



Secondary Beams of Radioactive Ions: The Isolde Experiments at CERN

P.G. Hansen, Aarhus and B. Jonson, CERN and the ISOLDE Collaboration

The use of a high-energy proton accelerator coupled to an "Isotope Separator On-Line" (ISOL) as a tool for studying problems in nuclear, atomic and solid-state physics.

The ISOL technique was first developed to enable nuclear physicists to make measurements on extremely short-lived and rare radioactive isotopes, the problems of which can be illustrated by considering the isotope ^{38}Ar , one of the few nuclei known with a proton excess of four units. The isotope which can be produced by the bombardment of vanadium with high energy protons, has a half-life of only 0.075s. With a primary beam intensity of $1\mu\text{A}$ ($\sim 6 \times 10^{12}$ protons/s), the typical yield is 0.3 atoms/s as against 10^{13} unwanted radioactive atoms/s. The experimental situation is thus very much like the search for a needle in a (very big) haystack.

In an ISOL experiment, the nucleides are produced in a target which is closely connected to the ion source of an electromagnetic separator (see Fig. 1). Short-lived products are liberated before they decay, and the characteristics of the ion source can be chosen so that only one element appears in the ion beam. After acceleration, the ion beam is mass-analyzed in a magnet, and only the mass of interest is delivered to the experimental measuring station: the needle has been separated from the haystack.

ISOL techniques are today being used in a large number of laboratories around the world to study specific problems in nuclear structure, especially in connection with nuclei that are extremely rich or extremely deficient in neutrons ("nuclei far away from the line of beta stability") and we shall give examples of such experiments in the following. However, we wish to emphasize a more general aspect.

A separator coupled to a high energy proton beam can supply a very wide range of masses and elements, providing beams of an intensity that is usually vastly superior to that which can be obtained by other methods. Such an installation is therefore capable of serving a large number of different experiments and these are not confined to nuclear physics. As we shall see, there is a wide application also in atomic and solid-state physics. Techniques have been developed to the point where ion beams of more than 40 elements can, in principle, be made available for experiments. For the moment, however, the ISOLDE installation at CERN's 600 MeV synchro-cyclotron is the only existing multi-purpose facility of the kind. The acceleration stage, the mass-analyzing system and the system for delivering beams to different experiments are shown in Fig. 2.

Sources

The most useful projectiles available at the CERN SC are 600 MeV protons and 910 MeV ^3He . The high penetration power of these beams makes it possible to use very thick targets, up to 150 g/cm^2 , which with a $1\mu\text{A}$ beam of protons give isotope-separated beams of more than 10^{11} ions for the most abundant species. The neutron deficient isotopes are best produced in spallation reactions, an example of which is the production of the lightest known isotope of caesium with mass 114, from a lanthanum target in the reaction:

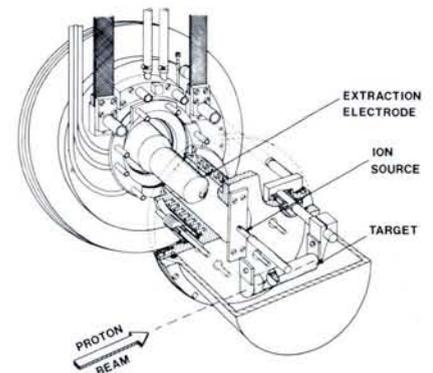
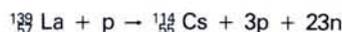


Fig. 1 — A disposable target unit designed for the production of isotopes of alkalis from molten-metals. Owing to the high radioactivity induced in these units, they are discarded in the case of a break-down. A vacuum entrance for the 600 MeV proton beam is not necessary; the beam penetrates the vacuum container and the 20 cm long target cylinder seen in the Figure. Also to be seen is the extraction electrode, through which the heavy ion beam enters the 60 kV acceleration stage of the isotope separator.

The neutron-rich isotopes are produced either from fission or fragmentation. In a target matrix of uranium carbide one may, for example, reach the heaviest known caesium isotope with mass 152 from fission, while fragmentation in the same target gives high yields of the heaviest lithium isotope which is stable against decay by particle emission, ^7Li .

