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# **ISOLDE** past, present and future

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# Abstract

The idea of production of short-lived radioisotopes with the on-line technique has it roots in the early 1950s. In 1964 this became a reality when CERN approved an experiment at the 600 MeV proton synchro-cyclotron, the SC. The first experiments were performed in 1967 and since then the ISOLDE programme has gradually developed into a major undertaking. Since 1992 the ISOLDE Radioactive Beam Facility is linked to an external proton beam from the PS Booster. Today this 50 years old 'lady' is more vital than ever. With the successful start of HIE-ISOLDE in 2015 one may conclude that the facility is ready to face the next half century with the boost of the success and the necessary knowledge to face new challenges. In this introductory article we give an overview of the history and pick up a few examples along the nuclear chart as illustrations of the experimental achievements.

Keywords: nuclear structure, radioactive decay and in-beam spectroscopy, radioactive beams, properties of nuclei

(Some figures may appear in colour only in the online journal)

# 1. Introduction

The ISOLDE Radioactive Ion Beam facility is one of the most senior installations at the CERN accelerator complex. With 50 years of successful operation ISOLDE now enters its second half-century with excellent prospects for its future. This laboratory portrait is a follow-up of a similar series of articles written almost 20 years ago [1]. The introductory chapter in that volume [2], gave an overview of the achievements during the first three decades of ISOLDE operation. When CERN celebrated its 60th anniversary in 2014 a book entitled '60

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**Figure 1.** The upper photo shows the new ISOLDE building as it was in 1992. In this camera angle the photo is to some extent dominated by the target area. The protecting soil on its top became a small forest in the following years. The large picture from 2016 is totally dominated by the cryogenic installations for HIE-ISOLDE with two huge liquid He dewars in front of the target-area forest to the left.

*Years of Experiments and Discoveries'* was edited [3] and the ISOLDE Facility got its obvious place also in that volume [4].

In this contribution we build upon these earlier chapters and give some examples of major developments and achievements from the mid-nineties, when the new ISOLDE Facility at the proton synchrotron booster (PSB) could deliver beams to the entire Community. We then continue with the successive progress towards today's installation and dare to give some thoughts about the coming years. We finally select experimental highlights, which we consider might be a guide for the expectations of the immediate and even the distant future.

#### 2. CERN and ISOLDE

The CERN management had decided that the operation of its first accelerator, the SC, should come to an end in 1990. The future of the ISOLDE physics at CERN had been discussed several times over the years [5], and even so in connection with the SC shut-down. With strong support from the Director General Carlo Rubbia and the Research Director Pierre Darriulat, the CERN Council decided to let the ISOLDE programme continue at CERN. The solution was that the ISOLDE Facility was moved to a new site closer to the main accelerator complex at CERN, with a dedicated new experimental hall and its targets irradiated with a 1 GeV proton beam from the CERN PS Booster [6]. Being part of the main CERN accelerator complex opened up new possibilities for the technical developments of the installation. The layout of the PSB ISOLDE building was carefully planned in its new environment so that it



**Figure 2.** Birds-eye drawing of the ISOLDE experimental hall. The inset shows a photo of an ISOLDE target-ion-source module.

gave perspectives for further growing and expansion (see figure 1). This foresight of the ISOLDE Technical Committee was indeed very important since the experiments of today, using beams from the HIE ISOLDE post-accelerator, are placed in the third successive extension of the experimental hall. The new technical challenges encountered at the PS-Booster, with pulsed beam, higher proton-beam energy and intensity gave new opportunities for expanding the experimental programme.

In the new layout of ISOLDE experimental hall, as shown in figure 2, the 1 GeV proton beam from PS Booster could be directed to either one of two independent target-ion-source units coupled to two conceptually different isotope separators. The first, of a more classical type, with one single 70° doubly focussing bending magnet, is referred to as the generalpurpose-separator. The ion beams from this separator pass an electrostatic switch-yard allowing data taking in up to three simultaneous experiments. The second separator is a reconstructed version of the two-magnet isotope separator, which was used for a relatively short time-period with its target placed in the SC vault, and referred to as ISOLDE 3 [6]. Due to space problems at the SC this separator had its magnets placed with the same bending direction making a double cross-over of the beam between them necessary. This could be avoided in the new hall. The reconstructed separator is referred to as the high resolution separator (HRS). Its two magnets, 90° and 60°, are coupled together, and with a sophisticated set of beam-correcting elements allowing a mass resolution of up to  $M/\Delta M \sim 7000$  [6].

The ISOLDE PS Booster Facility got an excellent start with one completed physics experiment already during the first scheduled beam-time in June 1992. This first experiment aimed at studies of the lightest bound Ne isotope, <sup>17</sup>Ne, which is a two-proton halo candidate. Its first-forbidden beta decay to the only bound excited state in <sup>17</sup>F and the role of the proton halo structure for the decay strength were studied [7].

A major change in the ISOLDE organisation came in the year 2000 when the different technical experts in the in-house group were incorporated in larger CERN groups with similar expertise. They were mainly attached to the PS Division, as the machine department was called at that time. This meant, to begin with, a loss of the benefits from a style, where all experts worked under the same roof to a new organisation where larger CERN groups took over tasks like vacuum, beamlines, beam instrumentation and diagnostic. Further the machine controls were gradually adapted to the CERN standard. At the same time the synergy gain and the mutual exchange of expertise knowledge were to the benefit of all parties. For ISOLDE all these changes slowly grew into what is now a well-functioning machinery.



**Figure 3.** The ISOLDE 'hot' area with the new MEDICIS building. The upper insets are (left) the robot used for target manipulations and (right) an ISOLDE target without vacuum container. The inset in the lower part of the figure shows the target storage shelves in MEDICIS.

