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Neutron-rich nuclei and fission; recent developments and future aspects

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Abstract. Production and studies of neutron-rich nuclei produced in fission are reviewed. Some recent experiments performed with the ISOL technique at the IGISOL and the ISOLDE facilities are reviewed. The manipulation of neutron-rich nuclei is discussed with a special focus on radioactive ion cooling and trapping techniques under construction worldwide. Perspectives of obtaining intense post accelarated beams of fission products are discussed.

PACS. 21.10.-k Properties of nuclei; nuclear energy levels – 24.75.+i General properties of fission – 25.85.-w Fission reactions

1 Introduction

Determination of properties of neutron-rich nuclei far from the valley of β-stability represents the first important step towards the understanding of nuclear structure and of possible new nuclear phenomena predicted to appear near the neutron drip line. Since production cross-sections of neutron-rich nuclei far from the valley of β-stability are generally low, efficient and selective experimental techniques are needed. Recently, intermediate-energy fission induced by low- as well as by high-energy protons combined with the ISOL technique have been shown to be powerful tools to search for and study neutron-rich isotopes of nearly all medium-heavy elements, including refractory ones. Some recent experiments performed at the IGISOL and the ISOLDE facilities will be reviewed. In the course of these studies, significant new information has been gained on the production cross-sections related to different modes of intermediate-energy fission.

Manipulation of neutron-rich nuclei is discussed with a special focus on cooling and trapping techniques for neutron-rich fission product ions. Perspectives of obtaining intense post-accelerated beams of fission products and their potential uses are briefly discussed.

2 Production of neutron-rich nuclides in fission

2.1 Thick target production at ISOLDE

At ISOLDE, radioactive nuclides are produced in thick high-temperature targets via spallation, fission or fragmentation reactions. For a review of ISOLDE physics the reader is referred to ref. [1]. The targets are placed in the external proton beam of the PS Booster, which has an energy of 1 or 1.4 GeV and an intensity of about 2 μA. Until now more than 600 isotopes of more than 60 elements ($Z = 2$ to 88) have been produced, in some cases with half-lives down to milliseconds and intensities up to 10^{11} ions/s. Neutron-rich nuclei are produced in fission reactions, either directly with very energetic protons or indirectly by the secondary neutrons of considerably lower energy. The commonly used targets are uranium or thorium carbide targets, which provide the optimum release properties for a large number of elements. In a typical experiment, protons bombard a 20 cm long target consisting of 50 g/cm^2 of uranium and 10 g/cm^2 of graphite. Depending on the element to be extracted the target can be heated up to 2000 °C temperatures. Radioactive atoms released are then diffusing through the transport line into a plasma or a surface ion source, or a hot cavity for resonant laser ionization. Presently, the laser ionization schemes developed include fission product elements Ni, Cu, Zn, Ga, Ag, Cd, In and Sn [2]. The release properties of various elements from the thorium and uranium targets are reported in ref. [3]. The shortest-lived fission products separated with this setup are very neutron-rich Rb isotopes with half-lives as short as 30 ms. Neutron-rich nuclei of refractory elements from yttrium to Pd are not available as beams at ISOLDE.

2.2 Thin target production at IGISOL

The combination of the ion-guide isotope separator on-line (IGISOL) technique and the intermediate-energy

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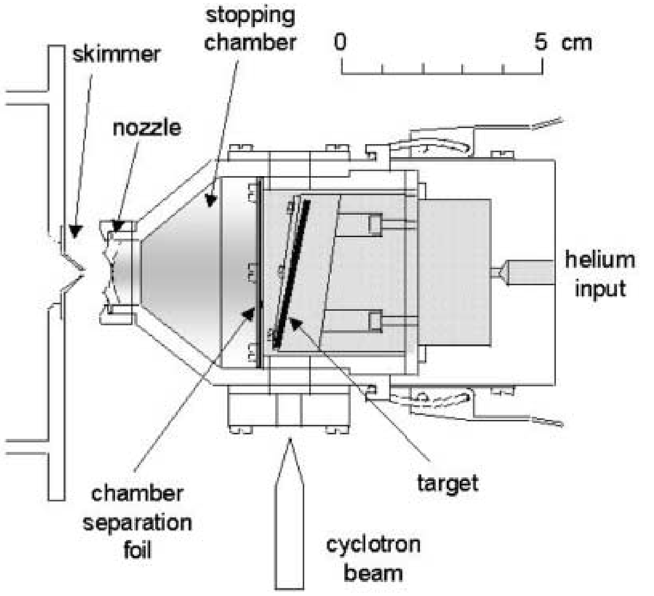