The ISOLDE target is normally kept at a high temperature so that the products of the reaction evaporate rapidly out of the target matrix and enter the ion source. The simplest type of ion source is based on high temperature surface ionization and will provide beams of alkali metals and alkaline earth metals. Chemically pure beams of noble gases may be obtained with a plasma ion source in combination with a cold line between the target and the ion source; all elements except the noble gases, condense on the cold line, and a chemically pure beam is the result. Intense and pure beams of halogens may be obtained, as has been shown recently, with a negative surface

ionization source, in which the active element is a LaB_6 surface which has a low work function for electrons, so that halogens pick up an electron from the surface and emerge as negative ions.

Applications

In keeping with the multidisciplinary nature of the subject, we have divided the presentation into self-contained examples of specific applications. More detailed treatments, especially of the physics of far-unstable nuclei, may be found in the review article and the proceedings of recent conferences quoted in references 1-6 (page 6).

One of the most striking applications of ISOLDE is the possibility it gives of studying a given nuclear property over a very large mass range, and the observation of patterns that would not emerge for a few cases measured near stability. This exploitation of what one might call "the isospin degree of freedom" is illustrated particularly well by several long series of experiments identifying the properties of nuclear ground states (and low lying isomeric states) such as nuclear masses (Panel 1), nuclear spins and moments (Panel 2) and charge radii (Panel 3). These experiments have also given insight into the interplay between single-particle structure and deformation effects in nuclei over a large part of the nuclear chart (Panel 4). Many studies of nuclear excited levels have contributed to the same end.

Far away from beta stability the radioactive decays are extremely complex and in a number of ways, different from those near stability. Often loosely referred to as "exotic" decay modes, their origin lies in the nuclear mass surface, which away from stability creates very high energy for the β -decay (Q_{β^-} for neutron rich and Q_{EC} for neutron deficient isotopes) and to particle separation energies that may be very low. The decays usually involve particle emission and the level densities often are so high that a statistical treatment is more appropriate than a consideration of individual levels. Examples of exotic decays and their interpretation are given in Panels 5 and 6.

Finally, the ISOLDE Installation is being used to an ever increasing extent for the study of non-nuclear phenomena. Implantation of radioactive ions is of growing importance in solid-state physics (Panel 7), and there are many applications in atomic physics (Panel 8). The use of radioactive ion beams in chemistry and for applied work (technology, medicine) in the not too distant future seems probable.

It is possible to give many more examples of applications of radioactive ion beams, but we feel that the cases cited here should suffice to give an impression of the possibilities. We invite all readers to consider new applications...

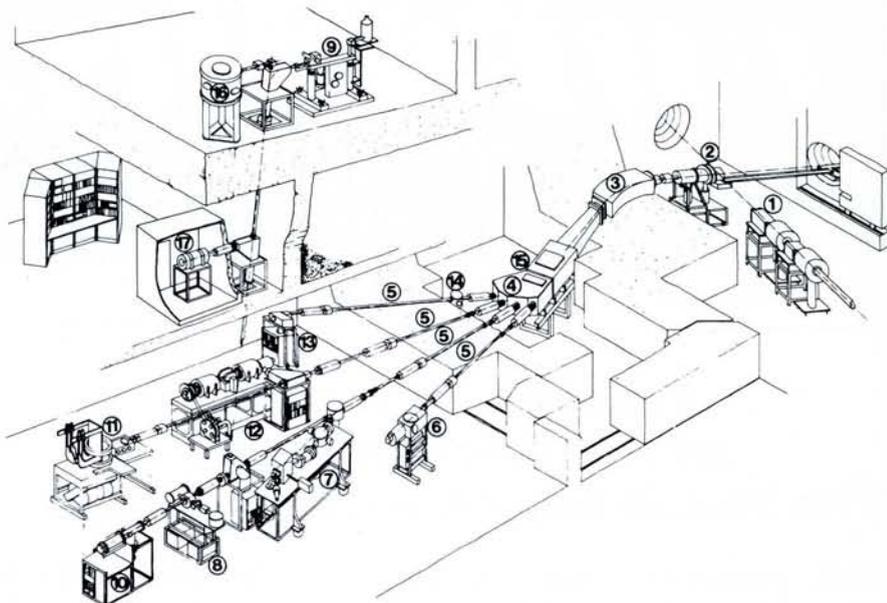


Fig. 2 — The isotope separator and a selection of typical experiments, separated from the target area by two heavy shielding walls with mid-plane iron. Not shown are electronics, power supplies, on-line computers, lasers, etc. The 600 MeV proton beam (or 910 MeV ^3He beam) (1) is focused on the target-ion-source unit (2) and the resulting (radioactive) heavy-ion beam is mass analyzed in the magnet (3). Individual masses are selected in the electrostatic switchyard (4) and distributed through the external beam lines (5) to the experiments, placed in three experimental halls.

