.7)$$

where t^0 is a constant of the order of 10^{-13} s, and Q_p the activation energy for desorption. For elements where $Q_p > 4$ eV the corresponding value of $t > 1$ s at 1800°C , and the transport through the capillaries of the powder bed becomes the rate-determining step. The mathematical treatment of this case is very similar to the case of solid-state diffusion. By defining an effective diffusion coefficient D_e , which determines the diffusion through the capillary length L_e , we can apply the diffusion law as described above. The result becomes:

$$F(t) = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} e^{-(2n+1)^2 \mu_p t} \quad (4.8)$$

$$\mu_p = \frac{\pi^2 D_e}{L_e^2} \quad (4.9)$$

$$p(t) = \frac{8D_e}{L_e^2} \sum_{n=0}^{\infty} e^{-(2n+1)^2 \mu_p t} \quad (4.10)$$

By means of the off-line studies [39], a series of fast high-temperature metal or carbide target matrices was found and the product elements could be classified according to the two rate-determining mechanisms as shown in fig. 4.2. These results were further supported in the on-line experiments [47] and by the fact that the determined diffusion parameters generally agreed quite well with those measured by Beyer et al. [49].

It is interesting to note here that the diffusion parameter μ_s values measured on line show a general trend to higher values than those measured off line. This effect is ascribed to the influence of the ionizing radiation and the nuclear reactions which create additional vacancies and interstitials. Such an enhanced diffusion was clearly seen in the case of Rb produced from Nb powder, where the determined diffusion parameter μ_s had increased by a factor of five as compared with the values measured in the off-line experiments.

The experimentally determined μ_s values allow the determination of the ratio between the observed and true -- i.e. decay-loss corrected -- yields Y_{obs}/Y by means of the simple equation derived in other papers [40,47], which for the case of $\lambda > \mu_s$ becomes

$$\frac{Y_{\text{obs}}}{Y} = \frac{3}{\pi} \sqrt{\frac{\mu_s}{\lambda}}, \quad (4.11)$$

where λ is the decay constant of the nuclide.

4.1.3 Release of reaction products from molten metals

A particularly favourable property of molten-metal targets is their high density, which in combination with the penetrating power of the 600 MeV proton beam

assures the highest possible production rates. In addition, their stability to the deleterious effects of the prolonged heating and irradiation is excellent. The first generation of ISOLDE liquid-metal targets [50] made use of the evaporation of the transition elements Zn, Cd, and Hg, from melts of the group 4A elements Ge, Sn, and Pb, respectively. In all three cases the chemical selectivity was assured since the neighbouring elements to the product of interest were not released at a noticeable rate. This target concept has been extended to chemically more reactive metals for the production of many more elements. In a series of off-line experiments [31] the volatilization and transfer of long-lived radioactive isotopes formed in a variety of different target melts were studied. The evaporation yields and the upper limit for the release rate were determined, and in particular the difficult problem of containment of the liquid metals was solved by suitable alloying and oven-construction techniques. These results extended the number of elements that could be produced from liquid-metal targets to 25 as shown in fig. 4.3.

In the on-line experiments with these targets it was found [31,40], in agreement with the measurements on Pb and Sn targets by Hagebø et al. [50], that the delay-time distribution of products from liquid metals to a good approximation could be written as a single exponential function of the form

$$p(t) = \mu_{\ell} e^{-\mu_{\ell} t} . \quad (4.12)$$

This function, which is shown in fig. 4.4, is well characterized by an average delay time $\tau = 1/\mu_{\ell}$, which experimentally is found to take values down to 30 s. It varies with temperature according to an Arrhenius relation and the determined activation energies are usually found to be close to the heat of evaporation of the pure product element. As shown in ref. 40 it allows the determination of the following expression for the release yield:

$$\frac{Y_{\text{obs}}}{Y} = \frac{\mu_{\ell}}{\mu_{\ell} + \lambda} . \quad (4.13)$$

In order to determine the relative importance of various possible rate-determining mechanisms, separate on-line experiments were made. In these the average delay

time was measured as a function of target volume, the free volume above the melt, the length of the transfer line between the target and ion source, the outlet opening of the ion source, and the degree of forced convection in the melt. Since none of these experiments affected the average delay time it was tentatively concluded, in agreement with Hagebø et al. [50], that the time-limiting step lies in the evaporation process.

Indeed the evaporation of pure metals in vacuum appears under most experimental conditions to be a purely surface process [51]. In the actual case where the question is to distil a microcomponent, the nuclear reaction products, out of a low vapour pressure liquid this assumption seems not to hold.

From kinetic theory considerations the maximum rate of evaporation of a component in a mixture can be derived [52]

$$J = \frac{S_0 p_1}{(2\pi M_1 RT)^{1/2}}, \quad (4.14)$$

where p_1 is the partial pressure of the volatile component, M_1 its molar weight, and S_0 the surface area.

This equation is only valid under the following assumptions:

- i) The conditions in the liquid should be turbulent enough for the surface concentration of the solute to be kept at the same value as the bulk concentration.
- ii) The pressure of the atmosphere above the liquid should be reduced to a low value, so that the motion of the vaporized atoms is not influenced to any appreciable extent by the atoms of any residual gas*).

*) In practice this is the case when the residual gas pressure is maintained at a value not much greater than 10^{-3} Torr.

- iii) The evaporating surface should be pure; surface films must be absent.
- iv) The solvent and the solutes should form ideal solutions.
- v) There should be no mechanical obstacle along the path of the vapours from the surface to the condenser.

In case of deviations from (ii) and (iii), eq. 4.14 must be modified by introducing an additional coefficient α . Experimental evidence [52] indicates that the evaporation coefficient α may assume values from 10^{-6} -1. If the alloy does not obey (iv), the molar fraction x should be replaced by the activity, which represents the actual ratio of the vapour pressure of the volatile component above its alloy to the vapour pressure of that component in the pure form. Since the solutions considered here are extremely diluted, Raoult's law is assumed to be valid. Deviations from (v) are taken into account by introducing the factor ϕ , which is the probability for re-entry into the liquid. By substituting in eq. (4.14) the partial pressure p_1 obtained from Raoult's law and the molar fraction x_1 :

$$p_1 = P_1 x_1 \quad (4.15)$$

$$x_1 = \frac{M_0 N_1}{V_0 \rho_0 A} \quad (4.16)$$

$$J = \frac{S_0 P_1 M_0 N_1}{V_0 \rho_0 A (2\pi M_1 RT)^{1/2}}, \quad (4.17)$$

where P_1 is the saturated vapour pressure of the product, N_1 its production rate, M_0 the molar weight of the target, V_0 its volume, ρ_0 its density, and A Avogadro's number. The mean escape rate τ_0 becomes:

$$\tau_0 = \frac{N/A}{J} \quad (4.18)$$

$$\tau_0 = \frac{V_0 \rho_0 (2\pi M_1 RT)^{1/2}}{S_0 P_1 M_0} . \quad (4.19)$$

Assuming the above-mentioned conditions (i)-(iv) to be fulfilled for the release of Cs and Xe from molten La at 1200°C , substitution of the numerical values gives $\tau_0^{\text{Cs}} = 4 \times 10^{-3}$ s and $\tau_0^{\text{Xe}} = 5.4 \times 10^{-6}$ s. This is in good agreement with the average lingering time measured for these elements on other metal surfaces [53].

Since the actual target and ion-source assembly [31] allows the evaporated atoms to recondense on the target surface, the effective average delay τ_{eff} becomes

$$\tau_{\text{eff}} = \tau_0 \phi . \quad (4.20)$$

From standard vacuum-flow considerations [51]

$$\phi = \frac{S_0}{S_i} \left(\frac{3\ell}{4d} + \frac{3L_i}{4D_i} \right) , \quad (4.21)$$

where ℓ , L_i are the length, and d , D_i the diameter of the transfer tube and the ion source, respectively, and S_i is the ion-source outlet area. The numerical value gives effective average delays

$$\begin{aligned} \tau_{\text{Cs}} &= 50 \text{ s} \\ \tau_{\text{Xe}} &= 6.8 \times 10^{-2} \text{ s} \end{aligned}$$

as compared with the experimental values from ref. 40 and fig. 4.4

$$\begin{aligned} \tau_{\text{Cs}} &= 30 \text{ s} \\ \tau_{\text{Xe}} &= 24 \text{ s} . \end{aligned}$$

The experimental facts that τ_{Cs} was independent of the geometry of the target and ion source, and that the agreement with the Xe results is poor, indicate that some other effect than the pure kinetic evaporation process is rate-determining. A good fit to the experimental data may be obtained by introducing different evaporation coefficients α for Cs and Xe, but this is in disagreement with the fact that evaporation coefficients from molten metals under the above-mentioned conditions generally are found to be unity [52].

It seems much more likely that the condition (i) does not hold, despite the fact that the stirring experiment described in ref. 31 had no effect on τ_{Cs} . Owing to the high rate of desorption of the volatile product from the La surface, the resulting impoverishment of this component in the surface causes the diffusion through the liquid to be rate-determining. This conclusion is further supported by the following two experiments.

The average delay of Rb from a molten Y-La alloy was measured as a function of V_0/S_0 by means of the off-line radioassay method described in section 4.1.2.

The samples of the alloy were melted in shallow crucibles under such conditions that the probability of re-entry was negligible. The result is shown in fig. 4.5 together with the τ_{Rb} measured from a typical on-line target, which allows the product nuclides to collide many times with the surface of the melt. It is seen that the short desorption times τ_0 expected in the off-line experiment on the basis of eq. (4.19) were not obtained. The fact that the on-line result fits the curve is taken as an indication that the nuclides are not reabsorbed in the melt, but may well be readsorbed on the surface and then released with the short lingering times given by eq. (4.19) and ref. 53.

In an on-line experiment on the release of Hg from a molten-Pb target, the yields of ^{179}Hg ($T_{1/2} = 1$ s) and that of ^{197}Hg ($T_{1/2} = 64$ h) were measured simultaneously as a function of the vertical position of the proton beam on the target. It was clearly seen that while the yield of the long-lived ^{197}Hg started to drop when the proton beam reached the surface of the melt, the delay time limited yield of ^{179}Hg rose to a maximum.

The description of the mass transfer from a liquid as in the case of a molten-metal target is different from that of solids which are limited purely by molecular diffusion. The liquid is usually not stagnant, but mixed or agitated by natural convection currents so that the transfer takes place by a combination of the slow molecular diffusion and the relatively fast eddy diffusion. The mathematical treatment of this situation is taken directly from the methods of reasoning used successfully to describe heat transfer from liquids. It is assumed that the rate limitation is due to a molecular diffusion through a stagnant or laminar flowing surface film [54]. The few available mass-transfer studies of liquid metals [55] strongly support the surface-film concept and so far no rate limitations due to surface- or chemical-transfer resistance on clean metal surfaces have been uncovered.

The average film thickness z , which is determined primarily by the liquid dynamics of the system, is usually incorporated into a grouping of parameters, defined as the mass transfer coefficient K , which replaces D/z in the diffusion calculations. The numerical value of z is typically $\sim 10^{-3}$ cm [56,57] in the case

of a liquid metal under natural convection conditions. As shown below, this compares very well with the z -values that can be calculated from the release data of a typical ISOLDE molten-metal target.

The diffusion rate through a plane stagnant liquid layer with the surface S_0 is given by Walas [54]

$$J_d = \frac{D_1}{z} (c_0 - c_1) S_0 , \quad (4.22)$$

where c_0 and c_1 are the concentrations on each side of the film. Assuming equilibrium and $c_0 \gg c_1$

$$c_0 = \frac{N_1 V_0 z}{S_0 D_1} . \quad (4.23)$$

The total amount of product present in the target volume is

$$V_0 c_0 = \frac{N_1 V_0^2 z}{S_0 D_1} . \quad (4.24)$$

The average delay time is

$$\tau_0 = \frac{V_0 c_0}{J_d} . \quad (4.25)$$

From eqs. (4.11), (4.12), and (4.13) the film thickness z is obtained:

$$z = \frac{S_0 D_1 \tau_0}{V_0} . \quad (4.26)$$

By introducing the numerical values from fig. 4.4 and a typical diffusion coefficient $D_1 = 2 \times 10^{-5} \text{ cm}^2/\text{s}$ for liquid metals [55], a film thickness of $2.5 \times 10^{-3} \text{ cm}$ is obtained. This very good agreement with diffusion-limited mass-transfer rate has the interesting implication that the film thickness and hence the delay time may be further reduced by the introduction of forced convection. However, the applied liquid velocities have to be much higher than was obtained in the vibration experiments [31].

4.1.4 Special cases of release from molten metals

The statements made above apply only to clean metal surfaces free from surface layers and adsorbed films of residual gases. If the surface is covered with

a layer of oxide which forms a solid diffusion barrier, the rate of release may be decreased: if covered by a reactive gas that forms a volatile product compound the release rate may be increased, as demonstrated in the chemical evaporation techniques [52]. These two cases have also been encountered [31].