Fig. 1. The fission ion guide presently in use at the Department of Physics, University of Jyväskylä.

fission has been shown to be a powerful as well as a universal tool to search for and study neutron-rich isotopes of all medium-heavy elements including refractory ones, for recent reviews, see refs. [4,5]. Radioactive isotopes of interest are produced in proton-induced fission of ^{238}U and mass separated using the upgraded ion-guide isotope separator IGISOL systems, mainly at Jyväskylä [4] and Leuven [6], but also at Sendai in Japan [7]. A schematic drawing of the fission ion guide operating at Jyväskylä is shown in fig. 1. Fission products emerging from the actinide targets following light-ion bombardments are emitted nearly isotropically. This allows easy separation of fission fragments from the primary beam, and can remove any potential problems due to ionization by the intense primary beam. The basic idea of the ion guide is to slow down and thermalize initially energetic fragment ions in gas, which is typically helium, sometimes argon. Ions are transported by a gas flow out of the gas cell and injected through a differentially pumped electrode system to the high-vacuum section of the isotope separator for further acceleration and separation by mass. Employing a 30 MeV proton beam, the fission rate in the target is about $3{\cdot}10^9$ fissions/s μC. A quantitative measurement of the yield for the $A = 112$ fission product ^{112}Rh gives a total efficiency of about 0.02 % with a beam intensity of 10μA. This corresponds to a yield of the order of 10^5 ions/s.

2.3 Mass and isotope distributions

Nuclear fission is a promising source for discovering, producing, and investigating exotic nuclei with high neutron excess. Several new neutron-rich nuclei have been discovered at ISOL facilities which employ proton-induced fission [8]. A large number of new fission products have been discovered in the projectile fission of uranium at relativistic energy on a Be-target using the fragment separator, FRS [9]. The fission process is also considered to be very promising for the production of neutron-rich nuclides in different Radioactive Nuclear Beam (RNB) projects. Among these the most important ones are

- thermal-neutron–induced fission of ^{235}U [10];
- fast-neutron–induced fission of ^{238}U [11];
- photon-induced fission of ^{238}U [12].

The intensity of RNB is determined by the expression

$$I = \sigma \times L \times \prod_i \varepsilon_i, \qquad (1)$$

where σ is the production cross-section (fission), L is the luminosity, and ε_i are efficiency coefficients for different stages of processing and extraction of the beam. Now we need a more deep insight into the mechanism of the fission process to predict the cross-section formation of neutron-rich fission products for a wide range of excitation energies and a large diversity of compound nuclei.

During the last years systematic studies on charge and mass distributions of fragments in proton, deuteron, and neutron-induced fission of ^{238}U at intermediate energy have been carried out at the Accelerator Laboratory of the University of Jyväskylä [13–16]. Using IGISOL technique independent yields of neutron-rich products in the very asymmetric proton-induced fission of ^{238}U at $E_{\rm p} = 25$ MeV have been measured [13]. A very large enhancement of yields was found in comparison with the low-energy neutron-induced fission. Enhancement of fission fragment yields in the far asymmetric region is also observed in time-of-flight measurements with HENDES setup with $E_{\rm p} > 20$ [14] and in the neutron-induced fission of ^{238}U using the reaction $^{238}{\rm U}(d, pf)$ [15]. In fig. 2 the fission fragment yieds at $A < 85$ measured in the proton-induced fission of ^{238}U at $E_{\rm p} = 20$ and 60 MeV [14], are compared with yields obtained with LOHENGRIN mass-separator in the thermal-neutron–induced fission of ^{235}U [17] and $^{242{\rm m}}$Am [18]. According to our theoretical model [13], developed for the calculation of fission product yields, in the case of a very asymmetric mass division at intermediate excitation energy ($E_{\rm c} \geq 20$ MeV) there are contributions from three components: the tails from the symmetric and the second asymmetric modes, and the superasymmetric mode. The superasymmetric fission mode is connected with nuclear shells $Z = 28$ and $N = 50$ (^{78}Ni mode). It is important for producing nuclides near the doubly magic nucleus ^{78}Ni detected in the fission induced by the peripheral collision of 750 MeV/nucleon projectiles of ^{238}U on a Be target [19].