During the first years of running at the PS Booster the ISOLDE targets were bombarded with protons with an energy of 1 GeV. In 2004 the energy was increased to 1.4 GeV, an increase that meant higher production cross sections for spallation and fragmentation reactions. With  $3.3 \times 10^{13}$  protons per pulse and an allocation of about half of the number of pulses from the so-called Super-cycle give a total of about 2  $\mu$ A average proton current to the ISOLDE target. This resulted in more intense radioactive beams delivered to the ISOLDE experiments to the benefit of the user community.

The handling of the ISOLDE targets is totally automated by using industrial robots, as shown in figure 3. The robots were renovated in 2013 during the first long shutdown of the CERN machines, LS1. In 2005 a Class A laboratory was built so that it became possible to safely handle open radioactive sources and targets. It is equipped with fume cupboards and has full protective measures and aerosol monitoring. It can, for example, handle 150 g UO<sub>2</sub> per day, corresponding to one ISOLDE target container.

In the years to come the energy of the PS Booster protons will be increased to 2 GeV. For ISOLDE this means that the dipoles of the proton beam transfer line need to be upgraded to cope with the higher energy. Further the ISOLDE targets might get as much as 6  $\mu$ A protons, which in turn implies a modification of the beam dump so that it can withstand a 13 kW deposited beam. With an increase in the production cross sections up to a factor of ten due to the energy boost and the linear increase by a factor of three in proton current one will obtain a factor 30 higher intensity for some of the delivered radioactive beams. This explains the 'I' in the acronym HIE: high intensity and energy is the name given to the upgrade of the facility.

#### 3. ISOLDE target-ion-sources and produced radioactive beams

The success of the ISOLDE experimental programme is to a very great extent due to a vital and continuous development of new radioactive beams and refinements of the existing ones. The target-ion-source (see insets in figures 2 and 3), where the main principles of construction have roots in the classical design from the initial phase of the ISOLDE 2 upgrade [8], is often referred to as 'The Heart' of the Facility. Target temperatures up to 2000 °C and different combinations of target material and dedicated ion-sources make the different target-ion-source units 'chemical factories' that give access to some 1300 different isotopes from 75 of the chemical elements in the periodic table.

Many of the elements produced at ISOLDE are released from oxide powders. A problem with such a target matrix is that the oxide sinters at high temperature, giving reduced release rate. To cure this problem zirconia and ceria targets consisting of oxide fibres were developed, giving significantly higher production yields for the short-lived isotopes of Cu, Ga and Xe [9].

The traditional ion sources used at ISOLDE were based on either surface or plasma ionisation. A giant step of improvement came with the development of RILIS<sup>5</sup>, which is a laser based ion source [10, 11]. Excitation schemes for 37 elements have been developed. As an addition to the RILIS technique a combination of the laser ion source with a trap (LIST) has been developed. By making the laser ionisation inside a radio-frequency quadrupole ion guide the surface-ionised contaminants may effectively be suppressed [12].

The first target material used for production of Ar isotopes was vanadium carbide, VC, which resulted in the first observation of a  $T_z = -2$  isotope, <sup>32</sup>Ar [13]. By replacing the VC matrix with CaO powder an increase by a factor 1000 in the production yield was obtained [14]. A problem with these CaO targets was, however, their unstable yields. To cure this problem a nano-structured CaO matrix was developed [15], which give rise to stable production yields over longer periods of time, since it could be operated at temperatures lower than traditionally used.

For the production of neutron-rich fission fragments a solid tungsten proton-to-neutron converter has been used for many years. This reduces the neutron-deficient isobaric contaminants since mainly fission fragments are produced. An improved converter design has been obtained based on simulations using energy-dependent cross-sections for proton and neutron induced fission. Recently this new type of converter was successfully tested experimentally [16].

For many experiments the beam quality is of major importance. To achieve reduced emittance and energy spread an ion cooler and buncher, ISCOOL [17], was developed and installed in the HRS separator section of ISOLDE. This resulted in strongly reduced emittance and energy spreads for all experiments downstream the beam line. Using its bunching capabilities, ISCOOL permits an increase in the sensitivity for experiments such as those devoted to collinear laser spectroscopy. The first physics run with this device took place in July 2008 when the isotope <sup>77</sup>Ga ( $T_{1/2} = 13$  s) was studied with the COLLAPS laser spectroscopy setup.

One of the latest elements for which a radioactive beam was produced at ISOLDE is boron (Z = 5). To illustrate the beauty of the technique we give here some details of the method of production that resulted in the observation of the isotope <sup>8</sup>B ( $T_{1/2} = 770$  ms) as a new ISOLDE beam in 2015. The target matrix consisted of 14.85 g of multi-walled carbon nanotubes. This resulted in an effective surface area of 4455 m<sup>2</sup>. To facilitate the release of the produced boron isotopes a SF<sub>6</sub> gas was introduced via a calibrated leak. The gas reacted

<sup>&</sup>lt;sup>5</sup> Resonance ionisation laser ion source.

with boron to form gaseous  ${}^{8}BF_{2}^{+}$  ions that were transferred to a VADIS 7, Versatile Arc Discharge Ion Source [18] via a cooled Cu transfer line. The future prospects for the  ${}^{8}B$  physics are great, the expected proton-halo ground state will be studied in reaction experiments at REX and HIE-ISOLDE, resonance scattering on a thick hydrogen target will yield information about excited states in  ${}^{9}C$  and studies of diffusion in semiconductors utilising  $\alpha$  emission channeling, these are two examples of future use of the  ${}^{8}B$  beam.

A new development, giving access to germanium (Z = 32) beams, was successfully achieved by extracting germanium as <sup>64</sup>GeS<sup>+</sup> from the target matrix. The use of molecular beams open new venues to extract refractory elements and it is an example in the quest to enlarge the palette of ISOLDE beams. For the nearest future a challenge is to produce a beam of the thorium isotope <sup>228</sup>Th. The beam-development programme is thus as vital as ever.