The average delay of Hg from Pb decreased from 120 s to 30 s by shaking the molten metal during irradiation. Off-line experiments showed that the Pb surface was covered with a skin (presumably of the oxide) which was disrupted by the onset of stirring. After prolonged heating under vacuum, this layer gradually disappears together with the shaking effect, so that the unstirred target is left with an average delay of ~ 30 s. The conclusion that the longer delay time is due to diffusion through a solid surface layer is in good agreement with general observations in metal distillation [58]. Similarly, experiments with a liquid-La target, as mentioned above, showed no such effect, since La has the particularity of dissolving its own oxide layer above a certain temperature [31].

The opposite effect -- the chemical reactive evaporation -- is of great interest for on-line targets, since it allows the release of otherwise non-volatile elements. This effect has been used by Sidenius [59] in isotope separation of stable elements. One such case is for the moment receiving attention -- the release of In as volatile InCl from molten Sn. By controlled addition of AgCl vapours to the Sn target surface, AgCl concentration-dependent average delay times of the order of 1 min have been observed in off-line tests [60].

4.2 Selective target and ion-source systems

As discussed in sections 4.1.2 and 4.1.3, a series of target materials have been found which allow about 40 nuclear-reaction-produced elements to be released sufficiently fast in a form acceptable to the requirements of an ion source. Most of these target matrices simultaneously release several neighbouring elements formed in the complex nuclear reaction. In order to achieve sufficiently high selectivity it is of fundamental importance to perform a chemical separation, preferably before the mass separation so that mono-isotopic beams can be delivered to the experiments. The separation according to the nuclear charge Z is basically obtained by the application of the following principles:

- i) vapour pressure differences;
- ii) differences in desorption activation energies;
- iii) characteristic ionization potentials or electron affinities;
- iv) formation of molecular ions;
- v) chemical reaction on the target surface.

In the following, the mechanical construction and some different selective types of target and ion-source combinations will be discussed individually.

4.2.1 The target design principles

As described in detail in ref. 31 and shown in fig. 4.6, the target and ion-source assembly is contained in a common vacuum chamber, which can be connected by remote control to the separator acceleration tank. This system allows a great variety of target and ion-source combinations to be mounted. Generally the target matrix is kept in a heated cylinder axial to the proton beam direction and outside the ion source. As seen from fig. 4.7 [30], products released from the target are brought to the ion source by molecular diffusion through a transfer tube. The extraction electrode is now fixed with respect to the ion source inside the central pumping port, which connects the target vacuum chamber to the acceleration tank. This is done in order to be able to remove the highly radioactive extraction electrode together with the target unit. The maximum target length was chosen to be 20 cm, which allows the use of molten-metal targets of one nuclear interaction length down to the element lanthanum.

4.2.2 Beam heating

The intense proton beam incident on the target deposits part of its kinetic energy in the target material, which causes considerable heating of the target. The main problem for the target design is to evaluate this effect in order to assure, if necessary, sufficient heat dissipation and to determine the maximum possible beam intensities which can be used with the present techniques.

In a calorimetric measurement, an iron target of 79 g/cm^2 with the typical ISOLDE geometry was mounted in a styrofoam calorimeter and irradiated with a 50 nA proton beam. The analysis of the cooling curve of the target as measured

by means of a thermocouple and the proton beam measured by means of Al foil activations gave an energy deposition of 9.3 W or 106 W/ μ A. This is in good agreement with a simple energy-loss consideration omitting corrections for the nuclear reactions and the secondaries not stopped in the targets, which would assume an average dE/dx of 2 MeV/g, i.e. 7.9 W for this experiment. The power dissipated in the targets by means of the heating current varies from 300 W for Pb to 3.6 kW for Ta. Clearly the Pb target would already at 2 μ A need additional cooling, whereas the Ta target would stand about 20 μ A without cooling. In both cases the main heat loss from the targets is by radiation from the outer of the 3 (Pb target) or 5 (Ta target) heat screens surrounding them. With present cooling technology, such as used in high-power-density machines like reactors (300 MW/m³) and high-current X-ray tubes (46 kW/cm²), it is estimated that the present target technique should be able to cope with proton beams up to 100 μ A.

4.2.3 Production of the alkalis

The phenomenon of surface ionization, in which atoms of a metal vapour impinging on a hot metal surface may lose an electron and re-evaporate as an ion [61], is a very simple and convenient way to attain the required high efficiency and selectivity for the alkali metals with their low ionization potentials (3.9-5.4 eV).

Since the on-line use of this principle by Amarel et al. [62], numerous groups have used this effect for on-line mass-separator studies especially of the fission products. At ISOLDE this principle has been used in the development of a series of target and ion-source systems by means of which all known (and many previously unknown) isotopes of the alkali elements have been produced.

The first generation of targets used molten metals or alloys, which because of their high density have particularly high production rates and are well suited for the production of intense ion beams of nuclei with half-lives down to a few seconds. In fact Cs⁺ beams with intensities of up to ~ 15 nA or 10^{11} s⁻¹ can be achieved with the 2.5 μ A proton beam available today. For the production of extremely short-lived nuclei the solid-state targets discussed in Section 4.1.2 have to be used. Owing to their low density and the practical limitation to 20 cm long

target containers, the effective target thickness and hence the production rates are typically a factor of 3 to 10 lower than for the liquid metals. For the short-lived nuclei this is outweighed by the fast delay times characteristic of these targets.

For the production of the neutron-rich alkalis, targets consisting of uranium and thorium carbides have been extensively used. The difference in release yields of Cs isotopes as produced from liquid and solid targets are shown in fig. 4.8 [47]. Similar improvements can be obtained for the neutron-deficient Cs nuclei by means of a LaC_2 target.

The usual surface ionization source, which has been widely used in mass spectroscopy and also used in the first isotope separator by Oliphant et al. [63], consisted of a flat emitting surface or a filament doped with the charge material. Later developments [64] led to the flat porous emitters through which the charge material diffuses from a reservoir behind. These source geometries are generally not very useful for on-line purposes where they have to communicate with a larger target volume. The yield, pumping speed, and delay time may be severely restricted. The use of an enclosed ionizing surface, as in the tubular surface ionization source with axial slit extraction described by Harrison [65] is much more favourable for on-line use. In fig. 4.7 is shown how this ion-source principle has been further developed and combined with a thick target for production of the alkali metals at ISOLDE. The surface ionization takes place to a great extent on the target-material surface or on the wall of the transfer tube, at the end of which a constriction on the tube serves as the ion source. In order to avoid ionization around the exit of the source, it is cooled down by means of a copper cone. It is interesting to remark, as discussed in ref. 31, the particular fast transport of the ions through the transfer line and the ion source, which is heated by a d.c. current. In fact the alkali ions drift towards the source outlet because of the heating potential put across the line and the ionizer. The ionization efficiencies obtained by means of this ion source for the alkali metals are close to 100%. Owing to escape of atoms through openings which compete with the extraction orifice, such as the mass-marker oven inlet and around joints, the typical figure drops to $\sim 70\%$.

4.2.4 The alkaline earth elements

The surface ionization technique by means of the tubular ion source described in the previous section can be extended to give good yields of elements with ionization potentials up to ~ 7 eV. In that case the Ta ionizer is exchanged for one made of materials with high work function (Re or W) and the ionizer temperature is raised to 1700-2800°C. The inset in fig. 4.7 shows how such a temperature may be achieved directly by ohmic heating. Depending on the temperature, efficiencies of 20-100% have been obtained for the alkaline earth elements. This is several times more than one would expect for pure surface ionization as calculated by the Saha-Langmuir formula. Latuszyński and Raiko [66], who have investigated this kind of ion source in detail, explain this effect qualitatively by a complex interaction between two effects, the space charge produced in the tube by the ions and electrons flowing from the wall, and the effective extraction orifice for ions due to the penetration of the extraction field into the tube. Under these conditions, however, the corresponding alkali metal is also fully ionized. In order to separate the alkaline earth elements from the alkalis we have used two different approaches.

From mass spectroscopic and thermochemical studies of alkaline earth and rare earth fluorides [67], it is well known that their monofluorides, contrary to those of the alkalis, form quite stable molecular ions and that they have ionization potentials which are equal to or lower than those of the pure metals.

By controlled addition of fluoride vapours to the target from a small oven, it has been possible to direct the radioactive alkaline earths into the surface-ionized monofluoride side band displaced by 19 mass units with respect to the alkali isobars. The second possibility of separating the two elements is to make use of the fact that only the alkali metal is fully ionized during its transport in the Ta transfer line. By application of a reverse potential for heating the line up to the constriction (see fig. 4.7), reduction of the alkali beam with respect to that of the alkaline earth by a factor of 4 has been achieved. This somewhat low value may be explained by a high gas pressure in the target and ion

source giving rise to neutralization. Much higher separation factors have been observed in tentative experiments with electrostatic extraction of the alkalis through a hole in the transfer tube. In both cases the over-all yields of the alkaline earth will become lower than those mentioned above. Owing to the finite stability of the monofluorides, the temperature dependence of their ion current exhibits a broad peak at ion-source temperatures below 2000°C, which might explain the up to 10 times lower ionization efficiency.

4.2.5 The rare earth elements

The fast on-line mass separation of the physically and chemically very similar rare earth elements presents a special challenge in view of their lengthy separation by conventional chemical methods. The variations shown in fig. 4.9 of their heats of sublimation, ionization potential, and dissociation energies for their monoxides and monofluorides indicate possible ways for their on-line separation.

As expected from the sublimation data in fig. 4.9 and the off-line studies discussed in ref. 31, the element pairs Sm, Eu, and Tm, Yb evaporate from targets of molten Gd-La alloy and Lu-La alloy, respectively. Combined with the high-temperature surface ionizer, prolific beams were obtained from the Gd-La alloy [31]; however, no means of separating the Sm and Eu isobars has been found. The analogous case of the extremely expensive Lu-La alloy never came to an on-line test, since in the meantime a high temperature Ta powder target was developed [39,41]. Combined with the ohmic-heated tungsten surface-ionizer (2800°C) shown in fig. 4.7, a very fast highly selective Yb producing target was achieved. The slow pore diffusion caused by the high desorption energies and affinity to oxygen trapped the neighbouring elements so that a decontamination factor of 10^3 was obtained. Since the diffusion rate of the rare earths in Hf at 1900°C is the same as that in Ta at 2400°C, whereas the surface desorption energies are the same [39,68], Hf should constitute an equally fast and more selective target material for production of Yb. Several attempts to verify this were unsuccessful, owing to the difficulty of obtaining sufficiently pure Hf powder.

The high stability and volatility of the rare earth monofluorides indicate a possibility to release and transport the surface desorption-rate-limited rare earths by means of the chemical evaporation technique described in section 4.1.4.

4.2.6 The halogens

There is no lack of target matrices that among many other elements will release also the halogens [31,39]. As in the previously discussed cases the major problem is to obtain chemical selectivity. A very elegant way to separate the halogens from their neighbouring elements is to make use of their high electron affinities $E_A \approx 3$ eV, which readily allow them to form negative ions in the inverse of the process of positive surface ionization. This inverse process is described by an emission equation analogous to the Langmuir equation for positive surface ionization [61]

$$\beta_- = \left[1 + \frac{g_0}{g_-} e^{\frac{\phi - E_A}{KT}} \right]^{-1}, \quad (4.27)$$

where ϕ is the work function of the ionizing surface, and g_0 and g_- are the statistical weights of the ground state of the atom and the negative ion, respectively. For iodine, which has a $2P^{3/2}$ doublet ground state with a degeneracy of 4, one has $g_0 = 4$, while the ions have a single state so that $g_- = 1$. Care must be exercised in applying this equation for the experimental case discussed below, because it is based on the assumption of a uniform surface, an equilibrium between the surface and the adsorbed atoms, and the presence of a single ionic and atomic species in the emitted current. The principal requirement for obtaining high ionization yields with negative surface ionization sources is to choose emitter surfaces with work functions smaller than 3 eV. In that case efficiencies of 40-100% for the halogens are predicted by eq. (4.27). Early attempts to use this effect in on-line mass separation of fission products [69] failed, probably because of too high a work function of the emitter. By taking advantage of the search for cathode surfaces with high thermionic emission properties carried out towards the end of the vacuum tube era, many surfaces with work functions ranging from 2-3 eV are available [70]. We have chosen LaB_6 to cover the internal surface of the tubular

surface ionizer for our first test of negative surface ionization. This material is particularly stable up to 1500°C, inert against the halogens, and has a work function in the range of 2.6-2.9 eV. Since this surface is a very efficient electron emitter, an intense electron beam unavoidably is extracted together with the negative ions. In order to avoid the possible problems with the electron beam, the electrons are separated from the ions by means of a magnetic field and dumped on a separate intermediate extraction electrode. In the first on-line test with this system we obtained a yield of 10% for bromine and an excellent long-term stability [71]. Work is now in progress to investigate the detailed properties of the source and other surfaces, but it is already clear that target and ion-source systems analogous to those of the alkali metals are feasible and that, by choice of proper surfaces, they possibly can be extended to the elements like S, Se, Te, and Po. Very high yields of negative molecular ions are reported in the literature [72,73] which open up possibilities for selective ionization also of other elements.