The experiments shown here comprise proton and alpha spectroscopy (6) (Panel 5), high-resolution mass spectroscopy (7) (Panel 1), beta-gamma spectroscopy (8) (9) (16), range measurements of ions in gases and sputtering studies (10), atomic spectroscopy through laser techniques (11) (Panel 1) and atomic beam magnetic resonance (12) (Panel 2), collections of samples for experiments in nuclear, atomic and solid-state physics (13) (14) (15) (in particular implantation studies, see Panel 8) and studies of beta-delayed neutron emission (17) (Panel 4).

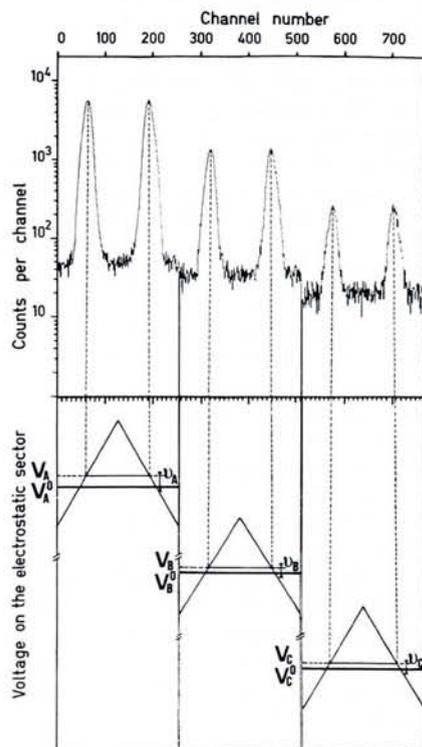
Panel 1

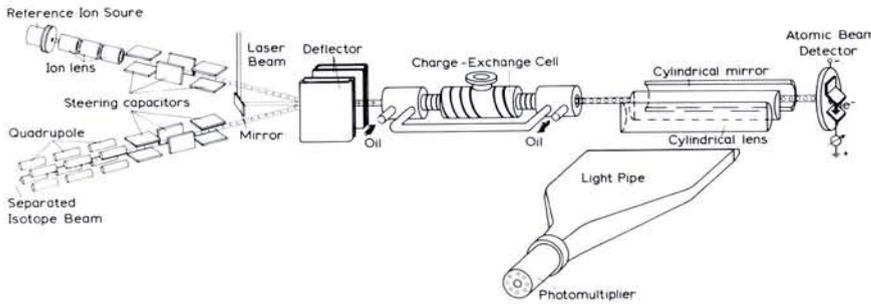
High Precision Mass Measurements by Direct Mass Spectrometry

Owing to the high intensities available at a high-energy proton accelerator, it has recently become possible to use mass spectrometry for determining the ground-state masses of radioactive atoms. The technique, which achieves a precision better than 100 keV for heavier nuclei ($A > 100$) is based on the double focusing Mattauch-Herzog spectrometer. The measurements are carried out for a constant magnetic field and are based on the property that if two ions of masses M_A and M_B follow the same trajectory in the system, all electric potentials V should obey the relation $M_A/M_B = V_B/V_A$. The mass measurement thus is converted to a precise measurement of a ratio of electrostatic potentials.

The figure shows the recording of the transmitted intensity for the three rubidium isotopes $^{95, 97, 98}\text{Rb}$ (denoted A, B and C). At each mass setting, the voltage was modulated by a "sawtooth" shape around the fixed potential V^0 , as shown in the lower part of the figure.

With this technique, extensive data have been taken for the alkali elements (Li-Fr). Among the very striking results has been the first evidence for a new region of deformed nuclei near $(Z, N) = (11, 20)$ and also strong evidence for the interplay of nuclear shapes and (spherical) shells in the rubidium and caesium isotopes.