# 4. The CERN-MEDICIS project

The use of radioisotopes for medical applications, both for diagnostics and for therapy, has a long tradition at ISOLDE. The possibility to use the know-how on radioactive-beam production and handling at ISOLDE for medical applications in a more continuous and organised way has also been considered over the years. Different scenarios and collaborations were discussed but none of them grew into a proposal for a full-scale project. This changed in 2012 when a project under the working name CERN-MEDICIS (MEDical Isotopes Collected from ISOLDE) was launched with the aim to produce innovative radioactive isotopes for medical research at ISOLDE. To give some numbers the ISOLDE experiments use about half of the protons from the PS BOOSTER. The proton beam loses only about 10% of its intensity and energy when hitting a standard ISOLDE target. Thus the protons passing through the target can still be used. For CERN-MEDICIS, a second target will be placed behind the HRS target to produce dedicated radioisotopes. An automated conveyor, designed for this purpose, will carry this irradiated target to an isotope separator in the MEDICIS building [19]. There it will be attached to the front-end of the separator, heated and the isotopes of interest are separated and collected. In this way the longstanding experience and knowledge, the knowhow of production and separation acquired at ISOLDE over the years will be applied for the production radio-isotopes to be used in innovative medical applications.

In September 2013 a ground breaking ceremony marked the beginning of the construction of the CERN-MEDICIS building that is placed in an extension of the Class A laboratory. The construction work has been completed in 2016, as shown in figure 3. The first irradiations of the second target are foreseen to take place in 2017.

#### 5. The Era of post-acceleration

The first scientific conference entirely devoted to the new possibilities offered to nuclear physics with the ISOL technique was held in Lysekil, Sweden in 1967. At that time the ground-work for the ISOLDE experiment at CERN was well advanced and the future physics opportunities were discussed. Already at that conference the possible new physics that could be achieved with energetic beams of exotic nuclei was mentioned [20]. In the *History of CERN* the contribution by Hansen provided more information about different discussions and initiatives for post-acceleration [5]. In parallel with the successive building up of the palette of different experiments at the ISOLDE PS-Booster Facility, the ISOLDE Community initiated a dedicated project aiming at post-acceleration of the radioactive isotopes [21].



**Figure 4.** The ISOLDE beam line, equipped with the first HIE-ISOLDE cryomodule in its light grey cryostat. Photo taken in May 2015.

### 5.1. REX-ISOLDE

The basic principle of the proposed post-accelerator can be summarised as follows: (i) the radioactive ions produced in the ISOLDE target-ion-source are mass separated and collected in an ion-trap, (ii) after cooling and bunching in the trap the ions are brought into a charge-state breeder (REXEBIS) using the technique of electron-beam ionisation, (iii) the charge-bred ions are then fed into a room temperature linear accelerator. In the linear accelerator the ions are first accelerated with a radio-frequency quadrupole (RFQ), followed by an Inter-digital H-type (IH) structure and finally three 7-gap resonators. This project was approved as an experiment and the new accelerator, REX-ISOLDE [22], built in a collaboration between CERN and several of the institutes from the ISOLDE Community. The first REX-ISOLDE beams were accelerated October 31st 2001, when a beam of <sup>26</sup>Na ( $T_{1/2}$ = 1.07 s) was accelerated to 2 MeV u<sup>-1</sup>. The accelerated ions were delivered to the reaction target of a nuclear spectroscopy set-up, including charged particle detectors and one unit of the future MINIBALL  $\gamma$ -ray detector [23].

Adding a 9-gap acceleration stage behind the 7-gap section gave in 2004 the first energy upgrade of the REX-ISOLDE radioactive ion-beams. This allowed ions to reach a maximal energy of 3 MeV  $u^{-1}$  for ions with A/Q = 3.5. This new addition to REX-ISOLDE was first used to accelerate a beam of <sup>22</sup>Ne to 2.9 MeV  $u^{-1}$ .

The availability of post-accelerated beams opened up new fields of research within the ISOLDE programme. REX-ISOLDE, originally planned for accelerating light isotopes up to mass around  $A \sim 50$ , has gone far beyond this goal and has provided post-accelerated beams covering the entire mass range from <sup>6</sup>He up to <sup>224</sup>Ra for nuclear reaction studies. The experiments have mainly profited from the opportunities offered by Coulomb excitation studies in inverse kinematics. REX-ISOLDE has, in its 15 years of operation, delivered accelerated beams of more that 100 different isotopes from more than 30 chemical elements. A detailed account of technical details of REX-ISOLDE and its physics results is given in [24].



**Figure 5.** Gamma-ray spectrum of <sup>74</sup>Zn, Coulomb excited at two beam energies: 4 MeV u<sup>-1</sup> beam on <sup>208</sup>Pb target (blue line) and 2.85 MeV u<sup>-1</sup> beam on <sup>196</sup>Pt target (red line) illustrating the enhanced population of higher-lying 4<sup>+</sup> state at the increased beam energy. Notice the higher intensity of the 4<sup>+</sup> $\rightarrow$  2<sup>+</sup> transition at 4 MeV u<sup>-1</sup> as shown in the inset. Reproduced with permission from A Illana.

#### 5.2. HIE-ISOLDE

Even with the great success of the initial REX-ISOLDE experiments the idea of an additional energy upgrade was latent and present in the discussions within the ISOLDE Community and this went even so far as to produce a memorandum of understanding (MoU) between CERN and ISOLDE. But as time passed the energy upgrade idea was superseded by a dream of a more global upgrade, including increased energy, intensity and beam quality. This resulted in a project with the working name HIE-ISOLDE. HIE-ISOLDE would be a major staged upgrade of the existing ISOLDE facility [25, 26]. It includes an energy increase of the REX post-accelerator up to 5.5 MeV u<sup>-1</sup>, which is already operative, followed by a further increase up to 10 MeV u<sup>-1</sup>, as well as an upgrade of the REX resonators. The beam quality will be improved e.g., with the installation of a new RFQ cooler. The beam purification upgrade foresees also to include a new HRS and a multi-reflection time-of-flight (MR-ToF), for mass separation. As mentioned before, the beam intensity will be increased by two independent parts of the injectors. First the production will linearly increase with the enhanced proton intensity delivered by the new Linac-4 and second the fragmentation and spallation cross sections are enhanced by the increase in energy of the PS Booster to 2 GeV [27]. The HIE-ISOLDE project was approved on 2 September 2009.