4.2.7 The noble gases

Noble gases emanate efficiently and selectively from a number of target matrices already at room temperature owing to their chemical inertness and high vapour pressure. It was therefore natural to choose these elements for the first on-line studies. Hydroxides of the elements Ti, Zr, Ce, and Th [38] were used very successfully as noble gas targets at ISOLDE-1. Since the loss of hydroxide water at ISOLDE-1 seriously limited the lifetime of the ion sources, these targets were abandoned at ISOLDE-2, where it is expected that this effect would become even more pronounced with the higher proton beam intensity. Instead, high production rate molten-metal targets and fast high-temperature solid-state targets with properties similar to those described for the alkali metal targets in section 4.2.3 were developed [31,47]. This incurred the disadvantage that the chemical selectivity was no longer granted by the evaporation process itself but had to be re-established by other means. In this case it is simply achieved by condensation or adsorption of all the other elements in a copper tube, inserted

between the target and the ion source, and kept at a temperature of 30°C. The discharge ion source used for ionizing the noble gases uses the Forced Electron Beam Induced Arc Discharge (FEBIAD) principle as developed by Kirchner and Roeckl [74] and is shown in fig. 4.7. This source has proved to be particularly useful for on-line use owing to its capability of working at pressures down to 2×10^{-5} Torr.

4.2.8 Mercury, cadmium and zinc

In the first-generation ISOLDE molten-metal targets, Pb, Sn, and Ge, for production of the elements Hg, Cd, and Zn, respectively [75], the chemical selectivity was ensured by the evaporation process itself, since no other elements with isobaric masses did evaporate. In fact the Pb target for production of Hg is one of the most successful ISOLDE targets in terms of physics output. Since it is also the most frequently employed target it has over the years undergone continuous development so that it today is one of the best understood targets. In its latest version, the Pb target is combined with the FEBIAD ion source, described in section 4.2.7, and a 50 Hz electromechanical shaking system, which moves the target axially with an amplitude of 0.2 mm. The ion-source yield is around 30%. The electromagnetic shaking described in ref. 31 is no longer in use, since the target is heated to its operating temperature of 800°C entirely by the proton beam current, so that the a.c. target-heating current flowing in the Pb itself is negligible or absent. As discussed in section 4.1.3, the shaking effect reflects the high sensitivity of the Pb target to oxygen contamination, a property which is shared by the Sn and Ge targets. The reason is probably that these three metals do not to any appreciable degree dissolve their own oxides, contrary to what is observed for all other (and, in fact, chemically more reactive) molten-metal targets. The oxide layers on Pb, Sn, and Ge eventually disappear during prolonged heating in vacuum, probably owing to dissociation and reduction. The delay-time distributions of these three targets exhibit a single exponential distribution typical for molten metals, with an average value of $\tau = 30$ s. Since these target elements as well as their heavier neighbouring elements do not form any

refractory compounds, it has not been possible to develop faster high-temperature spallation targets as in the case of many other elements. A reduction in the delay time may be obtained as indicated from the plot of $\log \tau$ versus $1/T$ [75] if the target temperature could be increased. The temperature of the lead target is limited by the Pb vapour pressure, but the possibility exists that intermetallic Pb compounds may exhibit a reduced Pb vapour pressure. Another interesting possibility is to make use of the ($^3\text{He}, \text{pxn}$) reaction on a molten-Au target which can be operated at a temperature of 1200°C .

4.2.9 Gallium, indium and thallium

Chemically selective arrangements for production of the group 3a elements, Ga, In, and Tl, with ionization potentials of 6.0 eV, 5.8 eV, and 6.1 eV, respectively, may in some cases be envisaged in combination with a high-temperature surface ionizer. Targets which release the neutron-deficient nuclei as formed by spallation of nearby elements have not been found for Ga and Tl, since the prospective heavier target elements have too high vapour pressure. A Sn spallation target for production of In, which makes use of chemical evaporation of In as InCl , is being developed. Preliminary tests of the release and ionization of Tl formed in a Au ($350 \text{ MeV } ^3\text{He}, \text{xn}$) reaction gave very promising results for the production of the light Tl isotopes with the $900 \text{ MeV } ^3\text{He}$ beam available at the CERN SC. Andersson et al. [76] and Chan et al. [77] have shown that the Ga and In fission products are readily released from a uranium oxide graphite matrix. Likewise it is shown that the neutron-rich nuclei of Ga, In, and Tl are released when formed by a high-energy proton reaction on the uranium carbide matrix described in section 4.1.2. In that case, however, a contamination from the isobaric neutron-deficient alkali isotopes, also formed in that reaction, cannot be avoided unless electrostatic suppression of the alkalis as described in section 4.2.3 is applied.

4.2.10 Chemical separation after the mass separation

In some cases, where an element separation cannot be performed in the target or the ion source, there still exists the possibility to single out one isobaric component from the mass-separated ion beam by means of a suitable separation

arrangement right in front of the measuring apparatus. The thermochromatographic separation method, which takes advantage of the differences in the vapour pressure of the products, and of their tendency to adsorb on a surface, has been successfully used on line by the Studsvik group [78,79]. There are other cases where such element separation is practical. For the case of short-lived radioactive daughters from originally isotopically pure samples, a simple apparatus for on-line separation of Hg and Pt has been developed [80]. The method which exploits the vapour pressure difference may produce platinum samples containing less than 0.1% of the original Hg activity within a separation time of 4 s.

4.2.11 Notations

The subscripts 0 and 1 refer to the target material and the product, respectively.

A	Avogadro's number	
c	Concentration	g cm^{-3}
D	Diffusion coefficient	$\text{cm}^2 \text{s}^{-1}$
D_e	Effective diffusion coefficient for diffusion in a porous system	$\text{cm}^2 \text{s}^{-1}$
D_i	Ion-source diameter	cm
d_t	Transfer-tube diameter	cm
$F(t)$	Fractional activity	
J_e	Release rate	$\text{atm cm}^{-2} \text{s}^{-1}$
J_d	Diffusion rate	$\text{atm cm}^{-2} \text{s}^{-1}$
ℓ_t	Length of transfer tube	cm
L_i	Length of ion source	cm
L_e	Capillary length	cm
M	Molar weight	g g-mole^{-1}
N	Nuclear-reaction rate	s^{-1}
P	Saturated vapour pressure	atm
p	Partial vapour pressure	atm
$p(t)$	Delay-distribution function	

Q_p	Activation energy for desorption	eV
Q_s	Activation energy for diffusion	eV
R	Gas constant	eV K ⁻¹ mol ⁻¹
r	Particle radius	cm
S	Surface area	cm ²
S_i	Ion-source outlet area	cm ²
t	Time	s
t^0	Pre-exponential factor in the Frankel equation	s
T	System temperature	K
$T_{1/2}$	Radioactive decay half-life	s
V	Volume	cm ³
x	Molar fraction	
Y	Decay-loss-corrected radioactive beam intensity	s ⁻¹
Y_{obs}	Observed radioactive beam intensity	s ⁻¹
z	Surface film thickness	cm
α	Evaporation coefficient	
λ	Radioactive decay constant	s ⁻¹
μ_ℓ	Delay constant for liquid-metal targets	s ⁻¹
μ_s	Diffusion parameter for solid targets	s ⁻¹
μ_s^0	Pre-exponential constant in the Arrhenius equation	s ⁻¹
ρ	Density	g cm ⁻³
τ	Average delay time $1/\mu_\ell$ for molten targets	s
τ_0	Average delay time for liquid systems as calculated from eq. (4.19)	s
ϕ	Probability for product re-entry into the target liquid	

5. EXPERIMENTS WITH FAR UNSTABLE NUCLEI

It has often been mentioned that about 2000 beta-unstable nuclei with half-lives longer than 1 ms remained undiscovered and that it would be of great interest to develop experimental methods that would make all these nuclei accessible for study. Especially the extension of the spectroscopy studies to the nuclei furthest

away from stability close to the proton and neutron drip-lines was one of the main motives behind most on-line isotope separators.

Now, more than a decade later, the techniques as described in the previous section have made a fair number of them available; often in such intensities that new experimental techniques could be used for their study. However, to identify them all has until now been far beyond our possibilities. Instead our efforts have been centred on experiments with the elements where the target techniques have been most successful, i.e. those which provide the highest yields and shortest delays. In the development work, we are directing our attention to possible new systems that could bring us to particularly interesting new regions like that of the expected doubly magic ^{100}Sn .

How far from stability we can get depends critically on a number of parameters. In comparing production methods it is often seen that emphasis is only placed on the reaction cross-section of the nuclides of interest and the incident particle intensity. However, the differences in range and other properties of the bombarding particles and the level of technical development of the target and ion-source system play an equally important role. The useful target thickness, the over-all separation yield expressed in terms of ion-source efficiency, and delay time losses are the pertinent factors to be taken into account when production yields far from stability are to be discussed. The prospects for detecting rare nuclei further depend strongly on the chemical selectivity of the target and ion-source system, the resolution of the mass separator, and the sensitivity of the detector system. In this section is discussed to what extent the above-mentioned goals have been reached, the limitation to detection methods and further identification of unknown nuclei, and the prospects for possible extension of proton- and ^3He -induced reactions to new regions of nuclei.

The experiments chosen to illustrate the state of the art represent only a few examples of the relevant work. The many experiments pursued with far unstable nuclei are covered in detail in the review paper by Hansen [16].

5.1 Yields and purity

In the present work the on-line testing of a new target system consists of a measurement of the entire isotopic yield curve as well as the delay-time distribution as described in section 4.1. In principle, the on-line mass separator is an almost ideal tool for the systematic study of nuclear reaction cross-sections [81], but so far there has only been a marginal preoccupation with this work owing to the conflict between our usual thick production targets and the thin targets required for reaction studies. A full account of the uncertainties involved in the present yield measurements are given in refs. 40 and 47.

The production yields as observed in the collector plane of the separator are given by

$$Y_{\text{obs}} = \phi N \sigma \epsilon \eta , \quad (5.1)$$

where ϕ is the proton beam intensity, N the number of target atoms/cm², σ the cross-section, ϵ the efficiency of the release, transfer and ionization processes, and η the correction for the delay losses as described in section 4. It must especially be remarked that owing to the long range of the 600 MeV protons, N may be of the order of 1 mol/cm² or 6×10^{23} atoms/cm², and beam intensities of $3 \times 10^{13} \text{ s}^{-1}$ can be used without major problems.

The system efficiency ϵ varies between 10 and 90% according to the element, and the delay time losses $\eta = Y_{\text{obs}}/Y$ for the fastest targets rise to $\sim 90\%$ for a nuclide with 100 ms half-life, as seen from fig. 4.8.

The isotopic yield distribution of an element Z formed by spallation of an element $Z + 2$ peaks at a cross-section of about 20-50 mb, as shown for the case of Hg from Pb in fig. 5.1. The purity of the samples depends on the discrimination in the target and ion-source unit against elements with isobaric masses and the optical properties of the separator. Shown in fig. 5.1 is the upper limit for contamination by the somewhat volatile element Tl. The curve is estimated by assuming the same reaction cross-section and ionization yield for Tl as for Hg and a discrimination factor of 10^6 equal to their partial vapour-pressure ratio at a target temperature of 800°C. The mutual contamination of the Hg isotopes is ultimately determined by the separator enhancement factor

$$n = \frac{C_{fw} C_{ip}}{C_{iw} C_{fp}}, \quad (5.2)$$

where C_{fw} is the final concentration of the wanted isotope, C_{iw} its initial concentration, and C_{fp} the final concentration of a parasitic isotope with an initial concentration C_{ip} . The good enhancement depends upon the resolving power of the separator

$$RP = \frac{DM}{b_{1/2}}, \quad (5.3)$$

where D is the mass dispersion and $b_{1/2}$ the full width at half maximum of the ion beam with mass M as measured in the focal plane. Depending on the conditions of operation, the resolving power of the ISOLDE machine varies between 400 and 1000. By separation of an element with many isotopes present, as in the case of on-line mass separation, the contamination of a given mass is caused by the sum of the contributions from all the others. It is, however, convenient to discuss the contamination of a given mass in terms of the general background which appears to be uniformly spread over all mass positions. This is mainly caused by gas scattering and the contamination from neighbouring masses and is determined by the number of ion-optical factors which contribute to the resolving power. A contamination of 3×10^{-5} of the yield of a given isotope is found uniformly distributed across the focal plane as illustrated by a horizontal line for one of the maximum yield nuclides in fig. 5.1. The neighbouring mass contamination to lighter mass is illustrated in fig. 5.1 by a yield curve shifted by one mass unit to the neutron-deficient side situated at a level of 4×10^{-4} of the measured yield.

5.2 Detection methods and limits

From fig. 5.1 it is clear that mainly the contribution from the uniformly spread contamination determines the purity of the ion beams at the extreme wing of the yield distribution. In the case of the Hg isotopes in fig. 5.1 single beta or gamma spectroscopy becomes exceedingly difficult from mass 181 and outwards. The use of detection systems with high selectivity becomes essential for

the study of lighter nuclei. For elements with $Z \geq 69$ the onset of alpha decay for very neutron deficient nuclei becomes important, and their detection by surface barrier detectors is now only disturbed by the much lower neighbouring mass contamination.