According to eq. (1) the beam intesinty depends on the intensity of the primary beam current, the target thickness and the efficiency of extraction of the secondary beam. In fig. 3 the intensities of Cu radioactive beams obtained at different RIB facilities are presented. One can see from fig. 3 that very high intensities have been achieved at ISOLDE due to favorable release and ionization conditions (full circles) [20]. Other ISOL facilities: GSI-ISOL (dotted circles) [21–24], LISOL (grey squares) [25], IGISOL

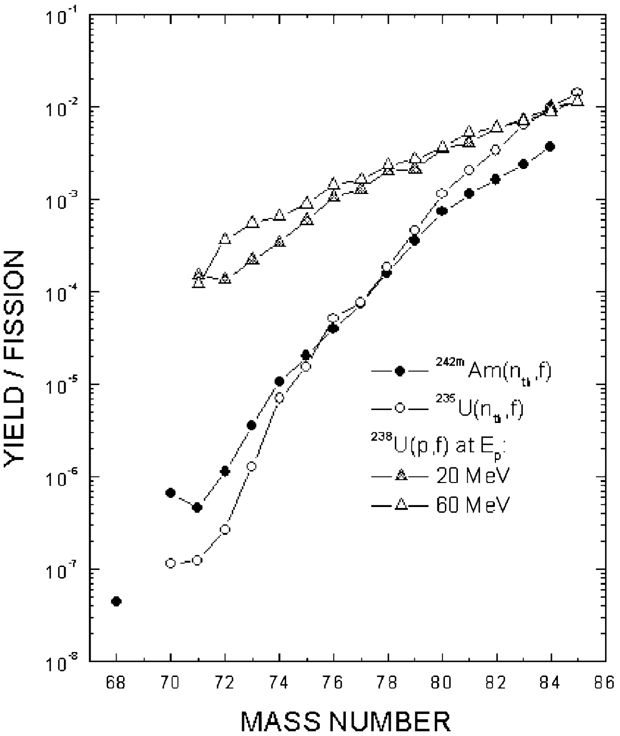

Fig. 2. Comparison between the fission fagment yields measured in the proton-induced fission of ^{238}U at $E_{\mathrm{p}} = 20$ and 60 MeV and the yieds obtained with LOHENGRIN mass-separator in the thermal-neutron–induced fission of ^{235}U and $^{242\mathrm{m}}$Am.

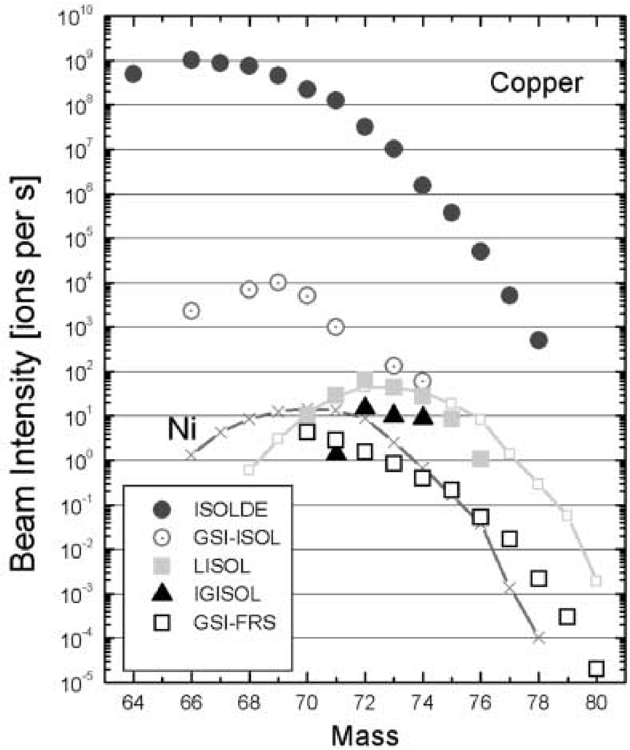

Fig. 3. Comparison of Cu yields at present RIB facilities: ISOLDE (full circles); GSI-ISOL (dotted circles); LISOL (grey squares); IGISOL (up-triangles); GSI-FRS (open squares). Lines present predictions for Co and Ni fission product yields in ^{238}U(p(30 MeV), f) normalized to LISOL data.