With the typical CERN efficiency to realise approved projects the construction of the HIE-ISOLDE additions to REX progresses fast. Figure 4 shows one of the cryomodules during its installation in 2015. The first step of the energy upgrade of REX-ISOLDE was tested successfully already in September that year. The first scheduled physics experiment was then performed in October–November 2015 by accelerating beams of  $^{74,76}$ Zn<sup>28+</sup> to an energy of 4 MeV u<sup>-1</sup>. The aim of this experiment was to measure the *B*(*E*2) value for the  $4^+ \rightarrow 2^+$  transition of the ground-state band populated in Coulomb excitation. This allows to

investigate the interplay between collective and single-particle degrees of freedom. One of the first measured gamma energy spectra from HIE-ISOLDE is shown in figure 5.

While writing this contribution the full energy of stage one, with two cryomodules reaching an energy of  $5.5 \text{ MeV u}^{-1}$  and two new beamlines, have become fully operational. The success of the first stage of HIE-ISOLDE was celebrated on 28 September 2016.

In the near future two more upgrade stages will be launched. First, two additional cryomodules will be installed, together with the present two, to reach an energy of  $10 \text{ MeV u}^{-1}$ . At the same time a new beamline will be constructed and is foreseen to be operative in 2017. The presently used general-purpose scattering chamber (SEC) will be moved to this third beamline, while its present location will be used for the ISOLDE solenoid spectrometer already being commissioned at CERN.

The next and final stage of the energy upgrade involves the manufacturing and installation of two more cryomodules, each housing six low- $\beta$  cavities and two superconducting solenoids, giving to the full accelerator a total of six cryomodules. These last two accelerating elements will replace the present 7-gap and 9-gap resonators as well as the IH-structure (IHS) of the present REX and allow also for deceleration of the beam. This will provide access to a wider range of beams accelerated to lower energies, which will widen the experimental possibilities.

It is further planned to add a pre-buncher to the RFQ accelerator at a subharmonic frequency, which would allow increased bunch spacing without a major transmission loss.

A beam chopper will be added between the RFQ and the IHS to clean the background of satellite bunches. With all these additions the astrophysics program at ISOLDE will certainly be boosted.

For the future it is envisaged to build and install a compact heavy-ion low-energy ring to be able to perform experiments with stored, cooled secondary beams that can be used for studies spanning from nuclear ground-state properties to reaction studies of astrophysical relevance.

#### 6. The hunt for the limits of nuclear existence

The number of identified particle-stable isotopes and unbound nuclei with distinct quantum properties has increased tremendously over the past decades [28]. Experiments at ISOLDE have over the years certainly contributed to this with discoveries of many earlier unidentified isotopes from many elements. But the main conquest of the limits of existence has come with the advent of heavy-ion beam accelerators [29]. The strategy of isotope identification at ISOLDE relies, however, not only on proving its mere existence in a Z versus A/Z plot. The identification is always supported by measurements of, for example, half-life, mass, spin or electromagnetic moments.

A nice example is given by the discovery of the isotope <sup>229</sup>Rn. In a measurement at the ISOTRAP mass spectrometer the masses of the heavy radon isotopes, starting at mass 223, were determined out to mass 228. The next radon isotope had not been observed before. With a combination of a half-life measurement and a trap measurement the new isotope <sup>229</sup>Rn could firmly be identified with a half-life of  $12^{+1.2}_{-1.3}$  s and a mass precision  $\delta m/m = 6 \times 10^{-8}$  [30]. This isotope was announced in the newspaper *Welt (24.3.2009)* as isotope number 3176 discovered on the chart of nuclides.



Figure 6. The distributed beam-time at ISOLDE for experiments in 2016.

# 7. Applications: materials science and biophysics

The diversity, purity and high intensity of ISOLDE beams places experiments at the facility in a unique position to undertake innovative research that yields applications in the fields of material and life sciences. The Material Science programme is a prime example of how nuclear research and the associated technologies are not only of interest to scientists, but create substantial benefits for society at large.

Studies focus on semiconductors, oxides, and, more recently, nanoparticles and metals, while the main techniques of investigation are Mössbauer spectroscopy, perturbed angular correlation (PAC), emission channeling and tracer diffusion studies [31].

ISOLDE is specially adequate for Mössbauer spectroscopy due to the pure and intense <sup>57</sup>Mn beam that allows for recording thousands of Mössbauer spectra per day. Emission channelling allows the identification of the lattice location of radioactive atoms using measurements of the charged particles from the probe decay [32].

PAC experiments focus on materials research, as well as on biophysics [33]. One, for example, studies diffusion processes of different ions and structural changes due to doping in, for instance, thin film solar cells. As example of PAC experiments in biophysics one should mention the research on the function of metal ions in biological systems and synthetic biomolecules, the study of the local structure of antimony, which is used for the treatment of Leishmaniasis, and investigations of artificial DNA nanostructures.

The new VITO beamline (Versatile Ion Techniques Online) combines several experimental techniques requiring both spin-polarised and unpolarised nuclei and uses them to perform experiments relevant to material and life sciences, but also to nuclear physics and fundamental interactions. At VITO the spins are polarised using lasers and the versatile techniques to be used cover beta-decay asymmetry studies and beta-detected NMR (with a polarised beam), and online PAC (unpolarised beams), both in solid and liquid samples. Successful commissioning of the beam line with polarised <sup>26</sup>Na beam was achieved in the autumn of 2016 [34].