The energy available for beta decay increases away from stability so that states in the daughter nuclei above the nucleon separation energy can be populated. The de-excitation may now proceed either via the emission of nucleons or groups of nucleons or via gamma emission. This phenomenon of beta-delayed particle emission serves, together with surface-barrier detectors, telescopes and neutron counters, as largely background-free and thus very sensitive detection methods for exotic nuclei in all mass regions. In fact, the highest detection sensitivity is obtained in charged particle detection, where it is possible to work with one decay per hour or 10^{-3} s^{-1} . In cases where the above-mentioned characteristic particle emission is absent, the high Q-values may serve as a characteristic "finger-print", as in the case of the light Rb and Kr isotopes, where values in the vicinity of 10 MeV lie much above those of likely contaminants and daughters. By means of a thick plastic scintillator operated in coincidence with a thin ΔE detector, the background of the E detector is considerably reduced. By analysis of the high-energy part of the beta spectrum recorded by the telescope, the presence of fast decaying nuclides can be determined. The detection limit is about 10^{-2} s^{-1} [82].

With the maximum obtainable proton beam intensity at the SC of $3 \times 10^{13} \text{ s}^{-1}$, an ion-source efficiency of 0.1, a target thickness of $6 \times 10^{23} \text{ atoms/cm}^2$ and assuming $\eta = 1$, the detection limit obtained from eq. (5.1) varies from 10^{-39} to 10^{-40} cm^2 , depending on the detection system.

5.3 The role of proton beam intensity and other particles

The increase in proton beam intensity of close to a factor of 100 provided by the reconstruction of the SC has, together with an estimated improvement of a factor of 10 due to the target technology, opened up a number of experimental possibilities as discussed in this and the following sections. From the discussion in section 4, it does not seem to be excluded that target and ion-source systems for beams of 20-100 μA could be developed. Since such beams are available at

several accelerators in the world, it is interesting to analyse the perspectives for their use. The yield curve in fig. 5.1 shows that the drop in yield at the extreme wing of the curve is two orders of magnitude for each mass number, so that proton beam increase of a factor of 100 would hardly bring us one mass unit further away from stability. One motivation in utilizing such high intensities would be to obtain higher amounts of very exotic nuclei, such as ^{32}Ar which is extremely hard to produce. However, the developments in ^3He beams and heavy-ion beams at lower energies seem to indicate substantially higher production yields for these nuclei than can be obtained with protons. Secondly, elements for which only low-yield target and ion-source systems can be envisaged may come into use by compensating with a high intensity beam. In any case the radioactivity problems in the target area would become quite formidable. With the present 2-5 μA proton beam there are already regions of the nuclidic map where 600 MeV protons, which usually, from a pure reaction cross-section viewpoint, were considered unfeasible, now become attractive production particles. The particularly versatile uranium carbide target discussed in section 4.1.2 allows high yields of fission products. In fig. 5.2 [83-85] are shown some typical maximum Cs yields obtained from on-line separator experiments with different particles which represent the state of the art. For the moment it seems clear that the heavy-ion reaction gives the highest yields at the extreme neutron deficient wing. For the time being ISOLDE technique is far superior in intensity on the neutron-rich side as compared with the use of nuclear reactors. The lower formation cross-sections in high-energy fission are outweighed by the combined effect of increased target thickness, higher beam intensity, and improved ion-source efficiencies. An extension of the ISOLDE experimental programme to neutron-rich nuclides has already been started. Another example where the uranium carbide target may come into use is for the production of neutron-rich nuclei of elements lighter than Ca. They are usually produced by fragmentation of uranium by protons. Since the threshold for this reaction is around 1 GeV, experiments with the heavy Li, Na, and K nuclides, as performed by Klapisch [81] have been made with 24 GeV protons, for which the cross-sections seem to be somewhat higher.

Shown in fig. 5.3 are the observed yields of K and Na nuclides as produced by fragmentation of ^{238}U with 600 MeV protons and for K also by spallation of Sc [86]. It is seen that for potassium the limit for production has been pushed out to ^{53}K and all previously known sodium isotopes are produced. It is now possible to make useful yields at ISOLDE of the neutron rich Ca, K, Ar, Cl, S, Na, Ne, Li and He nuclei.

The broad mass distribution of high-energy fission may also be used for production of neutron-rich products outside the thermal neutron-induced fission region like the heavy rare earths [47]. Another possibility of making neutron-rich nuclei outside the fission-product region is to use fast secondary neutrons generated in a converter target from high-energy protons; Katcoff et al. [87] found quite high cross-sections of neutron-rich species formed in $(n,2p_n)$ reactions. At ISOLDE one experiment has combined the fast neutron technique with mass separation. The first part of a 1 kg molten-Pb target served as a converter. The new isotope ^{207}Hg with a half-life of 2.9 min had a sufficiently high yield in the reaction $^{208}\text{Pb}(n,2p)$ to permit Nielsen and his group [88] to study the levels in ^{207}Tl (one proton removed from the doubly magic ^{208}Pb).

In Spring 1978 the first 910 MeV $^3\text{He}^{++}$ beam with an intensity of 0.8 μA was extracted from the SC and directed onto an ISOLDE target. Yield measurements of spallation and fission products from a uranium carbide target were carried out and indicated a very substantial increase of the yields for the light Fr isotopes shown in fig. 5.4. A first test of a Au target for the production $^{197}\text{Au}(^3\text{He},xn)\text{Tl}$ gave a maximum yield of about 10^7 atoms/s at mass 191 and permitted the observation of ^{183}Tl . If these features are confirmed with other targets it will have important consequences for future experiments.

5.4 Identification of unknown nuclei

A natural step in the testing of improved target and ion-source techniques is to determine to what extent new regions of nuclei have been made accessible. The total number of new nuclides which have been identified through ISOLDE work has exceeded 70. The alkali and the alkaline earth metals have been particularly

intensively studied at ISOLDE. In the case of Cs the limit for particle stability has been established [89] at ^{114}Cs as formed in the reaction $^{139}\text{La}(p,3p23n)^{114}\text{Cs}$ with a cross-section of 10^{-36} cm^2 . It was especially interesting that from 114-118 most of the Cs nuclides were delayed alphas or proton precursors, which allowed a first systematic study of a family of odd-Z precursors [89]. A search for ^{113}Cs , which by most mass formulae is predicted to be unstable towards ground-state proton emission, was negative. An upper limit for its production of 10 atoms/h or a formation cross-section of 10^{-39} -- much lower than expected from the semi-empirical spallation formula [20] -- led to the conclusion that at mass 113 one has reached the proton drip line for caesium. This is also in agreement with experiments at GSI [83], where despite much higher production yields (see fig. 5.2), the search for ^{113}Cs has been unsuccessful. As reviewed by Jonson [15], studies at ISOLDE have added significantly to the knowledge of beta-delayed particle emission from the heavy elements. With the development of the fast high-temperature carbide targets discussed in section 4.1.2, it is now possible at ISOLDE to study extremely proton rich nuclei in the light elements $Z < 20$. A first example is the delayed proton emitter ^{32}Ar , $T_{1/2} \approx 75 \text{ ms}$ [90]. Unlike the heavier proton precursors, one deals here with spectroscopy of individual levels. The isotope $^{32}_{18}\text{Ar}_{14}$ is the first observed nucleus with isospin projection $T_Z = -2$ and it is the most neutron deficient nucleus ever to have its radioactive decay recorded. The experiment permitted an accurate test for a cubic term in the isobaric-multiplet mass equation as applied to narrow $T = 2$ states.

The study of nuclei along the $N = Z$ line has now reached the heaviest self-conjugate nucleus detected until now by the observation of the beta decay of ^{74}Rb ($T_{1/2} = 64.9 \text{ ms}$), (see fig. 5.5). In the same experiment [82] an upper limit of 10^{-3} relative to ^{74}Rb or a formation cross-section of 10^{-36} cm^2 could be placed on the yield of the unobserved mirror nucleus ^{73}Rb . It is in fact likely that the limit of particle stability has been reached here for rubidium.

The heaviest known $T_Z = \frac{1}{2}$ nucleus ^{71}Kr has recently been identified. The half-life of this mirror nucleus $T_{1/2} = 102 \text{ ms}$ provides a possibility of extracting

the Gamow-Teller matrix element. Following the number of new targets taken into use over the last few years of ISOLDE the number of new identified alpha emitters has been steadily increased. Contributions to mass-determinations, measurements of transition probabilities, and other spectroscopic information in the heavy mass region are discussed in ref. 91. The earlier described high-temperature Ta target described has been further improved and gives now the yields shown in Fig. 5.6. Normalized to the cross-section for formation of ^{169}Yb [92], and corrected for decay losses according to eq. (4.11), the yield curve allows the deduction of the approximate isotopic cross-section distribution. It is interesting to remark that the last identified alpha precursor [93] ^{153}Yb , formed in the reaction $^{181}\text{Ta}(p,4p25n)^{153}\text{Yb}$ with a cross-section of 10^{-34} cm^2 , still is produced in a yield of more than 10^3 s^{-1} . The shortest lived isotopes studied at ISOLDE are found among the Fr alpha emitters [94], where the last identified ^{201}Fr $T_{1/2} = 48 \text{ ms}$, found in the reaction $^{238}\text{U}(p,6p32n)^{201}\text{Fr}$ with a cross-section of 10^{-35} cm^2 , is close to the mass-formula-predicted proton drip-line. The half-life of the more abundantly produced ^{211}Fr has been remeasured to be $27.5 \pm 2 \text{ ms}$. In a search for the reported $0.7 \pm 0.6 \text{ ms}$ ^{218}Fr it was found that there probably is an isomeric state in ^{218}Fr with 21 ms half-life, which decays to ^{214}At and only has a weak branch to the 0.7 ms ground state of ^{218}Fr . An α - α coincidence measurement on ^{214}At showed that, contrary to the estimated half-life given in the literature [95], there are at least two isomers in ^{214}At with half-lives of 790 and 565 ns.

Whereas the proton drip line has been reached for several elements as discussed above, the neutron drip line, which lies much further away from stability, seems for the heaviest case to be reached by the heaviest known isotope of the element lithium. This ^{11}Li , which was first identified by Roeckl et al. [96] in a uranium fragmentation reaction with 24 GeV protons, has a half-life of 8.5 ms and is bound against the emission by two neutrons by 170 keV. In a recent experiment at ISOLDE [97], this nuclide was produced in a yield of 15 atoms/s by irradiating the 13 g/cm^2 ^{238}U target with 1.6 μA protons. The beta-delayed neutron spectrum as recorded with high-resolution ^3He -filled neutron spectrometers presented in fig. 5.7 shows strong evidence of the first observation of beta-delayed two-neutron emission.

The study of nuclear excited states by the observation of the emitted photons or conversion electrons is being pursued at ISOLDE by means of a number of experimental techniques. Energies and intensities of lines are extracted from the direct spectra recorded with high resolution Ge and Si detectors and magnetic spectrographs. In particularly interesting cases complete level schemes of far unstable nuclei are built up on evidence from energies, intensities or coincidence experiments. An important use of such data is seen in the case of ^{118}Xe studied by Genevey-Rivier et al. [98] at ISOLDE. In recent attempts to establish a connection between the interacting boson model and the microscopic behaviour of nuclei, Arima and Iachello [99] predict the presence of the dynamical symmetries SU(5) in nuclei. In ^{118}Xe , the French ISOLDE group found all three members of the triplet, all five members of the quintuplet, and seven members of the higher multiplet, as predicted by the model.

In the course of this work we have only been able to exploit a few of the elements made accessible by the development work described in section 4. In spite of the very steep slope of the cross-section distributions caused by the rapid fall in proton binding energy we have for several elements been able to approach or reach the limit where the last proton is kept within the nucleus only by the effect of the Coulomb barrier. Thus even under improved experimental conditions we can hardly expect to reach further out than at most one unit in mass number to the neutron-deficient side.

6. THE USE OF RADIOACTIVE BEAMS OF HIGH INTENSITIES

An outstanding feature of an on-line separator connected to a high-energy proton accelerator is the possibility to produce very strong beams of mass-separated radioactive ions. In the effort of lifting up the extreme neutron deficient wing of the yield distribution (see fig. 5.1) by means of increased proton beam current and target development, the abundance of the beams in the region closer to stability has in many cases reached the level of 10^{10} - 10^{11} atoms/s. Such nanoampere beams make it possible to prepare mono-isotopic sources of the order of curies implanted to a depth of $10\text{ }\mu\text{g}/\text{cm}^2$ in a metal backing within an

area of only 0.5 mm^2 . These beams and sources are ideally suited for precision measurements of a number of ground-state properties in nuclei away from stability. Experimentally this new field which is mostly based on methods from atomic physics allows the study of the nuclear properties systematically over many nuclei differing in neutron number. They generally require beams of more than 10^6 atoms/s, but it is a typical trend that the experimental methods are continuously developed, so that some of them may already extend the measurements to much weaker beams.

6.1 Atomic beam magnetic-resonance measurements

The hyperfine structure measurements of short-lived nuclei by means of a conventional atomic beam magnetic resonance system (ABMR) are only possible for nuclei with production yields above 10^6 atoms/s, because of the low transmission of such an instrument. This requirement is fulfilled for a number of ISOLDE-produced nuclei and the Gothenburg-Uppsala group [100] has for the first time connected an ABMR apparatus on line to a mass separator. The crucial problem in the on-line ABMR apparatus, which is shown in fig. 6.1, is the efficient conversion of the 60 keV ion beam into a thermal atomic beam. In the conventional off-line apparatus the atomic beam is formed by means of a channel oven. Owing to the effusion losses through the ion beam entrance aperture and an increased delay time, the on-line experiments are performed with a flat ribbon "oven" heated by resistance heating. This gives a factor of 10 higher losses as compared to the off-line channel ovens. By means of this combined set-up, an impressive number of nuclear spins and moments in the elements Rb, Cs, Fr, and Au have been measured. A common feature of these elements is that they have stable isotopes close to a neutron shell closure ($N = 50, 82$, and 126) and their neutron-deficient isotopes extend into regions where nuclear deformation is expected. The measured nuclear spins and moments may be used to probe the change in the structure of the nucleus. In fact the experimental data give evidence for strongly deformed nuclear shapes of the light Cs isotopes [101].