(up-triangles) [13] give lower intensities. At the in-flight spectrometer FRS at GSI the very neutron-rich Cu isotopes were detected up to $A = 80$ but with considerably lower rates (open squares) [19]. Our predictions for Cu and Ni fission product yields, normalized to LISOL data, are shown in fig. 3 as lines with small symbols. The predicted ^{78}Ni nucleus production cross-section is about 0.8 nb, that is close to the value of 0.3 nb estimated in ref. [19].

Comparative investigations of neutron-rich fission product yields in different reactions are important for the development of RNB facilities. In fig. 4 calculated Ni isotope production cross-sections are compared for four fission reactions: $^{235}\mathrm{U}(n_{\mathrm{th}}, f)$, $^{238}\mathrm{U}(p, f)$ and $^{238}\mathrm{U}(n, f)$ at $E_{\mathrm{p(n)}} = 50$ MeV, and $^{238}\mathrm{U}(\gamma, f)$ at $E_\gamma = 14$ MeV. One can note that the production of very neutron-rich Ni isotopes in the fast-neutron–induced fission of ^{238}U is higher than in the thermal-neutron–induced fission of ^{235}U. This is due to the enhancement of mass yield for the very asymmetric mass division and higher mass dispersion. The neutron-rich Sn beams are at the top of the list of the requests from RNB facilities. Predictions of Sn isotope production cross-sections for the above-mentioned fission reactions are shown in fig. 5. Production of extremly neutron-rich nuclei is higher in the fission of a more neutron-rich compound nucleus. In this sense, the fast-neutron–induced fission of ^{238}U should provide a promising method for production and investigation of extremely neutron-rich isotopes.

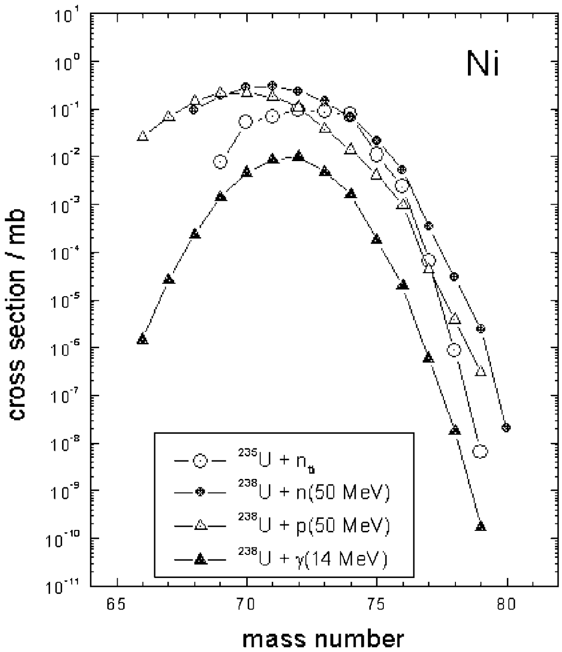

Fig. 4. The Ni isotope production cross-sections in reactions: $^{235}\mathrm{U}(n_{\mathrm{th}}, f)$ (open circles), $^{238}\mathrm{U}(n, f)$ (solid circles), and $^{238}\mathrm{U}(p, f)$ (open triangles) at $E_{\mathrm{p(n)}} = 50$ MeV, and $^{238}\mathrm{U}(\gamma, f)$ at $E_\gamma = 14$ MeV (full triangles).