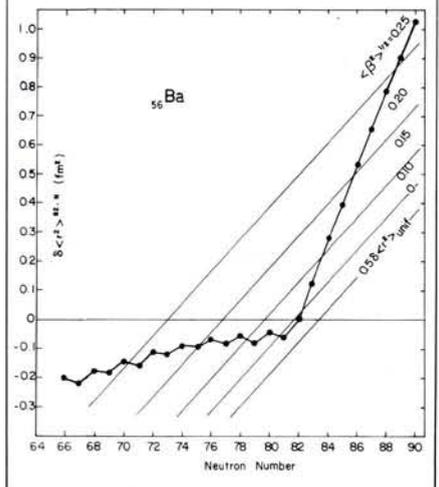


Although a number of different techniques have already been devised for determining nuclear spins, etc., the Fig. shows a recent and extremely powerful method, the so-called collinear laser technique first developed at Mainz University. An ion beam is brought into a charge exchange cell where it is neutralized by alkali vapour and converted into an atomic beam. A light beam from a single-frequency dye laser is superimposed on the atomic beam and resonance scattered photons from the observation region are detected in a photomultiplier tube. The collinear superposition of the beams gives not only the advantage of having the mass-separated beam directly as the spectroscopic sample, but it allows also an exploitation of the compression of the velocity distribution due to the acceleration. The energy spread of the ion beam:

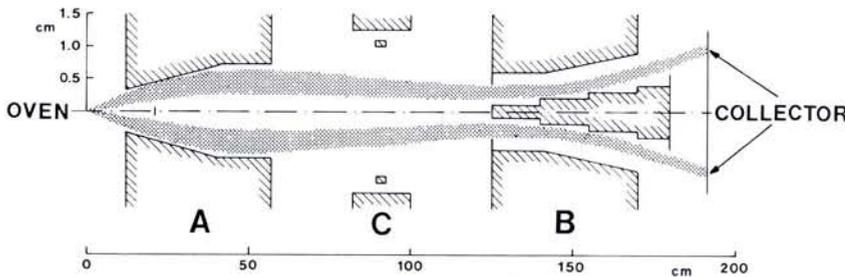
$$\delta E = \delta(mv^2/2) = mv\delta v$$

remains constant, which gives a narrowing of the velocity spread δv along the beam when the beam velocity $v = (2eU/m)^{1/2}$ is increased. (The acceleration voltage is here denoted U .) The absorption Doppler width is consequently decreased by a factor of the order $(kT/eU)^{1/2}$, corresponding typically to more than two orders of magnitude.

From the measured hyperfine structure one deduces the nuclear spin, I , the magnetic moment, μ_I , and electric quadrupole moment Q_S . Furthermore, the measured isotope shifts give the mean-squared charge radii. As an example of results obtained, the diagram shows the change in the mean square radii for a series of Ba isotopes. Note that the behaviour is far from the regular $A^{1/3}$ dependence predicted by the liquid-drop model, (indicated by the parallel lines) and furthermore, shows the effect of a spheroidal deformation characterized by the eccentricity parameter β .



Atomic Beam Magnetic Spectroscopy (ABMR)



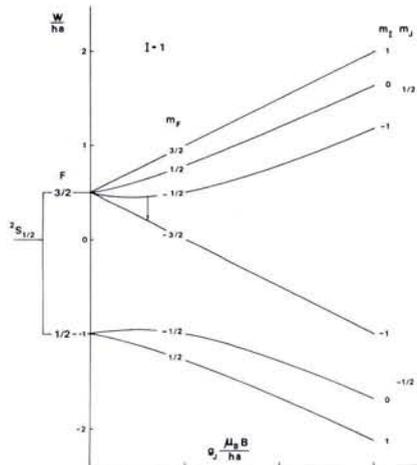
This technique (first introduced by Rabi in 1938) is the most powerful for determining nuclear spins and moments. The ion beam from the separator is directed on to a heated foil situated at the entrance of a six-pole magnet (A) and a beam of free atoms is formed by continuous evaporation from the foil. The beam is polarized in the inhomogeneous field from the A magnet and only atoms with negative effective magnetic moment pass into the central homogeneous field of the C magnet. In this region the orientation of the magnetic moment can be changed by radiofrequency-induced transitions. The four-pole B magnet with the centrally placed obstacle now serves as an analyzer so that only atoms that have changed the projection of the magnetic moment can pass through the machine.