6.2 Optical spectroscopy

Measurements of spins, moments, and optical isotope shifts have been performed on line for isotopes of mercury by the group of Otten [102]. By means of

their extremely sensitive isotope shift measurements, based upon beta-radiation-detected optical pumping, a sharp onset of nuclear deformation has been discovered [103] between ^{187}Hg and ^{185}Hg . This new region of deformed nuclei is interpreted as a transition from a slightly deformed oblate shape ($A \geq 187$) to a strongly deformed prolate shape ($A \leq 185$). Evidence for a coexistence and crossing of two bands, one built on an almost spherical shape and one built on a strongly deformed shape, has been found by gamma-spectroscopic measurements on the $I = 0$ isotopes ^{188}Hg [104,105], ^{186}Hg [106,107], and ^{184}Hg [108]. A strong odd-even nuclear shape staggering as shown in fig. 6.2 has recently been measured by the Otten group [109] in an isotope shift measurement of the even Hg nuclei by the purely optical technique of fluorescence excitation by means of a tunable dye laser.

This group is at present preparing laser spectroscopy on atomic and ionic beams from ISOLDE in a collinear geometry as shown in fig. 6.3. The advantage of this method is that it uses the velocity bunching phenomena to make Doppler free spectroscopy directly on the accelerated beam as demonstrated for the alkalis at the Mainz on-line separator [110]. The Doppler width, limited by the thermal energy spread of the ions, was reduced to the order of the natural line width of typical resonance lines, since the velocity distribution along the beam accelerated through a potential difference U is narrowed by a factor $R = \frac{1}{2} (kT/eU)^{\frac{1}{2}}$. For $T = 1500$ K and the ISOLDE 60 keV beam, $R = 7 \times 10^{-4}$. In addition this technique is expected to be 10^4 times more sensitive as compared to laser excitation of a collimated thermal atomic beam.

A group from Orsay has made a large series of laser hyperfine spectroscopic measurements on the alkali isotopes produced at ISOLDE [111]. By converting the beam into an atomic beam, as in the ABMR measurements, and optically pumping it with a laser, the populated spin states can, after analysis in a six-pole magnet, be detected in a mass spectrometer, as shown in fig. 6.4. The element Fr represented here a special problem, since the atomic excited levels were not known. In fact it is the last naturally occurring element, which, since its discovery in 1939, has resisted all attempts to determine its optical spectrum. The difficulty is obvious. The longest lived isotope ^{223}Fr ($T_{\frac{1}{2}} = 22$ min) occurs only in a weak

1.4% alpha-decay branch of ^{227}Ac which is a member of the actinium ($4n + 3$) series starting with ^{235}U . In fact 3 t of natural uranium contain in equilibrium only 10^{-11} g of Fr. But most experiments with Fr have only been made with not very pure samples of 10^{-13} to 10^{-15} g. ISOLDE is a continuous source of about 10^9 atoms/s of chemically and isotopically pure Fr, which allows a total of 4×10^{-10} g to be collected. With these amounts the Orsay group [112] succeeded in locating the D_2 resonance line of Fr at 717.7 nm, which allowed them to start laser spectroscopy of the Fr isotopes.

6.3 Experiments with radioactive targets

The radioactive beams with intensities in the nanoampere region allow the collection of 10^{12} - 10^{16} atoms of isotopes with half-lives ranging from minutes to days. Collected on a few mm^2 they correspond to a thickness of several $\mu\text{g}/\text{cm}^2$. As pointed out by Bondorf [113] already in 1966, this is sufficient for certain reaction-spectroscopy experiments with neutrons and beams from Van de Graaf accelerators. Especially interesting is the possibility to extend the range of targets with odd neutron and proton number for which comparatively little is known about the excited states since nature only provides a few stable ones. The first experiment of this kind has studied the capture of low-energy neutrons in ISOLDE-produced nuclei. In a joint experiment with the ILL high-flux reactor team in Grenoble a program to study the (n_{th}, p) and (n_{th}, α) reactions by irradiating a series of radioactive targets with a beam of thermal neutrons has been started. The target with the shortest half-life ($T_{1/2} = 16$ h) from which charged particles were observed consisted of 5×10^{12} atoms of ^{76}Br [114]. In order to extend these measurements to nuclei with shorter half-life, an on-line irradiation with a neutron generator is considered.

Experiments using radioactive targets in tandem Van de Graaf accelerators are in preparation.

6.4 Hyperfine effects in solids

The wide range of nuclei accessible at ISOLDE makes it an ideal location for the investigation of hyperfine interactions of impurities in solids by means of implanted radioactivity. In fact, ion implantation is now one of the standard

methods in this field. The properties of interest are the magnetic and electric moments, the magnetic and electric hyperfine fields, the isomer shifts and the Debye-Waller factors for the implanted impurity atoms. During the last few years such effects have been studied by means of several experimental techniques. In off-line measurements of nuclear orientation in a magnetic field at very low temperatures the ground state magnetic moments of the long-lived platinum isotopes ^{189}Pt and ^{191}Pt have been measured [115]. In fact, recent developments [116] show now the feasibility of on-line nuclear orientation experiments by means of a ^3He - ^4He dilution refrigerator. A series of experiments [117] with on-line time-differential perturbed γ - γ angular correlations aims at the determination of the electric field gradient resulting from the implantation of cadmium in gallium and antimony and the confirmation of the existence of a substitutional lattice site. Recently this field has been extended to Mössbauer spectroscopic measurements [118]. Apart from the high source strengths which, dependent on the detection method, are required by the Mössbauer technique and the possibility of extension to short-lived Mössbauer transitions, the on-line implantations have a particular advantage. By implantation of the beta-decay precursors of the Mössbauer nuclide, systematic studies of the dynamics in the host lattice of a series of impurity atoms may be performed. In the case of the Mössbauer transition of ^{119}Sn populated from members of the decay chain $^{119}\text{Cs} \rightarrow ^{119}\text{Xe} \rightarrow ^{119}\text{I} \rightarrow ^{119}\text{Te} \rightarrow ^{119}\text{Sb} \rightarrow ^{119}\text{Sn} \leftarrow ^{119}\text{In} \leftarrow ^{119}\text{Cd} \leftarrow ^{119}\text{Ag}$, the semiconductor relevant elements I, Te, Sb, In and Cd can be studied [118].

6.5 Shifts in the energies of K X-rays

The interest in the study of X-rays has recently been revived and in the last decade a number of new phenomena have been discovered in X-ray physics as reviewed by Dyson [119]. It has been shown [120,121] that the energies of K X-rays from the heavier elements to a small extent depend on the isotopic and chemical composition of the source. The isotope shifts originate in the nuclear finite size effect and serve to give information on nuclear radii, while the chemical shifts come from changes in the screening due to the valence electrons.

In 1976 experimental evidence was reported by a group from CalTech [122] that the energies of Ta K X-rays depend on the excitation mechanism: the rays induced by photo-ionization of Ta have about 1 eV higher energy than those originating in the electron-capture beta decay (EC) of ^{181}W . The experiment was interpreted as demonstrating a selective shake-off of 4f electrons in the beta-decay process; it was soon pointed out that there are strong theoretical reasons [123] to expect outer shell shake-off to be very weak in the EC process and that the sign of the CalTech measurement in fact was consistent with that expected for shake-off in the photo-ionization process. The quantitative explanation of this measurement still remains an interesting puzzle. For these effects, the characteristic energy shifts are 10^{-2} - 10^{-3} of the natural line width; consequently such experiments require not only high intensities but also special precautions to eliminate systematic errors.

The β -decay of the neutron-deficient nuclides proceeds either by emission of a positron or by the process of orbital electron capture (EC). Since the latter alternative creates an inner shell vacancy the subsequent production of X-rays is a consequence of this decay mode. The emission of the characteristic X-rays is a widespread phenomenon in the ISOLDE-produced nuclei, except perhaps in the lightest elements where the fluorescence yields are very low. The obtained sources are ideally suited for precision measurements by means of a curved-crystal spectrometer of the DuMont type, which provides the highest obtainable transmission. In a forthcoming paper we describe such a spectrometer which has been developed and built by the CERN-Jülich group and installed at CERN in order to carry out detailed studies of various effects which give rise to small K X-ray shifts and where essential use is made of the strong sources produced by ISOLDE.

6.5.1 The crystal spectrometer

The instrument, which is schematically shown in fig. 6.5, is equipped with a 4 mm thick quartz crystal, which is reflecting the photons at the (110) plane ($d \approx 2.5 \text{ \AA}$) through a window of 5 cm diameter. The crystal is bent between clamping blocks to a focal length of 4.64 m. The angular setting is accomplished

with a double sine screw, the position of which is controlled through an electronic regulation circuit combined with an angular decoder. The detector is a thin NaI(Tl) scintillation counter, which is shielded against the undiffracted beam by means of a Soller slit collimator. An optical control unit keeps it in the correct position with respect to the diffracted X-ray beam, by means of a laser beam. The new and essential feature of the instrument is the source arrangement. In order to minimize the systematic errors and achieve a high relative accuracy, the source holder is double and equipped with a shutter system as shown in fig. 6.5. This system, which permits only one source to be seen by the crystal at a time, allows the study of two different sources relative to each other. The reflections of the rays from the two sources are recorded point by point as a function of the Bragg angle θ in an alternating manner, since the shutter gates the detector signal several times per minute to separate scalers. The source arrangement allows use of sources from ISOLDE, from neutron capture, and from fluorescence excitation of inactive foils by means of a 40 Ci ^{169}Yb source.

Still the accuracy of the position of the two sources does not suffice for a determination of the energy shift from one scan alone, since a 3 μm displacement of one source relative to the other in the direction of the dispersion typically is equivalent to a 10 meV energy shift. This effect is balanced out by scanning both the positive and negative Bragg angles θ . By applying corrections for the instrumental asymmetry and an analysis of the data which is independent of small changes in the line shape, as described elsewhere [124,125], a precision of 3×10^{-7} of the K X-ray energy (or 3×10^{-4} of the natural line width) has been reached.

6.5.2 New effects influencing the energies of K X-rays

In the first measurement on the pair of isotopes $^{131},^{132}\text{Cs}$, a shift of 112 ± 11 meV was found [124]. This difference in the energy of the Xe K X-rays is interpreted as being due to a selection rule in EC beta decay, which gives a non-statistical population of the atomic hyperfine levels as illustrated in fig. 6.6. As the magnetic moments of the final levels (in isotopes of Xe) are known, it is possible to calculate a theoretical hyperfine shift of 113 ± 6 meV,

to which should be added 13 meV as the estimated contribution from isotope and isomer shifts. There is thus good agreement with the experiment, which represents the first observation of $1s$ hyperfine shifts and provides a new tool for measuring magnetic moments of nuclear states, even with extremely short half-lives.

In a second experiment [126], the energies of the Au $K_{\alpha 1}$, $K_{\alpha 2}$, and $K_{\beta 1}$ produced in the EC decay of ^{197}Hg were compared with those obtained by fluorescence excitation of a Au foil by means of a 40 Ci ^{169}Yb source. Of the resulting ~ 600 meV shift, a 200 meV shift persists even after allowance has been made for contributions from the hyperfine effect and from a small nuclear volume effect, which could be evaluated from Mössbauer data. The reason for this additional shift is tentatively interpreted as being due to the different probabilities for outer-shell shake-off by the two excitation mechanisms. In the fluorescence excitation process the resulting unresolved satellite lines as shown in fig. 6.7 play an important role, while the gentle EC decay gives a near absence of outer-shell excitation and thus no observable satellite shift.

Since the lifetime of a $1s$ hole is extremely short (10^{-16} – 10^{-17} s), the atom which by EC decay is transformed from an element ($Z + 1$) into an atom of the element Z retains the configuration of the mother atom until the K X-ray is emitted. In the simplest case this will mean that the (daughter) X-ray is emitted in the presence of an additional electron in a certain orbit nl_j , so that the energy shift will be identical to that shown in fig. 6.7, but of opposite sign. This atomic structure effect is expected to be particularly large when inner shells are filling. An example of the role played by all three effects mentioned above is demonstrated using the pair of rare earths Er and Ho, which are believed to have the electron configurations Er ("Xe core" $6s^2$, $4f^{12}$) and Ho ("Xe core", $6s^4$, $4f^{11}$). In this third experiment [127] the energies of photo-ionization X-rays from Ho metal were compared with those from a neutron-capture-produced metallic ^{165}Er source. An important point here is that the two metals have essentially the same crystal structure and lattice parameter; therefore it seems reasonable to assume that chemical shifts will be entirely absent. The experimental results are shown in fig. 6.8. A remarkable feature of the measurement is that it has been possible

to measure also the weak lines originating in the outer shells, so that a "fingerprint" characteristic of the additional electron is obtained. When the effect of this extra 4f electron present after the EC decay (to Er) is taken into account (in a Hartree-Fock-Dirac calculation) good agreement is obtained between theory and experiment. For the Hg-Au case, discussed above, a (much smaller) 6s effect is expected; it turns out that when it is taken into account the agreement is quantitative.