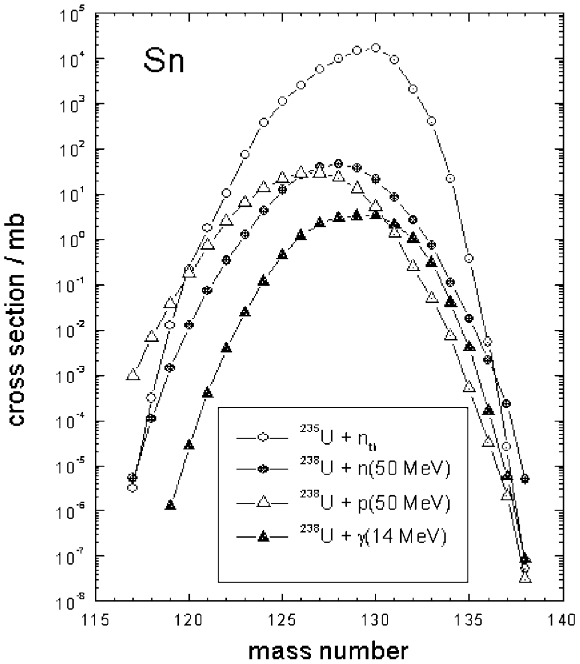

Fig. 5. The Sn isotope production cross-sections in reactions: $^{235}\mathrm{U}(n_{\mathrm{th}}, f)$ (open circles), $^{238}\mathrm{U}(n, f)$ (solid circles), and $^{238}\mathrm{U}(p, f)$ (open triangles) at $E_{\mathrm{p(n)}} = 50$ MeV, and $^{238}\mathrm{U}(\gamma, f)$ at $E_\gamma = 14$ MeV (full triangles).

3 Some recent spectroscopy studies of fission products

Interest in studies of neutron-rich nuclei in the intermediate mass region is manyfold. The evolution of the nuclear shell structure towards very large neutron numbers is a key element in these studies. Precise and broad systematic investigations are needed on ground-state properties such as masses, radii and moments as well as on excited states. These studies are necessary for the full understanding of the evolution of the nuclear potential and related phenomena like pairing and spin-orbit coupling. These experiments carry also relevance for understanding the synthesis of chemical elements via the rapid neutron capture process.

Recent experiments at ISOLDE have mainly concentrated on very light neutron-rich Mn fission products [26] as well as extremely neutron-rich Ag and Cd isotopes at or near the r-process waiting point $N = 82$. These studies also include the mass measurements with ISOLTRAP on cesium isotopes up to ^{142}Cs and barium isotopes up to ^{144}Ba [27] as well as those for $^{128-130,132}$Sn isotopes [28].

The experiments at Jyväskylä and Louvain-la-Neuve, using the IGISOL technique, have concentrated on decay and structure properties of neutron-rich nuclei between deformed Zr and spherical Sn isotopes, and on neutron-rich isotopes of Co, Ni and Cu close to the doubly magic nucleus ^{78}Ni, respectively. All in all more than 40 new decays and isotopes have been studied in these experiments. In the course of these studies significant new information has also been gained on the production cross-sections related to the intermediate-energy symmetric fission and a highly asymmetric fission, as described above in sect. 2.

3.1 Recent studies at the frontier of neutron-rich nuclei

The yield studies described in sect. 2 show that intermediate-energy fission provides the most promising approach for spectroscopy towards ^{78}Ni. A standard IGISOL employed at Jyväskylä and Sendai has had difficulties in studies of these nuclei, mainly due to the strong contaminants caused by the doubly charged fission products with masses around 140 to 150. This problem is overcome in the resonant-laser scheme where ions are first neutralized before selective ionization. Studies performed at Louvain-la-Neuve have yielded information on excited sates and transitions of $^{66-70}$Ni [29–31] and $^{68-74}$Cu [32]. An important general conclusion, drawn from the comparison to the valence mirror nuclei in the $N = 50$ region as well as from the shell-model calculations, is that collective excitations play increasingly important role with increasing occupation of the $\nu g_{9/2}$ orbital. Recent experiments at ISOLDE have demonstrated very high yields for copper isotopes, which were detected up to ^{79}Cu, and provide great potential for future studies of these nuclei [33].