# 8. The distribution of the allocated beam-time

The experimental capabilities at ISOLDE make experiments in different branches of physics feasible as illustrated in figure 6. A major part of the experimental programme has over the



**Figure 7.** Left: transition probability into the  $\alpha + d$  continuum after  $\beta$  decay of <sup>6</sup>He. The red dots represent the data measured with the OTPC chamber [38], illustrating its superior performance at low energy. Right: decay scheme for <sup>11</sup>Be where the  $\beta^-p$  emission may be considered as a quasi-free neutron decay of the valence neutron [43]. Reproduced with permission from Ronja Thies, 2016.

years been devoted to nuclear physics experiments with a major emphasis towards the unique features encountered by the exotic nuclear species towards the dripline regions and, now in the post-accelerator era, even beyond. The long chains of isotopes available for many elements provide important sources of information about the evolution of nuclear structure. As an example ISOLDE can provide beams of the element Caesium (Z = 55) from mass 114–152. In total the nuclear physics experiments, including specialised experiments of relevance for weak-interaction studies, takes about three quarters of the scheduled beam-time at ISOLDE. The remaining beam-time is distributed between experiments in solid-state physics, atomic physics, biology, medicine and finally a fair time slot for the very important technical developments of new beams.

#### 9. Picking flowers in the nuclear landscape

In this section we shall give examples of some experimental results and highlights that according to our taste reflect the recent history of ISOLDE. We do this by taking a 'walk' over the chart of nuclides from <sup>6</sup>He to <sup>224</sup>Ra. On our way we shall pick up some examples of the beauty of the ISOLDE physics

The element helium (Z = 2).

A characteristic feature of nuclei approaching the dripline regions is the increase of Q values available for beta decay combined with decreasing separation energies for one or several particles. This permits the study of beta-delayed particle-emission processes for the most exotic nuclei on both sides of the beta-stability line. Several of the more rare beta-delayed decay modes were first observed at ISOLDE like  $\beta 2n$ ,  $\beta 3n$ ,  $\beta t$  and  $\beta \alpha$  [35]. As an addition to this group of exotic decays the beta-delayed deuteron emission was also observed for the first time at ISOLDE [36] in the decay of the lightest radioactive isotope produced at ISOLDE, <sup>6</sup>He ( $T_{1/2} = 807$  ms). The search for this decay mode was triggered by the proposed halo structure of <sup>6</sup>He [37]. In the first experiment the  $\alpha + d$  were separated by a set of magnets and detected in opposite directions by a channeltron and Si-detector. The determined branching ratio was a factor of hundred smaller than expected indicating large cancellation between the halo and core wave functions even at very large distances. In the following experimental attempts done at different facilities, the <sup>6</sup>He beam was either implanted on the

window of a gas-Si detector telescope or in a highly segmented Si detector to follow the decay. Nevertheless the deuteron energy spectrum could not be studied at energies below 0.5 MeV. In order to improve the low-energy data to test the different theoretical calculations, a recent experiment was set up utilising the optical time projection chamber (OTPC) technique for the charged particle detection [38]. The produced <sup>6</sup>He activity, accelerated in REX-ISOLDE to an energy of 3 MeV u<sup>-1</sup>, entered the OTPC through a window and was stopped in a gas mixture of atmospheric He/N<sub>2</sub>. The tracks from the charged particles following the decay were recorded with a CCD camera. This permitted the detection of alphas and deuterons of very low energy. Figure 7 shows the recorded  $\alpha + d$  events transformed to transition probability as a function of the  $\alpha + d$  energy, with data down to 150 keV. Since <sup>6</sup>He is the simplest Borromean two-neutron halo nucleus consisting of an  $\alpha$  particle surrounded by two neutrons, these kind of data are relevant for understanding the coupling to the continuum.

The element lithium (Z = 3).

The last bound lithium isotope, <sup>11</sup>Li ( $T_{1/2} = 8.2 \text{ ms}$ ) was the first isotope where a halo state in its ground-state was proposed and observed. Early experiments on spins, magnetic moments ( $\mu$ ) and electric quadrupole moments (Q) for <sup>9,11</sup>Li were important ingredients in the support of the halo suggestion [29]. The observation of similar electric quadrupole moments for <sup>9</sup>Li and <sup>11</sup>Li indicated an inert charged <sup>9</sup>Li core surrounded by a pair of neutrons in <sup>11</sup>Li. The laser spectroscopy work at ISOLDE has over the years been subject to successive refinements of the experimental technique. The  $\mu$  and Q measurements of <sup>9</sup>Li and <sup>11</sup>Li were revisited in experiments where the isotopes were implanted in Si and Zn lattices, respectively [39]. This resulted in a tenfold better precision of the ratio of the quadrupole moments,  $|Q(^{11}Li)/Q(^{9}Li)| = 1.088(15)$ , which opened up for more detailed theoretical work about the influence of the neutron halo on the <sup>9</sup>Li core in <sup>11</sup>Li.

The element beryllium (Z = 4).

Another major experimental success was achieved when the collinear laser technique was utilised to determine the nuclear charge radii for beryllium isotopes. With a beam of Be<sup>+</sup> ions, a frequency comb and measuring the absolute transition frequencies for parallel and antiparallel geometry of the ion and laser beams, a hitherto unreachable 1 MHz precision was obtained. The beauty of the technique is that the rest frame frequency,  $\nu_0$ , is obtained independent of the acceleration voltage by combining the measured absolute transition frequencies for parallel ( $\nu_p$ ) and antiparallel ( $\nu_a$ ) laser beams so that  $\nu_p \nu_a = \nu_0^2 \gamma^2 (1 + \beta)(1 - \beta) = \nu_0^2$ . The charge radii were observed to decrease for the isotopes <sup>7</sup>Be to <sup>10</sup>Be and then increase for <sup>11</sup>Be. This increase of the charge radius is expected since the centre of mass and the centre of charge do not coincide in the one-neutron halo nucleus <sup>11</sup>Be [40, 41].