The present investigations have brought out a number of new effects which indicate that further studies may be very fruitful. It seems interesting to concentrate the new experiments on the atomic structure effects and to extend the measurements to other pairs of elements, notably those in the actinide region. An entirely open question is the extent to which solid-state effects play a role. The possibility at ISOLDE of implanting very large amounts of radioactivity should provide unique opportunities for investigating the implant-host relationship.

7. CONCLUSION AND OUTLOOK

As can be noted in the foregoing discussion, the field of high-intensity radioactive beams is a very rapidly progressing one. New experimental approaches have opened up the possibilities for many experiments, of which only a few have been pursued at the time of writing this article. Active discussions on the future prospects belong to the daily life in this field. On the production side the 86 MeV/amu ^{12}C beam under preparation at the SC may make new target-projectile combinations possible, thereby enlarging the range of available elements, and possibly lead to increased yields of products far from stability. The latter effect clearly can be achieved by means of an increased proton beam intensity.

A new phase of the research possibilities may be represented by the idea of using accelerated radioactive nuclei produced by an on-line separator for nuclear reaction studies. This idea, which already occupied the concluding discussions at the Lysekil Conference in 1966 [113], has since fascinated many physicists. Mallory and McNeily [128] propose using a heavy-ion-based on-line separator as an injector to a heavy-ion isochronous cyclotron, in order to obtain accelerated

radioactive beams with an intensity of 10^5 s^{-1} . However, the basic limitation for the physics which one could hope to do with such an installation is intensity.

In the light of the technical developments described in this paper, it is clear as pointed out by Hansen [129] that only in conjunction with a modern high-energy proton accelerator may useful beam intensities, i.e. 10^{10} s^{-1} , be achieved. The time has now come where this is technically feasible and the kind of physics that can be done with such beams should be considered.

In conclusion, one can say that this is still a large and fascinating field where further research inevitably will pay off in the future.

REFERENCES

- [1] O. Kofoed-Hansen and K.O. Nielsen, K. Danske Vidensk. Selsk. Mat.-Fys. Medd. 26 (1951), No. 7.
- [2] J. Koch, Lecture at the Second Scandinavian Isotope Separator Symposium, Uppsala, 1955 (unpublished); G. Andersson, private communication.
- [3] O. Kofoed-Hansen, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 65.
- [4] G. Rudstam, Nucl. Instrum. Methods 38 (1965) 282.
- [5] I. Bergström, Nucl. Instrum. Methods 43 (1966) 116 and 43 (1966) 129.
- [6] Proc. Internat. Symposium on Why and How should we investigate Nuclides Far Off the Stability Line, Lysekil, Sweden, 1966 (eds. W. Forsling, C.J. Herrlander and H. Ryde), Ark. Fys. 36 (1967) 1.
- [7] Proc. Internat. Conf. on Electromagnetic Isotope Separators and the Techniques of their Applications, Marburg, 1970 (eds. H. Wagner and W. Walcher), Bundesministerium für Bildung und Wissenschaft Forschungsbericht BMBW-FB K 70-28 (1970).
- [8] Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Gothenburg, Gothenburg, 1973).
- [9] Proc. Internat. Conf. on the Properties of Nuclei Far from the Region of Beta Stability, Leysin, 1970, CERN 70-30 (1970), 2 Vols.
- [10] A. Kjelberg and G. Rudstam (eds.), CERN 70-3 (1970).
- [11] Aa. Bohr and B.R. Mottelson, K. Danske Vidensk. Selsk. Mat.-Fys. Medd. 27, (1953), No. 16.
- [12] Aa. Bohr, K. Danske Vidensk. Selsk. Mat.-Fys. Medd. 26 (1952), No. 14.
- [13] E. Marshalek, L.U. Person and R.K. Sheline, Rev. Mod. Phys. 35 (1963) 108.
- [14] S.G. Nilsson, K. Danske Vidensk. Selsk. Mat.-Fys. Medd. 29 (1955), No. 16.
- [15] Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976).
- [16] P.G. Hansen, Far unstable nuclei, to be published in Annu. Rev. Nucl. Sci.

- [17] N. Trautmann, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 30.
- [18] R.D. Macfarlane and W.C. McHarris, *in* Nuclear Spectroscopy and Reactions, Pure and Applied Physics (ed. J. Cerny) (Academic Press, New York, 1974), Vol. 40A, p. 244.
- [19] P. Armbruster, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 3.
- [20] G. Rudstam, Z. Naturforsch. 21a (1966) 1027.
- [21] S. Yamaguchi, Proc. Phys. Math. Soc. Japan 23 (1941) 264.
- [22] T. Alväger and J. Uhler, Progr. Nucl. Techniques Instrum. 3 (1968) 161.
- [23] K.O. Nielsen, Proc. Internat. Conf. on Mass Spectroscopy, Kyoto, Japan, 1969 (University of Tokyo Press, Tokyo, 1970), p. 310.
- [24] F.K. Wohn, Report IS-4270 (1977), p. 16. Available from National Technical Information Service, U.S. Dept. of Commerce, Springfield.
- [25] G. Rudstam, Nucl. Instrum. Methods 139 (1976) 239.
- [26] D.D. Bogdanov, A.V. Demyanov, V.A. Karnaukhov, L.A. Petrov, A. Plohocki, V.G. Subbotin and J. Voboril, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 299.
- [27] K.H. Burkard and E. Roeckl, Nucl. Instrum. Methods 133 (1976) 187.
- [28] K. Kirchner, O. Klepper, G. Nyman, W. Reisdorf, E. Roeckl, D. Schardt, N. Kaffrell, P. Peuser and K. Schneeweiss, Phys. Lett. 70B (1977) 150.
- [29] B.W. Allardyce, A. Fiebig, G. Le Dallic, J.H.B. Madsen and P.H. Standley, Status report on CERN SC, to be published in Proc. 8th Internat. Conf. on Cyclotron and their Applications, Bloomington, USA, 1978.
- [30] S. Sundell, P.G. Hansen, B. Jonson, E. Kugler, H.L. Ravn and L. Westgaard, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Gothenburg, Gothenburg, 1973), p. 102.
- [31] H.L. Ravn, S. Sundell and L. Westgaard, Nucl. Instrum. Methods 123 (1975) 131.

- [32] E. Kugler, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Gothenburg, Gothenburg, 1973), p. 382.
- [33] G. Andersson, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Gothenburg, Gothenburg, 1973), p. 356.
- [34] E. Kugler, private communication.
- [35] G. Herrmann and H.O. Denschlag, Annu. Rev. Nucl. Sci. 19 (1969) 1.
- [36] O. Hahn and F. Strassmann, Naturwissenschaften 28 (1940) 54 and 61.
- [37] T. Bjerge, Nature 138 (1936) 400 and 139 (1937) 757.
- [38] P. Patzelt, Proc. Internat. Conf. on Electromagnetic Isotope Separators and the Techniques of their Applications, Marburg, 1970 (eds. H. Wagner and W. Walcher), Bundesministerium für Bildung und Wissenschaft Forschungsbericht BMBW-FB K 70-28 (1970), p. 158.
- [39] L.C. Carraz, I.R. Haldorsen, H.L. Ravn, M. Skarestad and L. Westgaard, Nucl. Instrum. Methods 148 (1978) 217.
- [40] H.L. Ravn, S. Sundell, L. Westgaard and E. Roeckl, J. Inorg. Nucl. Chem. 37 (1975) 383.
- [41] H.L. Ravn, L.C. Carraz, J. Denimal, E. Kugler, M. Skarestad, S. Sundell and L. Westgaard, Nucl. Instrum. Methods 139 (1976) 267.
- [42] M.L. Andersen and B. Scharff, Proc. Internat. Symposium on Why and How should we investigate Nuclides Far Off the Stability Line, Lysekil, 1966 (eds. W. Forsling, C.J. Herrlander and H. Ryde), Ark. Fys. 36 (1967) 151.
- [43] F. Hansen, A. Lindahl, O.B. Nielsen and G. Sidenius, Proc. Internat. EMIS Conf. on Low Energy Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers University of Technology and University of Gothenburg, Gothenburg, 1973), p. 426.
- [44] I. Amarel, R. Bernas, J. Chaumont, R. Foucher, J. Jastrzebski, A. Johnson, R. Klapisch and J. Teillac, Ark. Fys. 36 (1967) 77.

- [45] S. Borg, I. Bergström, G.B. Holm, B. Rydberg, L.-E. De Geer, G. Rudstam, B. Grapengiesser, E. Lund and L. Westgaard, Nucl. Instrum. Methods 91 (1971) 109.
- [46] J. Crank, The mathematics of diffusion (Clarendon Press, Oxford, 1957).
- [47] L.C. Carraz, H.L. Ravn, A. Skarestad, S. Sundell and L. Westgaard, Nucl. Instrum. Methods 158 (1979) 69.
- [48] J.H. De Boer, The dynamical character of adsorption (Clarendon Press, Oxford, 1969), p. 29.
- [49] G.J. Beyer, W.D. Fromm and A.F. Novgorodov, Zentralinst. für Kernforschung, Rossendorf, Dresden, Report ZfK-309 (1976).
- [50] E. Hagebø, Proc. Internat. Conf. on Electromagnetic Isotope Separators and the Techniques of their Applications, Marburg, 1970 (eds. H. Wagner and W. Walcher), Bundesministerium für Bildung und Wissenschaft Forschungsbericht BMBW-FB K 70-28 (1970), p. 146.
- [51] L. Holland, W. Steckelmacher and J. Yarwood, Vacuum manual (E. and F.N. Spon, London, 1974).
- [52] C.F. Powell, J.H. Oxley and J.M. Blocher, Vapour deposition (Wiley, New York, 1966).
- [53] M.D. Scheer and J. Fine, J. Chem. Phys. 38 (1963) 307.
- [54] S.M. Walas, Reaction kinetics for chemical engineers (McGraw-Hill, New York, 1959).
- [55] D.R. Olander and A.D. Pasternak, Nuclear Met., Met. Soc. AIME 15 (1969) 467.
- [56] A.D. Pasternak and D.R. Olander, AIChE J. 14 (1968) 235.
- [57] N. Schwarz, Berichte der Österreichischen Studiengesellschaft für Atomenergie Forschungszentrum Seibersdorf, SGAE Ber. No. 2174 (1973).
- [58] V.Xe. Ivanov, I.I. Papiro, G.F. Tikhinskii and V.M. Amonenko, UCRL-Trans-10036 (1965).
- [59] G. Sidenius and O. Skilbreid, Proc. Internat. Symposium on Electromagnetic Separation of Radioactive Isotopes, Vienna, 1960 (Springer Verlag, Vienna, 1961), p. 234.

- [60] J. Heinemeyer, private communication.
- [61] I. Langmuir and K.H. Kingdom, *Science* 57 (1923) 58.
- [62] I. Amarel, R. Bernas, J. Chaumont, R. Foucher, J. Jastrezbski, A. Johnson, R. Klapisch and J. Teillac, *Ark. Fys.* 36 (1967) 77.
- [63] M.L. Oliphant, E.S. Shire and B.M. Crowther, *Proc. Roy. Soc.* A146 (1934) 922.
- [64] H.L. Daley, J. Perel and R.H. Vernon, *Rev. Sci. Instrum.* 37 (1966) 473.
- [65] M.F. Harrison, AERE Report GP/R 2505 (1958).
- [66] A. Latuszyński and V.I. Raiko, *Nucl. Instrum. Methods* 120 (1974) 69.
- [67] K.F. Zmbov and J.L. Margrave, *Advanced Chem. Ser.* 72 (1968) 267.
- [68] G.J. Beyer and A.F. Novgorodov, Zentralinst. für Kernforschung, Rossendorf, Dresden, Report ZfK-303 (1976).
- [69] A. Venezia and S. Amiel, *Nucl. Instrum. Methods* 87 (1970) 307.
- [70] V.S. Fomenko, *Handbook of thermionic properties* (Plenum Press, New York, 1966).
- [71] B. Vociki, private communication.
- [72] J.L. Beauchamp, *J. Chem. Phys.* 64 (1976) 929.
- [73] I.N. Bakulina, E.Ya. Zandberg and N.I. Ionov, *Soviet Phys. Tech. Phys.* 10 (1965) 437.
- [74] R. Kirchner and E. Roeckl, *Nucl. Instrum. Methods* 127 (1975) 307 and 131 (1975) 371.
- [75] E. Hagebø, A. Kjelberg, P. Patzelt, G. Rudstam and S. Sundell, CERN 70-3 (1970), p. 93.
- [76] Ch. Andersson, B. Grapengiesser and G. Rudstam, The Swedish Research Council's Laboratory, Nyköping, Sweden, Research Report LF-45 (1973).
- [77] K.C. Chan, B.P. Pathak, L. Nikkinen, L. Lessard and I.S. Grant, *J. Inorg. Nucl. Chem.* 39 (1977) 1915.
- [78] L. Westgaard, G. Rudstam and O.C. Jonsson, *J. Inorg. Nucl. Chem.* 31 (1969) 3747.
- [79] B. Grapengiesser and G. Rudstam, *Radiochim. Acta* 20 (1973) 85.