The resonance transition frequency indicated in the diagram is given by the expression:

$$\nu = B \cdot f(I, J, F, g_J) + B^2 g(I, J, F, g_J, a, b) + \dots$$

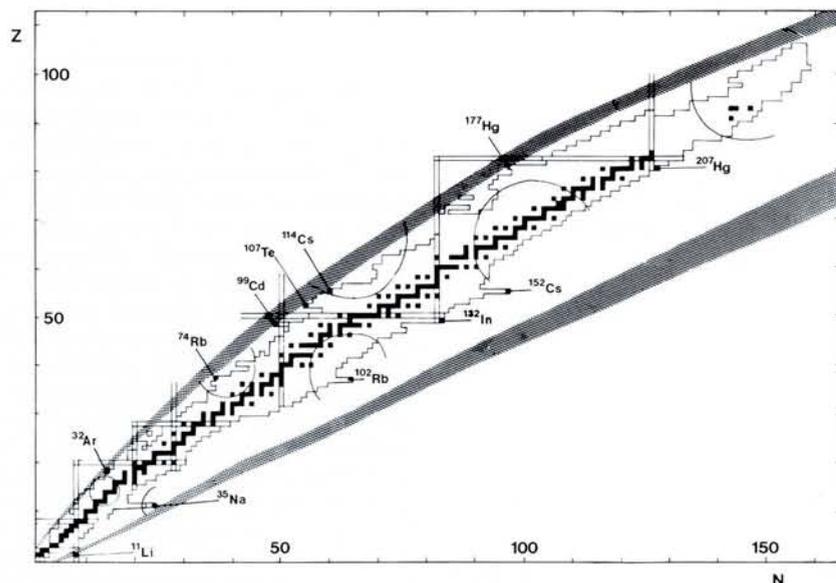
where B is the external magnetic field and a, b the magnetic and electric hyperfine coupling constants, respectively. If the electronic angular momentum J and the splitting factor g_J are known, a measurement of ν in a weak external field gives the nuclear spin I . Measurements in intermediate fields give in addition the a and b factors from which the magnetic dipole moment and the electric quadrupole moment can be determined.

The changes in nuclear spins and moments through long mass chains, give sensitive indications of the interplay between collective and single-particle phenomena in determining individual nuclear properties.



The Nuclear Chart: Closed Shells and Regions of Deformation

Panel 4



The "magic numbers" (2, 8, 20, 28, 50, 82, 126) corresponding to closed shells of neutrons/protons are indicated by the thin double lines. The cross-hatched areas correspond to the (predicted) limits of particle stability. Examples are given of the most neutron-deficient or neutron-rich isotopes found until now. With the exceptions of ^{107}Te (found at GSI, Darmstadt) and ^{132}In (found at the Studsvik reactor), all of the examples shown were first observed at CERN.

The nuclei near to the magic numbers have spherical equilibrium shapes while those far away from magic N, Z values tend towards strong ellipsoidal deformations and often show well-developed rotational spectra. The curved lines in the diagram indicate approximately the deformed regions; those near ^{35}Na and ^{102}Rb have been studied extensively at CERN during the past few years. The doubly magic ^{132}Sn ($Z = 50, N = 82$) is of special interest and has been the subject of detailed studies, both at CERN, at Studsvik (Sweden) and at the Kernforschungsanlage Jülich (FRG).

New Radioactive Decay Modes: Beta-Delayed Two-Neutron and Three-Neutron Emission

(Panel 5)