Since in the early 80's very little information existed on neutron-rich nuclei in the $A = 100$–120 mass region, the first experiments at IGISOL were the search experiments for new isotopes and isomers as well as measurements of gross properties of their decays, especially their β-decay half-lives [34,35]. Later these measurements were extended to new β-delayed neutron emitters, such as ^{104}Y, ^{110}Nb and ^{114}Tc, about 16 neutrons away from the β stability [8,36]. The β-decay half-life ($T_{1/2}$) and the delayed neutron emission probability (P_{n}) are the first measurable gross β-decay properties for nuclei very far from stability produced in low production yields. They contain important nuclear-structure information; $T_{1/2}$ is sensitive to low-lying β-strength and P_{n} carries information on strength just above the neutron separation energy.

Neutron-rich nuclei in the Sn region are produced via fission in high yields at ISOLDE. The experiments employing selective laser ionization have been able to detect silver isotopes up to $^{130}\mathrm{Ag}_{83}$ [37], cadmium isotopes up to $^{132}\mathrm{Cd}_{84}$ [38], indium isotopes up to $^{134}\mathrm{In}_{85}$ [39] and tin isotopes tentatively up to $^{137}\mathrm{Sn}_{87}$ [40]. The fast release of the uranium carbide target is demonstrated by the observation of half-lives less than 100 ms for Ag and Cd. These new data provide a basis for estimates of the β-strength distributions and decay Q-values, and are of high value for the astrophysical calculations concerning the r-process scenario. It is of interest to note that, using the IGISOL technique, the heaviest palladium isotope observed is $^{120}\mathrm{Pd}_{74}$ [41] and that of rhodium is $^{118}\mathrm{Rh}_{73}$ [35], *i.e.* several neutrons closer to the valley of stability than the heaviest observed Ag-Sn isotopes.

The status of our knowledge on neutron-rich nuclei near $N = 50$ and 82 is shown in fig. 6. It is quite obvious that in the future, experiments will have to work with

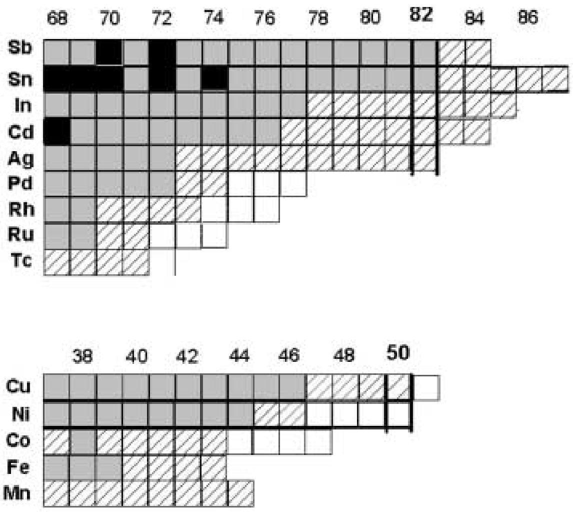

Fig. 6. Sections of the chart of nuclides below $N = 50$ and $N = 82$ displaying our present knowledge. The black squares denote stable isotopes. The shaded and lined squares denote isotopes whose β-decays have been observed. The lined squares are isotopes with half-lives less than 1 s. The open squares denote isotopes identified only in projectile fission experiment at GSI.

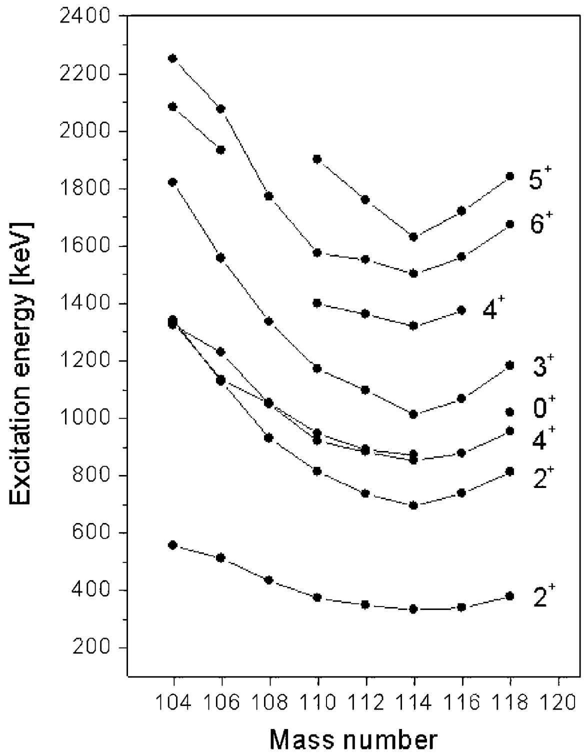

Fig. 7. The systematics of collective low-lying states of neutron-rich palladium isotopes. All data from $A = 112$ to $A = 118$ are from IGISOL experiments, (see refs. [35,5]).

isotope-pure sources which can be provided by two different approaches, by employing selective laser ionization or by employing purification and bunching by ion traps.