- [80] P. Hornshøj, B. Jonson, H.L. Ravn and L. Westgaard, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Gothenburg, Gothenburg, 1973), p. 379.
- [81] R. Klapisch, Nuclear spectroscopy and reactions, Pure and Applied Physics (ed. J. Cerny) (Academic Press, New York, 1974), Vol. 40A, p. 213.
- [82] J.M. D'Auria, L.C. Carraz, P.G. Hansen, B. Jonson, S. Mattsson, H.L. Ravn, M. Skarestad and L. Westgaard, Phys. Lett. 66B (1977) 233.
- [83] E. Roeckl, R. Kirchner, G. Nyman and W. Reisdorf, Recent developments and results with the GSI on-line separator at the UNILAC, Contribution to the Isotope Separator On-Line Workshop, Brookhaven, 1977, to be published as a BNL Report.
- [84] W. Reisdorf, Gesellschaft für Schwerionenforschung, GSI-Bericht-M-2-78 (1978).
- [85] L. von Reisky, J. Bonn, S.L. Kaufman, L. Kugler, E.-W. Otten, J.-M. Rodriguez-Giles, K.P.C. Spath and D. Weskolt, An on-line mass separator for fission-produced alkali isotopes, to be submitted to Nucl. Instrum. Methods.
- [86] T. Bjørnstad, L.C. Carraz, J. Heinemeyer, H.L. Ravn, M. Skarestad and L. Westgaard, New high temperature carbide targets for ISOLDE, to be published.
- [87] S. Katcoff, J. Gilat, P.E. Haustein, E.-M. Franz, N.A. Morcos, T.E. Ward, H.A. Smith, Jr. and R.F. Petry, Proc. Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 528.
- [88] O.B. Nielsen, private communication.
- [89] J.M. D'Auria, J.W. Grüter, E. Hagberg, P.G. Hansen, J.C. Hardy, P. Hornshøj, B. Jonson, S. Mattsson, H.L. Ravn and P. Tidemand-Petersson, Nucl. Phys. A301 (1978) 397.
- [90] E. Hagberg, P.G. Hansen, J.C. Hardy, A. Huck, B. Jonson, S. Mattsson, H.L. Ravn, P. Tidemand-Petersson and G. Walter, Phys. Rev. Lett. 39 (1977) 792.

- [91] P. Hornshøj, P.G. Hansen, E. Hagberg, B. Jonson, S. Mattsson, O.B. Nielsen, H.L. Ravn, P. Tidemand-Petersson and L. Westgaard, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 171.
- [92] R. Silberberg and C.H. Tsao, Naval Research Laboratory, Washington DC, NRL Report 7593 (1973).
- [93] E. Hagberg, P.G. Hansen, J.C. Hardy, P. Hornshøj, B. Jonson, S. Mattsson and P. Tidemand-Petersson, Nucl. Phys. A293 (1977) 1.
- [94] L.C. Carraz, G.T. Ewan, E. Hagberg, B. Jonson, S. Mattsson, H.L. Ravn, M. Skarestad and P. Tidemand-Petersson, Fast target technique for studies of alpha decay from very short-lived francium isotopes, to be published.
- [95] W.W. Meinke, A. Ghiorso and G.T. Seaborg, Phys. Rev. 81 (1951) 782.
- [96] E. Roeckl, D.F. Dittner, C. Détraz, R. Klapisch, C. Thibault and C. Rigaud, Phys. Rev. C 10 (1974) 1181.
- [97] R.E. Azuma, L.C. Carraz, P.G. Hansen, B. Jonson, K.L. Kratz, S. Mattsson, G. Nyman, H. Ohm, H.L. Ravn, A. Schröder and W. Ziegert, Beta-delayed two-neutron emission from ^{11}Li and other recent results in delayed particle spectroscopy, to be published in Proc. 8th Internat. Workshop on Gross Properties of Nuclei and Nuclear Excitations, Hirschegg, Austria, Jan. 1979 (Institut für Kernphysik, Technische Hochschule, Darmstadt, 1979).
- [98] J. Genevey-Rivier, A. Charvet, G. Marguier, C. Richard-Serre, J.M. D'Auria, A. Huck, G. Klotz, A. Knipper and G. Walter, Nucl. Phys. A283 (1977) 45.
- [99] Phys. Today 31 (1978) 17.
- [100] C. Ekström, S. Ingelman and G. Wannberg, Nucl. Instrum. Methods 148 (1978) 17.
- [101] C. Ekström, G. Wannberg and J. Heinemeier, Nuclear spin of a new isomer in ^{119}Cs and magnetic moment of ^{120}Cs -- evidence for strongly deformed nuclear shapes of the light cesium isotopes, submitted to Phys. Lett. B.
- [102] H.-J. Kluge, Proc. 3rd Internat. Conf. on Nuclei Far from Stability, Cargèse, 1976, CERN 76-13 (1976), p. 177.
- [103] J. Bonn, G. Huber, H.-J. Kluge, L. Kugler and E.-W. Otten, Phys. Lett. 38B (1972) 308.
- [104] J.H. Hamilton et al., Phys. Rev. Lett. 35 (1975) 562.

- [105] C. Bourgeois et al., J. Phys. (NY) 37 (1976) 49.
- [106] D. Proetel, R.M. Diamond, P. Kienle, J.R. Leigh, K.H. Maier and F.S. Stephens, Phys. Rev. Lett. 31 (1973) 896.
- [107] R. Béraud, C. Bourgeois, M.G. Desthuilliers, P. Kilcher and J. Letessier, J. de Phys. 36 (1975) Colloq. C5-101.
- [108] N. Rud, D. Ward, H.R. Andrews, R.L. Graham and J.S. Geiger, Phys. Rev. Lett. 31 (1973) 1421.
- [109] T. Kühn, P. Dabkiewicz, C. Duke, H. Fischer, H.-J. Kluge, H. Kremmling and E.-W. Otten, Phys. Rev. Lett. 39 (1977) 180.
- [110] K.R. Anton, S.L. Kaufman, W. Klempt, G. Moruzzi, R. Neugart, E.-W. Otten and B. Schinzler, Phys. Rev. Lett. 40 (1978) 642.
- [111] G. Huber, F. Touchard, S. Büttgenbach, C. Thibault and R. Klapisch, Phys. Rev. Lett. 41 (1978) 459.
- [112] S. Liberman, J. Pinard, H.T. Duong, P. Juncar, J.L. Vialle, P. Jacquinet, G. Huber, F. Touchard, S. Büttgenbach, A. Pesnelle, C. Thibault and R. Klapisch, CR Acad. Sci. B286 (1978) 253.
- [113] J. Blomquist, Ark. Fys. 36 (1967) 681.
- [114] G. Andersson, M. Asghar, A. Emsallem, E. Hagberg, B. Jonson and P. Tidemand-Petersson, Study of the reaction $^{76}\text{Br}(n_{\text{th}}, p)^{76}\text{Se}$, Phys. Scripta 18 (1978) 165.
- [115] I. Berkès, private communication.
- [116] P. Herzog, H.-R. Folle, K. Freitag, A. Kluge, M. Reuschenbach and E. Bodenstedt, Nucl. Instrum. Methods 155 (1978) 421.
- [117] H. Haas, private communication.
- [118] G. Weyer, private communication.
- [119] N.A. Dyson, X-rays in atomic and nuclear physics (Longman, London, 1973).
- [120] O.I. Sumbaev, Proc. Internat. Conf. on Inner Shell Ionization Phenomena, Atlanta, 1972 (USAEC Technical Information Center, Oak Ridge, Tennessee, 1972), Conf-720404, p. 346.
- [121] F. Boehm, *in* Atomic inner shell processes (ed. B. Crasemann) (Academic Press, NY, 1975), Vol. 1, p. 411.

- [122] A.A. Hahn, K.-C. Wang, F. Boehm, H.E. Hendrikson and P. Vogel, Phys. Rev. Lett. 37 (1976) 1741.
- [123] H. Primakoff and F.T. Porter, Phys. Rev. 89 (1953) 930.
- [124] G.L. Borchert, P.G. Hansen, B. Jonson, H.L. Ravn, O.W.B. Schult and P. Tidemand-Petersson, Phys. Lett. A63 (1977) 15.
- [125] P.G. Hansen, Nucl. Instrum. Methods 154 (1978) 321.
- [126] G.L. Borchert, P.G. Hansen, B. Jonson, I. Lindgren, H.L. Ravn, Phys. Lett. A65 (1978) 297.
- [127] G.L. Borchert, P.G. Hansen, B. Jonson, I. Lindgren, H.L. Ravn, O.W.B. Schult and P. Tidemand-Petersson, Phys. Lett. A66 (1978) 374.
- [128] M.L. Mallory and G.S. McNeilly, Nucl. Instrum. Methods 138 (1976) 421.
- [129] P.G. Hansen, The acceleration of short-lived, radioactive nuclei, unpublished.

Table 3.1

Essential parameters for comparing on-line separators

	Nuclear reactor	Heavy-ion accelerator	High-energy proton accelerator
Particle	n_{th}	HI	p
Energy	Thermal	~ 10 MeV/A	~ 600 MeV
Maximum available intensity (particles/s)	10^{15}	6×10^{12}	10^{15}
Nuclear reaction	Thermal fission of ^{235}U	(HI,ypxn)	(p,ypXn)
Cross-section (cm^2)	$< 2 \times 10^{-23}$	$< 2 \times 10^{-25}$	$< 2 \times 10^{-26}$
Effective target thickness (atoms/ cm^2)	1×10^{18}	10^{19}	6×10^{23}
Highest beam intensity used in an on-line separator experiment (particles/s)	10^{11}	10^{12}	10^{13}
Maximum production rates (atoms/s)	10^8	10^6	10^{11}

Table 3.2

Operational statistics for ISOLDE

	1.7.1976 to 30.6.1977	1.1.1977 to 31.12.1977	1.1.1978 to 31.12.1978
Number of scheduled shifts	187.5	211.5	228.5
Number of shifts used	163	186	227
Number of user shifts	428	429	350
Average number of users	~ 2.6	~ 2.3	~ 1.5
Running periods	20	19	17
Number of scheduled experiments	109	97	~ 100
Experimental groups/run	~ 5.5	~ 5	~ 3

Table 3.3

Total number of shifts scheduled
for different ISOLDE targets from 1975 to 1977

Product (target)	Number of shifts	% of total	Target units used
Cs(La)	118.5	29.0	12
Hg(Pb)	114.0	27.9	9
Fr/Cs/Rb(UC)	54.5	13.3	7
Rb(Nb)	34.0	8.3	2
Ar(VC)	28.5	7.0	5
Cd(Sn)	21.5	5.3	2
Yb(Ta)	19.0	4.7	6
Xe(La)	10.0	2.4	3
Kr(Nb)	5.5	1.3	2
Br(Nb)	3.0	0.7	2

Figure captions

- Fig. 2.1 : The chart of nuclides. The stable isotopes are represented by the black squares. The region of known nuclides unstable against beta decay is enclosed by the broken contour. The diagonal smooth lines indicate the approximate limit of proton or neutron stability ($B_p \approx 0$ being the upper line; $B_n \approx 0$ being the lower line; B denoting nucleon binding energy). The circular lines are isodeformation curves showing the domain where nuclear deformation occurs.
- Fig. 2.2 : Isotopic yield distribution for the spallation of La by 600 MeV protons, calculated according to the semi-empirical formula of Rudstam [20].
- Fig. 3.1 : Schematic view of an on-line isotope separator experiment.
- 1) Target and ion-source unit traversed by the particle beam.
 - 2) Acceleration chamber.
 - 3) Analysing magnet.
 - 4) and 5) Deflector chamber with electrostatic switchyard.
 - 6) External beam line.
- Fig. 3.2 : SC layout in Summer 1978.
- Fig. 3.3 : Layout of the ISOLDE-2 separator and experimental area. Separator and beam lines: BL = external beam lines 1-4; C = collimators in the focal plane 1-4; D = switchyard electrostatic deflectors 1-4; FP = fore pumps; G = ionization vacuum gauges; IG = electrostatic beam gates; LA-LD = electrostatic quadrupole triplets; LE = magnetic quadrupoles in the proton beam; ME = deflector magnets in the proton beam; N = nitrogen inlet; ScA-ScC = oscillation vane scanners; TC = thermocouple vacuum gauge; TP = turbomolecular pumps; TV = television cameras; V = valves.

Fig. 3.4 : Remote-handling device for target exchange.

- 1) Acceleration chamber.
- 2) Lever mechanism for positioning of the target.
- 3) High-voltage screen to take up charges induced in the air by the passage of the proton beam.
- 4) Concrete door.
- 5) Shielded vault.
- 6) Fibre-glass rail and spindle.
- 7) Carriage for transport of the target.
- 8) Target and ion-source unit.
- 9) Turntable accommodating up to four targets.

Fig. 3.5 : The arrangement of the four electrostatic deflector plates of the switchyard, viewed from above and in the direction of the external beam lines.