The one-neutron halo nucleus <sup>11</sup>Be constitutes another example where a very rare decay has been identified. The neutron-separation energy in <sup>11</sup>Be is as low as  $S_n = 504$  keV, which means that  $\beta^-$ -delayed proton emission is energetically allowed, as illustrated in figure 7. The energy required for this process to occur is  $Q_{\beta p}(MeV) = 0.782$ - $S_n(MeV)$  [42]. With a proton spectrum extending up to about 280 keV only, it is very difficult to use conventional counting techniques to observe this decay. Further, theoretical estimates pointed to a branching ratio of the order of 10<sup>-8</sup>. An elegant method to get hold of the probability for this decay was to apply atomic mass spectroscopy (AMS). In a sample collected at mass position 11 one expects the presence of the long-lived delayed-proton emission daughter <sup>10</sup>Be. The amount of the isotope <sup>10</sup>Be in the collected sample, measured at the VERA facility in Vienna, was found to correspond to a branching ratio of (8.3 ± 0.9) × 10<sup>-6</sup>. The result of this experiment is convincing and at the same time very surprising [43] and may indicate the presence of a resonance in <sup>11</sup>B in the decay window. To understand the branching ratio, which is a factor of 100 higher than predicted, further theoretical work is needed. A plausible explanation may include a quasi-free neutron decay, that is a decay direct into the continuum. The relatively high branching ratio will allow a future study of the delayed-proton spectrum down to 100 keV using the already mentioned OTPC devise.

The element neon (Z = 10).

The masses and charge radii for <sup>17–22</sup>Ne [44] were measured in a joint venture including the groups specialised in Penning trap spectroscopy and in collinear laser spectroscopy. For <sup>17</sup>Ne, which is the last bound light neon isotope, this was at the time the lightest nuclide investigated at ISOLTRAP and also its first Penning trap mass determination. For the charge radius measurement the required very sensitive detection was obtained by using ion counting. The two-proton halo candidate <sup>17</sup>Ne was found to have a large charge radius. Calculations using the fermionic molecular dynamics model suggest that the large charge radius was due to an extended proton configuration in the ground state, with a 40%  $s^2$  component.

The element magnesium (Z = 12).

An interesting case of beta-delayed multi-particle decay has been observed for <sup>21</sup>Mg, produced in a SiC target. The beta-delayed particles were measured using a gas-Si telescope opposed to a Si(DSSSD)-Si telescope [45]. The presence of beta-delayed protons and alpha particles were detected and separated in the two-dimension  $\Delta E - E$  spectrum. Using a sophisticated statistical analysis the very rare decay mode  $\beta p \alpha$  could unambiguously be identified for the first time [46]. This prompted a general analysis of systematic trends in the  $\beta p$ ,  $\beta p \alpha$  and  $\beta \alpha$  emission processes. Future experiments on possible candidates for  $\beta p \alpha$ emission were found to be feasible, especially in  $T_z = -5/2$  nuclei like <sup>23</sup>Si and <sup>27</sup>S. The region of the nuclear chart around <sup>31</sup>Na (N = 20) is called the Island of Inversion

and has been subject to great interest since the early nineties. At ISOLDE this region may be entered by using a <sup>30</sup>Mg beam from REX-ISOLDE, produced in an UC<sub>x</sub> target and ionised with RILIS. This beam was accelerated in REX to  $2.25 \text{ MeV u}^{-1}$  and Coulomb excited in a Ni target inside the MINIBALL  $\gamma$ -ray spectrometer [47]. This was the first Coulomb excitation experiment using the full MINIBALL setup and was referred to as a 'safe' Coulomb experiment. Here safe means that the experiment is performed in an energy regime and angular range such that the main excitation process is electromagnetic with a negligible nuclear contribution. With this beam it is possible to enter the island of inversion region by populating states in  ${}^{32}Mg$  in (t, p) reactions. The data revealed an excited state at 1058 keV excitation energy and the angular distribution was compared to DWBA calculations. It was concluded that it was a  $0^+_2$  state and with a spherical shape as the <sup>30</sup>Mg ground state [48]. This means shape coexistence between spherical and deformed  $0^+$  states in  ${}^{32}Mg$ .

The element argon (Z = 18).

The most neutron deficient bound Ar isotope is <sup>31</sup>Ar ( $T_{1/2} = 15.1$  ms) with isospin projection  $T_z = -5/2$ . It is produced in a CaO target with a production rate of 1–2 atoms s<sup>-1</sup> only. With highly efficient  $\gamma$  ray detectors and Si telescope detectors, covering almost  $4\pi$  sr, the decay of this nucleus has been studied in greatest detail. By recording  $\beta p$ ,  $\beta 2p$ ,  $\beta 3p$  and beta-delayed  $\gamma$ 's a very complete decay scheme could be constructed. The spectrum of the beta-delayed particles were used to reveal how the different decay modes evolve with increasing excitation energy in  $^{31}$ Cl. In particular it was shown that 3p events were a very sensitive probe of the high-energy part of the Gamow-Teller strength and that as much as 30% of the total strength resides in the  $\beta 3p$  decay mode [49].

The element calcium (Z = 20). The neutron-rich isotope <sup>52</sup>Ca, has been predicted as a doubly magic nucleus with N = 32 as a new magic number far from stability. Its magicity has been tested in nuclear

spectroscopy experiments as well as in direct mass measurements. Recent experiments at TITAN/TRIUMF gave new results for the masses of the two isotopes <sup>51,52</sup>Ca, results which showed the need for going further away form stability to pin down the theory. In order to go beyond <sup>52</sup>Ca a new device was added to the three ion traps that constitute the ISOLTRAP spectrometer. This addition was a newly developed MR-ToF mass spectrometer [50], increasing the effective time-of-flight path to several kilometers in a time of about 10 ms. This gives access to nuclides with low production rate and half-lives in that time domain. With this the masses of <sup>53,54</sup>Ca could be determined [51], and the results showed that pronounced structural effects are in play also for very exotic nuclei and that shell effects remain. The results provide key information for Chiral effective field theory and show that a description of extreme neutron-rich nuclei can be closely connected to a deeper understanding of nuclear forces.