3.2 Structure studies of Pd and Cd isotopes

Neutron-rich Ru, Pd and Pd isotopes belong to a transitional region between the spherical closed-proton-shell Sn isotopes and the strongly deformed Zr and Sr nuclei at the proton midshell. Beta-decays of odd-odd neutron-rich isotopes of Nb, Tc, Rh and Ag have provided a wealth of information on the low-lying collective structures of their daughter isotopes. These experiments have produced also crucially important experimental information on the low-lying states on which band structures have been built by employing prompt fission fragment gamma-ray coincidence techniques.

The systematics of low-lying levels of neutron-rich palladium isotopes have been studied extensively at IGISOL [5] and recently also by using in-beam studies of prompt gamma-rays emitted in fission and correlated with fragments [42]. This has allowed the identification of the yrast band structures up to spins as high as 18^+. Decay studies have provided important information on the low-lying states on which the bands extending to higher spins could be observed and identified. Recently, we have reported on the first observation of the β-decay of ^{118}Rh and the low-lying level structure of ^{118}Pd [35].

Beta-decays of even-even Rh isotopes are characterised by the existence of one or more isomeric states leading to the feeding of a rich variety of states, exhibiting collective and multi-quasiparticle features. The systematics of energy ratios, like $E(4^+)/E(2^+)$ and $E(6^+)/E(2^+)$ studied in ref. [43] for even $^{110-116}$Pd implies an increasing trend up to ^{116}Pd. At $N = 68$ the ground-state deformation reaches its maximum. This is almost in the middle of the shell, which is located at $N = 66$. The systematics of collective low-lying states of neutron-rich Pd isotopes is shown in fig. 7. New results show a smooth transition to higher excitation energies. This suggests decreasing deformation when the neutron shell $N = 50$–82 starts to fill. This change can be interpreted as a transition from the $SU(5)$ (vibrator) to the $O(6)$ (γ-unstable) symmetry. Data for ^{118}Pd show that the $O(6)$ symmetry has reached its maximum at $A = 116$ ($N = 70$) and sudden transition back to the $SU(5)$ symmetry occurs between $A = 116$ and $A = 118$.

In a study of excited structures of heavy even-even cadmium isotopes ^{126,128,130}Cd, Kratz *et al.* have claimed for the observation of the possible weakening of the $N = 82$ neutron shell far from stability [37]. This important effect has only been deduced from the comparison of the $E(2^+)$ and $E(4^+)/E(2^+)$ level systematics and model predictions for Pd, Cd and Te isotopes. Any further firmer interpretation would require substantially more detailed spectroscopic studies of the low-lying collective level structures of these isotopes. Such measurements have been recently initiated at IGISOL in Jyväskylä. The first results on ^{116,118,120}Cd isotopes provide a substantial amount of new information on the collective states in even-even Cd isotopes. Among other features, these results include the completion of the three-phonon quintuplet in ^{116}Cd and show the very narrow energy spread, only 70 keV between the lower and upper members of the multiplet [44]. Extension of the knowledge on two- and three-phonon multiplets to heavier Cd isotopes could provide a new insight into the evolution of possible collectivity as suggested by Kratz *et al.*

4 Manipulation of fission product ions

New concepts to improve the purity and the quality of the ion beams are the buffer-gas filled RFQ for ion beam cooling and bunching and the Penning trap for isobar purification. Recently, a concentrated effort has been taken to develop these techniques for improving the quality of low-energy radioactive ion beams. Following the pioneering work at ISOLDE several projects are underway in Europe [45,46]. In the following we concentrate on the system under construction at IGISOL in Jyväskylä for improving the experiments on neutron-rich nuclei from fission.