Fig. 3.6 : Mass spectrum of the most intense radioactive Hg beams and ^{204}Pb as measured with an oscillating needle scanner.

Fig. 3.7 : Perspective view of the experiments at ISOLDE-2.

- 1) Target and ion-source development.
- 2) Nuclear spectroscopy (α , β , γ).
- 3) High-resolution mass spectrometer.
- 4) Optical pumping and laser spectroscopy.
- 5) Atomic-beam magnetic resonance.
- 6) Collection of radioactive sources for off-line work (hyperfine interactions in solids, determination of shifts in X-ray energies, targets for nuclear-reaction studies).
- 7) Beta-delayed particles.
- 8) Range measurements of ions in gases.

Fig. 4.1 : The figure shows the release of Yb from irradiated Ta powder (20 μm particle size). The points represent the fraction of the isotope

^{169}Yb remaining in the Ta sample as a function of the heating time. The curves have been fitted to the experimental points according to eq. (4.2).

- Fig. 4.2 : Periodic system of the elements, indicating high-temperature powder or fibre targets together with the possible product elements and the rate-determining mechanism.
- Fig. 4.3 : Survey of the elements available from molten-metal targets.
- Fig. 4.4 : Integral delay curve obtained by sampling the yield of ^{118}Xe in regular intervals after the proton beam is switched off. The points marked with crosses are the background-corrected count rates. The measured average delay time $\tau_{\text{Xe}} = 24$ s. The target data were the following: lanthanum target 120 g/cm^2 ; temperature = 1473 K ; $V_0 = 15 \text{ cm}^3$; $S_0 = 60 \text{ cm}^2$; $S_1 = 0.07 \text{ cm}^2$; $\ell = 1 \text{ cm}$; $L = 5 \text{ cm}$; $d = 0.3 \text{ cm}$; $D = 1 \text{ cm}$.
- Fig. 4.5 : The average delay time τ_{Rb} as a function of the volume-to-surface ratio V_0/S_0 for the Yb-La target melt. The open circles denote off-line measurements. The filled circles represent the value for the typical on-line target and ion-source assembly.
- Fig. 4.6 : Coupling of a target and ion-source unit to the separator acceleration chamber. The compact assembly consists of a water-cooled base plate (similar to that of a standard vacuum evaporation plant) with feedthroughs, pumping port, and quick connectors. It carries the target and ion source and is enclosed in an aluminium bell jar (for economical reasons a commercial cooking casserole is used). By means of an insulating rail and spindle the unit is moved to the acceleration tank, where the coupling force is taken over by pneumatically operated levers.

Fig. 4.7 : The mechanical construction of the ISOLDE-2 target and ion-source unit.

- 1) Target container surrounded by heat screens.
- 2) Transfer line.
- 3) Ion source.
- 4) Copper cone.
- 5) Terminals for target support and heating.
- 6) Terminal for the line heating.
- 7) Thermocouple.
- 8) Extraction electrode.
- 9) Magnet coil.
- 10) Base plate with feedthroughs.
- 11) Vacuum chamber.

Inset A: Low-temperature Ta surface ionizer.

Inset B: High-temperature W surface ionizer.

Inset C: Forced electron-beam-induced arc-discharge source.

Fig. 4.8 : Release yields of Cs isotopes from different uranium target matrices. The release yields, Y_{obs}/Y , are given as a function of the half-life of the nuclide.

Fig. 4.9 : Variations with atomic number of the heats of sublimation of the rare-earth metals (a), and the ionization potential of the metals (b) and monofluorides (c).

Fig. 5.1 : Experimental yields of Hg isotopes produced by irradiating a 170 g/cm^2 molten-Pb target with a $1.5 \text{ }\mu\text{A}$ proton beam. The yield points (filled circles) which extend over 11 decades are given as atoms per second in the beams at the collector side of the separator. A horizontal line at 10^6 represents the level of contamination spread uniformly over the mass range, whereas curve a illustrates the amount of contamination from the next heavier neighbour. Curve b shows the upper limit of contamination from the element Tl.

Fig. 5.2 : Present status of the Cs isotopic yields as obtained with different bombarding particles and targets. Denoted by means of open and filled triangles and open squares are the yields of the heavy-ion reactions $^{58}\text{Ni}(^{58}\text{Ni},\text{pxn})\text{Cs}$, $^{89}\text{Y}(^{40}\text{Ar},2\text{pxn})\text{Cs}$, and $^{63}\text{Cu}(^{58}\text{Ni},2\text{pxn})\text{Cs}$, respectively. These measurements are made with the separator in Darmstadt on line to the 5 MeV/amu, $\sim 10^{12} \text{ s}^{-1}$ heavy-ion beams from the GSI, UNILAC [83,84]. The crosses represent the strongly cumulative thermal fission yields as produced in the on-line experiment at the Triga reactor in Mainz [85]. The 200 mg ^{235}U target is located close to the core in a neutron flux density of $1.7 \times 10^{11} \text{ s}^{-1} \text{ cm}^{-2}$. Illustrated by means of filled and open circles are the production yields at ISOLDE in 1 μA proton irradiations of a 120 g cm^{-2} molten-La target and a 13 g cm^{-2} ^{238}U target. It should be noted that the ISOLDE yields from ^{146}Cs and outwards are the neutron saturation count rates corrected for the estimated neutron branching ratio and normalized to the yield of ^{146}Cs , measured by β^- counting. The curves are only drawn to guide the eye.

Fig. 5.3 : Observed yields of sodium and potassium isotopes, corresponding to a 1 μA 600 MeV proton beam impinging on a 14 g/ cm^2 uranium-carbide target and a 3.2 g/ cm^2 scandium carbide target. The yields below ^{27}Na and ^{50}K are measured by means of a 4 πsr beta counter, those above are estimated from measurements of beta-delayed neutrons and normalized to the beta yields of ^{29}Na and ^{50}K , respectively.

Fig. 5.4 : Production yields of light Fr isotopes obtained by irradiation of a 13 g/ cm^2 target of ^{238}U -loaded graphite fibres with 600 MeV protons (triangles) and 910 MeV ^3He (filled circles). The yields have been normalized to correspond to 6×10^{12} particles/s. Note that the yield of the 48 ms ^{201}Fr has increased by almost a factor of 20 with ^3He as projectile.

Fig. 5.5 : Decay of beta rays of energy greater than 5 MeV from ^{74}Rb detected in a multiscaling mode. The experimental arrangement is shown schematically in the inset. During the counting periods, the ^{74}Rb ion beam was intercepted by an electrostatic beam gate placed far upstream in the beam line. During the collections, the beam was directed electrostatically to an aluminized mylar foil placed in front of a counter telescope consisting of a 1 mm thick ΔE counter of 1 cm² followed by an 8 cm diameter $\times$ 5 cm thick NE110 plastic scintillator. The over-all efficiency of the telescope including solid angle was 20%, determined for 3.5 MeV betas.

Fig. 5.6 : Yields and cross-sections of Yb isotopes (filled circles) produced in a 114 g/cm² Ta powder target, irradiated in a 1 μA proton beam. The open circles denote the yields corrected for delay losses according to eq. (3.11). The triangles and the crosses represent the contamination from the elements Lu and Tm, respectively. The cross-sections are obtained by normalizing the yields to the thin target experimental value of 11 mb for ^{169}Yb taken from ref. 92.

Fig. 5.7 : a) Pulse-height spectrum of beta-delayed neutrons from 8.5 ms ^{11}Li as measured with a ^3He spectrometer of 20 keV resolution. The inset shows the low-energy part of the spectrum. The double-humped distribution of neutrons ranging from the low-energy end of the spectrum up to about 1600 keV is interpreted as beta-delayed two-neutron emission from the 8.84 keV state in ^{11}Be .
b) Decay scheme of ^{11}Li where the neutron transitions that give the main contribution to the energy spectrum are indicated as E_1 to E_6 .

Fig. 6.1 : Top view of the ABMR apparatus. The ion beam (1) from ISOLDE passes through an off-line sample collection box (2), an electrostatic deflector (3), the beam tube (4), a beam observation box (5), and a valve (6) before it is focused onto the oven section (8) by means of an electrostatic quadrupole triplet (7). The stopped ions are

re-evaporated as an atomic beam which passes through a polarizing sextupole magnet (9), a dipole magnet (10) where the spin-flipping radio-frequency field is applied, and finally through an analysing quadrupole magnet (11) to the detection system (12-14). In the lower part of the figure the beam path of atoms in resonance is shown, i.e. those that have undergone a transition from a focusing to a defocusing state. In off-resonance, the atoms hit the central obstacle in the analysing magnet.

Fig. 6.2 : Changes of charge radii of Hg isotopes relative to ^{204}Hg and plotted against atomic number. The abrupt change at $A = 185$ was discovered by the optical pumping technique. Full circles represent charge radii of ground states, and open circles indicate those of isomers. The arrows point to the results from laser spectroscopy, which reveal an odd-even shape-staggering unknown in other mass regions.

Fig. 6.3 : Schematic diagram of collinear laser spectroscopy on an isotope separated beam. Since the resonance lines of ions mostly are in the ultraviolet region, which is inaccessible with present single-mode dye lasers, the ion beam is neutralized in the charge-exchange cell. The hyperfine structure of the spectral lines is measured at constant laser frequency by tuning the particle velocity in the charge-exchange cell which is connected to a scanning voltage. The reference beam may come from a separate ion gun, but it seems to be most advantageous to take it directly from the isotope separator.

Fig. 6.4 : General scheme of the atomic-beam laser spectroscopy arrangement. The mass-separated alkali ion beam is stopped, neutralized, and formed into an atomic beam by means of an oven with yttrium-covered surfaces. The laser-beam-induced hyperfine sublevels of the ground state of the atoms are analysed by a six-pole magnet, after which the atoms are re-ionized and detected by a high-sensitivity mass spectrometer.

Fig. 6.5 : Principle of the crystal spectrometer and its double-source arrangement. The lower is the photo-ionization source.

Fig. 6.6 : The F selection rule for the population of the hyperfine levels. It is assumed that the initial state has a nuclear spin of $(I - 1)$ and a full K shell, so that the total angular momentum is $F_i = I - 1$. The nucleus decays by an allowed (or "quasi-allowed" forbidden) K-capture transition to a state with spin I . The neutrino carries away $\frac{1}{2}$ unit of angular momentum, and it is immediately seen that the remaining $1s$ electron can only be in a state antiparallel to the nuclear spin. This is the consequence of the selection rule $\Delta F = \pm \frac{1}{2}$. Consequently, the observed energy shift is $-A(I + 1)/2$, where A is the usual hyperfine parameter proportional to the nuclear g factor.

Fig. 6.7 : Unresolved satellite lines for the 68.8 keV gold $K_{\alpha 1}$ X-ray produced in photo-ionization. The main lines (at zero) originate in the $2p_{3/2} \rightarrow 1s_{1/2}$ transition. The presence of a "spectator" hole with the quantum numbers indicated will give rise to a shifted component with calculated energy and intensity as shown. The natural width of the K X-rays (50 eV) is very much larger than the shifts involved, so that the net effect only is a slight displacement of the composite peak. This displacement may be observed by comparing the energy with that of X-rays from electron capture, for which process the intensities of the satellite lines are approximately two orders of magnitude lower.

Fig. 6.8 : Measured energy shifts (points, full-drawn line) for holmium K X-rays for the transition from np states to the $1s$ state as a function of n . Three calculations are shown: the hyperfine shift (hfs), the sum of hyperfine shift and shake-off effect (so) and, finally, a calculation which includes also the effect of the additional $4f$ electron.

Fig. 2.1

Fig. 2.2

Fig. 3.1

62618

Fig. 3.2

Fig. 3.3

Fig. 3.4

Fig. 3.5

Fig. 3.6

56899

Fig. 3.7

Fig. 4.1

GROUP IA																				VIII A										
H	IIA																				He									
Li	Be											B	C	N	O	F	Ne													
Na	Mg	IIIB	IVB	VB	VIB	VII B	VIII			IB	IIB	Al	Si	P	S	Cl	Ar													
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr													
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe													
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn													
Fr	Ra	Ac																												
LANTHANIDES—		Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu															
ACTINIDES—		Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr															

- Metal powder target
- Carbide target
- Product with diffusion-controlled release rate
- Product with desorption-controlled release rate

Fig. 4.2

PERIODIC TABLE OF THE ELEMENTS

GROUP IA																	VIIIA	
	H																He	
	Li	Be															Ne	
	Na	Mg															Ar	
	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
	Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac																
LANTHANIDES			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu		
ACTINIDES			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr		

Molten metal target

Product element

☐ Molten metal target
☒ Product element

Fig. 4.3

Fig. 4.4

Fig. 4.5

Fig. 4.6

Fig. 4.7

Fig. 4.8

Fig. 4.9

Fig. 5.1

Fig. 5.2

Fig. 5.3

Fig. 5.4

Fig. 5.5

Fig. 5.6

Fig. 5.7

Fig. 6.1

Fig. 6.2

Fig. 6.3

Fig. 6.4

Fig. 6.5

$\Delta l = +1$

68415

$$\Delta E = -\frac{A(l+1)}{2}$$

Fig. 6.6

68412

Fig. 6.7

68293

Fig. 6.8