#### The element zinc (Z = 30).

The Penning trap mass spectrometer ISOLTRAP was used to measure masses of zinc isotopes in the mass range  $^{71m,72-81}$ Zn, thus including masses on both sides of the N = 50 waiting point nucleus,  $^{80}$ Zn [52], where the astrophysical *r*-process responsible for the synthesis of the heavy elements is slowed down. In the measurements the Zn ions were separated in the HRS and brought into and bunched in a He-gas-filled RFQ, then transferred into a Penning trap for isobaric purification and finally injected into a precision trap, where the cyclotron frequency was measured. This nucleus was the first of the few major waiting points along the rapid neutron-capture path where both the neutron-separation energy and the capture *Q*-value were experimentally determined. In a second attempt, to reach heavier masses the full ISOLDE machinery was put into place to deliver pure beams: neutron converter, temperature-controlled quartz line and lasers, nevertheless the A = 82 beam contained over a factor of 600 more <sup>82</sup>Rb than the nucleus of interest <sup>82</sup>Zn. The addition of a new device to the ISOLTRAP system the MR-ToF allowed one to separate of the isobars and the determination for first time of the the mass of <sup>82</sup>Zn [53]. The new data were incorporated into model calculations that enabled to determine the crust composition of a neutron star.

The element gallium (Z = 31).

The element Ga has been studied over a broad range of isotopes with collinear laser techniques. In a first experiment [54] nuclear spins and moments were determined from N = 36-50. The data show anomalous ground state spins for <sup>73,81</sup>Ga (I = 1/2 and 5/2, respectively) and a sign change in the quadrupole moments for <sup>75,77,79</sup>Ga, which suggest a changing shell structure at N = 42. In more recent studies the mean-square charge radii were extracted from isotope shift data [55] in the region N = 32-51. These results form together a very complete systematic study where the N = 50 shows a shell effect while the N = 32 did not show any anomaly.

#### The element rubidium (Z = 37).

To the group classified as fundamental-interaction experiments belong the study of superallowed  $0^+ \rightarrow 0^+ \beta$  decays. These decays offer stringent tests of CVC when very precise values of half-life, beta-decay Q value and branching ratio are provided. Examples of such input data are the precise ISOLTRAP mass values that have been obtained for the isotope <sup>74</sup>Rb [56] and its beta-decay daughter <sup>74</sup>Kr [57]. These data are part of the 20 cases included in the 2014 critical survey by Hardy and Towner [58]. For the CKM matrix this survey proposes an up–down quark mixing matrix element of  $V_{ud} = 0.97417(21)$  and an unitary test of the upper row matrix elements give  $|V_{ud}|^2 + |V_{ud}|^2 = 0.99978(55)$ .

The element cadmium (Z = 48).

The region around the doubly magic pair, Z = 50 and N = 82, attracts considerable interest. ISOLDE is able to attack the problem with all existing tools: measurements of



**Figure 8.** Left: background-subtracted Doppler-corrected  $\gamma$ -spectrum for Coulomb excitation of <sup>126</sup>Cd (2<sup>+</sup><sub>1</sub>  $\rightarrow$  0<sup>+</sup><sub>g.s.</sub>, 652 keV) in a <sup>64</sup>Zn target. Right: 1  $\sigma$  region for the diagonal  $M_{22} = \sqrt{\frac{7}{2\pi}} \frac{5}{4} e Q_s(2^+_1)$  and non-diagonal  $M_{20} = \sqrt{B(E2; 0^+_{g.s.} \rightarrow 2^+_1)}$  matrix elements for the 2<sup>+</sup><sub>1</sub> state in <sup>126</sup>Cd obtained in a maximum likelihood analysis [59].

ground states properties: masses, spins, electromagnetic moments, beta decay studies and reactions.

The observation of an anomalous behaviour of the  $2_1^+$  state with a decrease of its energy passing from <sup>126</sup>Cd to <sup>128</sup>Cd prompted a Coulomb excitation experiment at REX ISOLDE with 2.85 MeV/u beams of <sup>122,124,126</sup>Cd. The reaction targets were selected so that the Coulomb excitation was 'safe', which means that the nuclei involved should be kept at a distance such that the minimum impact parameter should be  $b(\theta) \ge R_1 + R_2 + 5$  fm. The reduced transition probabilities  $B(E2; 0_{g.s.}^+ \rightarrow 2_1^+)$  and limits for the quadrupole moments for the triplet of even isotopes <sup>122,124,126</sup>Cd were determined [59], see figure 8. The results were discussed in terms of onset of collectivity in this region.

The spin and electromagnetic moments of  $^{107-129}$ Cd isotopes reaching N = 82 shell closure have been measured recently [60]. Deep ultraviolet excitation and radioactive beam bunching provided the required sensitivity, even allowing for the discovery of long lived isomers in  $^{127,129}$ Cd. The quadrupole moments on isotopes with spin 11/2 related to the  $h_{11/2}$ orbital show a lineal increase with the neutron occupation as expected in an extreme shell model description. The linear trend extends beyond the filling of this orbit indicating a certain degeneracy with the neighbouring orbitals. It is amazing to find such a simplicity in these complex systems. In addition, the difference in radii between the ground state and the isomeric one shows an unusual simple parabolic behaviour when moving from a slightly oblate to a prolate shape [61].

Measurements of the masses for the three isotopes  $^{129-131}$ Cd were performed at the ISOLTRAP spectrometer [62]. The most tricky case here is  $^{131}$ Cd due to low efficiency and short half-life ( $T_{1/2} = 68$  ms) but the mass could be determined by using the faster MR-ToF technique. The results show deviations of more than 400 keV as compared to earlier beta-decay data. These data give evidence of a significant reduction in the N = 82 shell gap for Z < 50 and, maybe most interesting, give more reliable description of the *r* process nucleosynthesis with these higher-precision data. As the authors of [62] write "...Given the large number of data required in *r*-process calculations, it is remarkable that only three masses make an observable impact on the predicted abundances...'. This shows the importance of these kind of data at the 'precision frontier''.