Two novel techniques are under development, the facility for collinear laser spectroscopy and the buffer-gas RFQ cooler [47] followed by the Penning trap [48] for isobar separation and mass measurements. Both of these technologies will provide unique opportunities for spectroscopic studies of very short-lived nuclei of all elements, including highly refractory ones. The universality and the speed of the IGISOL principle can be maintained and the beam quality improved by orders of magnitude. The isotopic purity is important for the low-production rates necessarily encountered when the studies are being pushed further from the present boundary of known neutron-rich nuclei. The construction of the ion cooler has been completed. The first successful tests performed with radioactive ^{112}Rh ions have demonstrated successful beam cooling with more than 60 % transmission [47].

The success of the on-line collinear laser spectroscopy approach for refractory elements was recently demonstrated by the measurements on radioactive ^{170}Hf, ^{172,173,174}Hf isotopes produced in proton-induced fusion reactions [49]. The next step will be to extend these measurements to refractory neutron-rich nuclei in a similar manner as was done for the neutron-rich Ba isotopes at IGISOL [50]. These measurements are expected to benefit tremendously from the improvement of the IGISOL beam quality when cooled and bunched in the RFQ cooler.

5 Fast radioactive beams of neutron-rich nuclei

There are two basic methods to produce fast radioactive beams. One relies on the Isotope Separator On-Line (ISOL) method and the other one is called In-Flight, see ref. [25] for extensive presentations of both techniques. In the ISOL method radioactive isotopes are first produced at rest by a driver accelerator in a thick target, a catcher or an ion guide. After releasing from the target and following ionization and acceleration up to a few 10 keV energy through a mass selective device, these nuclei are accelerated in a post-accelerator typically up to 2–50 MeV/u energy. In the In-Flight method the initially energetic (50–1000 MeV/u) ions are fragmented or fissioned in flight and subsequently transported to a secondary target after their purification in mass, charge and momentum distribution. The resulting beams at the ISOL and In-Flight RNB facilities are truly complementary in their energy, intensity and isospin range. Therefore both types of facilities are intensively pursued world wide today.

At CERN the ISOLDE facility, which operates using the 1–1.4 GeV protons from the PS-Booster to produce radioactive ion beams of more than 60 elements, is used to deliver low-energy radioactive ions to the REX-ISOLDE LINAC-type post accelerator. This concept employs a novel charge breeding principle based on the coupled Penning trap and the EBIS charge breeder. In addition, the ISOL-based facilities providing beams of fission product ions are being launched in Europe at the new high-flux reactor in Munich [10], at the SPIRAL II facility at GANIL [11] and in Dubna [12]. In these concepts, fission products are to be produced in fission induced by high-energy protons and secondary neutrons (CERN), protons or deuterons and secondary neutrons (GANIL), or by photons (Dubna, GANIL). Furthermore, a new plan is proposed in the UK employing a 200 MeV proton driver accelerator.

In North America, ISOL-based RNB facilities or projects are planned or are operational at ORNL in Oak Ridge, at TRIUMF in Vancouver, at ANL in Argonne and at LBNL in Berkeley. At ORNL, the driver accelerator is a 100 MeV proton cyclotron and the post-accelerator is a 25 MV Tandem. The facility has recently started its physics programme and accelerated fission product ion beams are available with intensities in the 10^5 to 10^6 ions/s range. The ISAC project at TRIUMF uses a 500 MeV, 100 μA proton synchtrotron as a driver accelerator. The post-accelarator is LINAC producing RNBs up to 1.5 MeV/u, a second stage up to 7 MeV/u was recently approved.

The future plans in Europe include a substantial upgrade of the GSI facility to produce ultimately 2 GeV/u uranium beams with intensity up to 10^{12} ions/s with a fully upgraded fragment separator as well as cooler and electron rings for versatile programmes on exotic nuclei. Similarly, the EURISOL study project funded as the RTD project of the European Union aims at producing a preliminary design for the second-generation ISOL-based RNB facility with qualifications second to none existing at the moment. Major projects for the next-generation ISOL RNB facilities are also proposed in the US as the RIA project and in Japan at RIKEN and the Japan hadron factory.

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