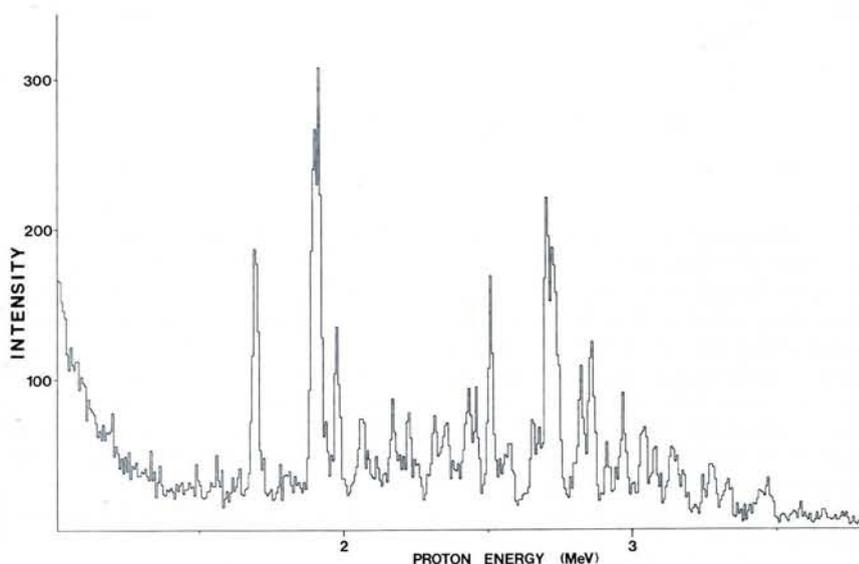
The extremely neutron-deficient or neutron-rich nuclei produced at ISOLDE are characterized by very high nuclear beta decay energies. At the same time, the daughter nuclei have low particle separation energies so that the beta decay may feed excited states that are particle unbound. The decay may then proceed via particle emission rather than gamma emission. Emission processes involving protons, neutrons and also alpha particles have been studied in detail (see next Panel) at ISOLDE. It has also been predicted that the most neutron-rich nuclides would allow the emission of several neutrons so that one would have beta-delayed multi-neutron emission. The first experimental observations of these new radioactive decay modes have been made at ISOLDE during the past two years. It has been found that the nucleus ^{11}Li exhibits both two-neutron and three-neutron emission. The detection of these new decay modes rests on measurements of neutron energies and coincidences by time correlation techniques.

Beta-delayed two-neutron emission has also been observed from three heavy sodium isotopes ($^{30-32}\text{Na}$) and it appears that this decay mode is quite common far from stability, and should be taken into account in calculations of the astrophysical r -process, the process through which heavy elements are assumed to have been created.

The abundance of the 2-n events is a sensitive test of the beta strength function at high energy.

Average and Statistical Properties of the Decays of far Unstable Nuclei: Nuclear "Noise Spectra"

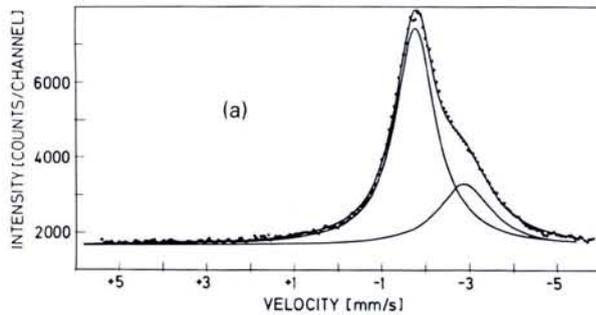
Panel 6



The high Q_{EC} , Q_{β} -values for far-unstable nuclei lead to extremely complex decays with very many intermediate states being populated. In a situation frequently encountered, the individual excited levels are still distinct and resolvable (although not resolved) in the experiments. The complexity, however, is such that the most profitable approach is one in which the nuclear structure is described in terms of local averages ("strength functions") and the fluctuations around the averages, an approach first developed in neutron physics. Because of many similarities to the properties of noise in electrical circuits, it is tempting to refer to the fluctuation phenomena as "nuclear noise".

As an example of fluctuation phenomena, the figure shows the spectrum of beta-delayed protons from the electron capture, β^+ decay of ^{99}Cd . The protons are emitted from excited levels at 4-6 MeV in the daughter nucleus ^{99}Ag . From the amplitude distribution of the peaks it is possible to derive the level-density parameter for ^{99}Ag . The low value obtained, $a = 8.5$ as compared with $a = 16-18$ for the heavier silver isotopes, is an indication that the influence of the doubly-magic ^{100}Sn is beginning to be felt, and the experiment provides — together with α -decay data — the first evidence for the expected stability of the 50-50 shells.