**Figure 9.** Left: systematics of mean-square radii  $\delta < r^2 >$  for Hg isotopes measured at ISOLDE. The red dots represent measurements from 2015 [70]. Right: low-energy part of the <sup>182</sup>Hg level scheme. The  $0_1^+$  ground state is found to be weakly deformed and predominantly of oblate nature while the  $0_2^+$  at an energy of 335 keV is more deformed.

The beta decay of the *r*-process waiting point nucleus <sup>132</sup>Cd was recently measured at the ISOLDE decay station (IDS) [63]. The beta decay strength distribution of waiting point nuclei is extremely valuable to anchor nuclear models necessary to calculate half-lives farther away from stability. Previous measurements of the decay of <sup>132</sup>Cd at ISOLDE [64] and RIKEN [65] did not observe gamma-rays, indicating a neutron branching ratio close to 100%. Due to this expected large neutron branching ratio, the neutron time-of-flight detector VANDLE [66] was installed at IDS. The largest neutron transition was identified to be neutron emission from the first 1<sup>+</sup> state in <sup>131</sup>In. This single beta transition, with log(ft) = 4.3, dominates the decay strength and the half-life of <sup>132</sup>Cd, which is 97 ms.

The element hafnium (Z = 76).

The element hafnium is produced in a Ta/W metal foil target. The produced radioactive Hf isotopes are brought to the ion source with CF<sub>4</sub> as supporting gas. The Hf isotopes are then delivered to the experiments in form of a HfF<sub>3</sub><sup>+</sup> beam. With the NICOLE online nuclear orientation dilution refrigerator the  $\gamma$  decay of the isomeric <sup>180</sup>Hf<sup>m</sup> state was studied. The results for the 501 keV 8<sup>-</sup> $\rightarrow$  6<sup>-</sup> transition gave an *E2/M2* mixing ratio confirming the presence of parity violation in this decay [67].

The element mercury (Z = 80).

One very early success in the ISOLDE history is the RADOP experiments on Hg isotopes that revealed the famous odd–even staggering of the isotope shifts and deduced radii [68]. Figure 9 shows the present status of the derived  $\delta < r^2$ > where the 1972 results still stands out as unique. The even mercury isotopes <sup>182–188</sup>Hg accelerated to 2.85 MeV u<sup>-1</sup> in REX-ISOLDE were studied via Coulomb excitation in targets of Sn, Ag and Cd [69]. The magnitudes and relative signs of *E*2 matrix elements that couple ground states and low lying states were determined. It was found that the ground states of <sup>182,184</sup>Hg are slightly deformed and oblate while the the second 0<sup>+</sup> state has a larger deformation. The data were interpreted within a two-state mixing model and the presence of two different co-existing structures in the light even-mass Hg could be established firmly.

The element astatine (Z = 85).

Astatine is the least abundant chemical element found on the Earth's crust. The minute quantity of radioactive astatine of 0.07 g that exists in nature originates in the decay chains



**Figure 10.** Laser scan across the ionisation threshold of At, the observed Rydberg resonances converge towards the ionisation limit. The indicated principal quantum numbers (n) are based on the assumption of an *nd* series.

from <sup>238</sup>U. To study its basic chemical and physical properties, artificial production is required. In the quest to develop new ionisation schemes for the users, in this case At, the RILIS team realised that the atomic properties, and in particular the ionisation potential, were not known. The At isotopes were produced in a UC<sub>x</sub> target and in-source laser spectroscopy was performed using the RILIS ion source. The observed high-lying Rydberg states, as shown in figure 10, permitted the determination the ionisation potential for the first time [71]. The developed ionisation schemes then enabled the first in-source laser spectroscopy study of the astatine isotopic chain at ISOLDE. In addition beta-delayed fission of <sup>194,196</sup>At [72] was measured. The fission-fragment mass distribution in the Po daughter nuclei indicated a triplehumped structure making the transition between the asymmetric fission found in Hg and the symmetric one in the light Ra–Rn nuclei.

The element radium (Z = 88).

A radioactive beam of the isotope  $^{224}$ Ra ( $T_{1/2} = 3.66$  d) accelerated in REX-ISOLDE is the hitherto heaviest post-accelerated beam. In a 52<sup>+</sup> charge state from REX-EBIS its was accelerated to 2.83 MeV u<sup>-1</sup> and studied in a Coulomb excitation experiment with MINI-BALL (see figure 11) using targets of  $^{60}$ Ni and  $^{120}$ Sn. In a paper presenting the results for  $^{224}$ Ra together with those for  $^{220}$ Rn the deduced *E*3 transition strengths were used to conclude that  $^{224}$ Ra has a permanent octupole deformation in its ground state [73]. This is important for the search of atomic electric-dipole moments (EDM), since a non-zero EDM would indicate *T* or CP invariance due to physics beyond the standard model. These 'drops of nuclear matter' do not have the same centre of mass and centre of charge which favours the search for static EDM and other new physics [74].

# 10. Final remarks

While writing this chapter we have witnessed how nature turned from green to different nuances of yellow, orange and red. The leaves start to fall from the trees and cover the ground. At ISOLDE the next to final beam-time before the annual shut down is devoted to a HIE-ISOLDE run using a 6.8 MeV  $u^{-1.9}Li^{3+}$  beam. The aim of this experiment is to study the <sup>11</sup>Li binary subsystem <sup>10</sup>Li in a (*d*, *p*) reaction. We are looking forward to a spring with a brand new set of experiments. A bit more into the future we are looking forward to the completion of the HIE-ISOLDE project both in the energy and intensity part as well as to the



**Figure 11.** The MINIBALL gamma detector array at ISOLDE. The inset shows a graphic representation of the octupole shape of  $^{224}$ Ra with the deformation deduced parameters.

future installation of a compact heavy-ion low energy ring. With all these upgrades ISOLDE will remain in the coming years at the forefront of nuclear research. We hope that this chapter will give inspiration for further reading of the following more in-depth chapters of this ISOLDE focus issue.

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