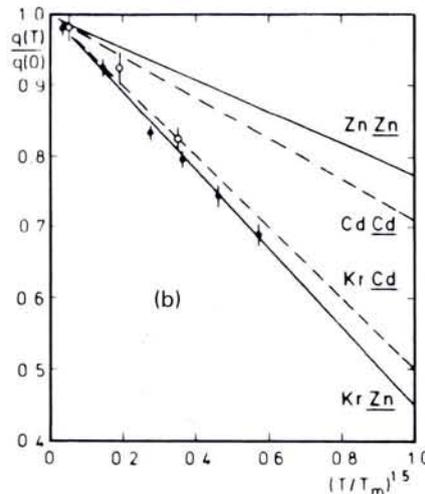
Radioactive isotopes produced by ISOLDE are implanted as probe atoms into solids. Nuclear hyperfine methods utilizing radiation emitted in the nuclear decay of these isotopes like Mössbauer spectroscopy and perturbed angular correlation techniques are applied to determine various properties of the impurity atoms (electronic densities, electric quadrupole and magnetic dipole interactions, lattice vibrations). It is inherent to these methods that defects formed with radioactive parent isotopes are probed with a daughter isotope. It has been shown that in this way radiogenic defects, which are not necessarily formed with the daughter, can be created. An absolute number of 10^{10} - 10^{13} impurities is sufficient for such experiments. A Mössbauer spectrum of the 24 keV γ -radiation of ^{119}Sn recorded within a few minutes from $\sim 10^{11}$ implanted ^{119}In ions in a GaAs single crystal is shown in Fig. (a).



The two lines correspond to Sn on substitutional Ga sites (high-intensity line) and Sn associated with vacancies that are created in the implantation process. The implantation of radioactive ^{119}In and ^{119}Sb into III-V semiconductors has been found to result in a selective population of III or V sites, respectively. Thus the nuclear transmutation to ^{119}Sn leads to a site-selective doping of these semiconductors with amphoteric Sn atoms acting as donors or acceptors, respectively. By this technique the electronic configuration of Sn dopants has now been studied by Mössbauer spectroscopy.

The interactions of lattice defects with impurity atoms are of considerable interest in semiconductors and metals. Since various impurity-defect structures are formed in the ion-implantation process (cf. Fig. a), studies of these defects are a natural field of interest at ISOLDE.

Fig. (b) shows, as an example of the results obtained from γ - γ perturbed-angular-correlation experiments, the temperature dependence of the electric field gradient of ^{79}Kr impurities in Zn and Cd in a normalized representation (on a $T^{1.5}$ scale normalized to the melting point T_m), together with the trend for the pure metals. The ^{79}Kr was inserted on substitutional sites by ion implantation of ^{79}Rb source activity. The steep slopes of the curves for Kr in Cd and Zn can be explained by low frequency local modes, characteristic for the inert noble gases impurity.



Atomic Physics Experiments

A combination of atomic beam and laser techniques has been used to detect the first optical transition in the element francium ($Z = 87$), the D_2 line at a wavelength of 7179.7 Å. This element was the only one with $Z < 100$ for which no optical transition had been reported; the difficulties were due to the short half-lives (the longest lived francium isotope has $T_{1/2} = 22$ min) and low production rates. The ISOLDE experiment was made with a separated beam of 10^8 atoms/s which gave an intensity sufficiently high for the optical experiments, which have now opened the way for a study of francium nuclei through their hyperfine structure. Further studies of the atomic physics of this rare element are being planned.

In another series of experiments, performed with a bent crystal spectrometer, the energies of K X-rays were measured with very high precision. It was already known that for the heavier elements, the energies depend to a small extent on the isotopic and chemical composition of the sample and the new experiments aimed at demonstrating additional contributions.

(i) The first effect observed was a selection rule in electron capture beta decay, which gives a non-statistical population of the 1s hyperfine levels. An illustration of the effect is shown in the Figure: the initial state is a nucleus with spin of $I-1$ and orbital angular momentum of zero, which gives a total angular momentum equal to $I-1$. The nucleus undergoes beta decay, with capture of an electron, to a state with nuclear spin I . The neutrino carries away half a unit of angular momentum, so that the remaining inner electron can only have its spin anti-parallel to the nuclear spin and the $F = I-1/2$ hyperfine component is populated, while the component with $F = I+1/2$ is forbidden.

(ii) A contribution from unresolved satellite lines has been shown to increase the energies of X-rays from photoionization with typically 200 meV.

(iii) A third effect observed originates in the extremely short lifetime of a 1s hole (10^{-16} - 10^{-17} s) which gives rise to a peculiar phenomenon: the atom left after electron capture beta decay with atomic number Z essentially retains the configuration of the atom ($Z+1$) until the X-ray is emitted. The X-ray energies are influenced in a characteristic way due to this effect and from the observed pattern of the energy shift one can learn about the electronic structure of the atoms involved in the decay.

Panel 8

