Fast-timing spectroscopy at ISOLDE

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Abstract

The advanced time-delayed $\beta\gamma\gamma(t)$ method has been used at ISOLDE since the relocation of the facility from the synchrocyclotron to the CERN proton synchrotron booster, a quarter of a century ago. The method was designed for precision measurements with low-intensity beams, achieving good efficiency and excellent time resolution with a compact setup. Over this time the technique has evolved to cope with the challenge of measuring lifetimes of complex level schemes of increasingly exotic nuclei populated in $\beta$ decay. The ISOLDE facility provides unsurpassed opportunities to study many regions of the nuclide chart. The physics case encompasses topics of interest across the nuclide chart, including the evolution of shell structure around neutron shell and subshell closures such as $N = 20$, $N = 40$ and $N = 50$, shape coexistence, and octupole correlations in heavy nuclei. The recently commissioned ISOLDE decay station provides enhanced capabilities that will be fully exploited with the increased beam intensities available at the upcoming HIE-ISOLDE facility.

Keywords: ISOLDE, advanced time delayed $\beta\gamma\gamma(t)$ method, level lifetimes, reduced transition probabilities, shell closures, islands of inversion, octupole correlations

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

The atomic nucleus is an excellent laboratory to test quantum physics in action in a many-body system made up of fermions of two flavours, protons and neutrons. Apart from the fermionic degrees of freedom, strong correlations rule the low energy phenomena that happen in nuclei. In spite of the steady advancement of nuclear models, a consistent description of nuclei across the nuclear chart based on first principles is still lacking. *Ab initio* nuclear models to date are constrained to light nuclei and to simple nuclei around closed shells. Large-scale shell-model calculations [1] have been successful in describing many of the nuclear properties, not only close to stability but also for exotic species. In this framework the main ingredient is the single-particle energy levels, which can group in given magic numbers of protons and neutrons (2, 8, 20, 28, 50, 82 and 126) to form the shell structure of the nucleus. The single particle motion in the average potential of the nucleons is able to account for shell effects, but the residual interaction beyond the mean field and correlations are advocated to explain collective phenomena.

Since decades it is known that severe modifications to the standard single particle energies exist in regions of the nuclear chart far from the stability line. The magic numbers for nuclides with large asymmetry in protons and neutrons do not longer match those known at stability, and new shell gaps develop. Experimental and theoretical nuclear structure physics is devoted to the understanding of the new structures and the reasons behind them. The understanding of these structure modifications will be a step forward in the quest for nuclear models valid across the nuclear chart and with strong predictive power.

Experimentally the mapping of the changes in nuclear structure relies on observables that do not depend on modelling, and that need to be systematically obtained in vast regions of the nuclear chart, specifically for neutron-rich nuclei. The basic spectroscopic information given by excitation energies, typically obtained from γ-ray measurements, is one of the first observables accessible after the identification of nuclei in the laboratory. More stringent benchmarking of nuclear theory can be achieved by the knowledge of matrix elements for transitions between excited nuclear levels.

These absolute transition matrix elements can be obtained by the direct measurement of the lifetimes of nuclear levels. Therefore, several experimental methods for the measurement of nuclear lifetimes, which can be tailored to the production mechanism and to the magnitude of the nuclear lifetime, are put into play. Electronic timing methods, such as the advanced time-delayed (ATD) $\beta\gamma(t)$ method or Fast Timing is very well suited for the measurement of lifetimes of excited levels ranging from a few ps to several ns in nuclei populated in β-decay. The technique has been successfully exploited at ISOLDE.

This article will start by stressing the impact of transition rates in our understanding of nuclear structure. The ATD $\beta\gamma(t)$ method and the latest technical improvements will be then reviewed. The physics programme at ISOLDE will be described by highlighting recent results in several areas of the nuclear chart.

2. Nuclear transition rates

Electromagnetic transition probabilities between nuclear states furnish unique structure information. For instance, transition rates can provide an indication of the ground state deformation of even–even nuclei from the quadrupole moment extracted from the reduced $B(E2)$ probability of the transition from the first excited $2^+$ to the $0^+$ ground state. Similarly collective quadrupole and octupole modes may be identified from the magnitude and
dependence of transition rates. Shape coexistence in nuclei is revealed by the transition probabilities as a function of the wave function content of nuclear states. Many other examples could be listed.

As it is well known, the interaction of the electromagnetic field with the nucleons can be expanded into an infinite series of electric and magnetic terms \[2\], out of which only the lowest moments (dipole, quadrupole and hexadecapole) provide significant contributions. For a given magnetic or electric \((X = M \text{ or } E)\) operator \(\mathcal{M}\) of a defined multipolarity \(\lambda\), that involves the transition from an initial \(|\psi_i\rangle = |\alpha J M_i\rangle\) to a final state \(|\psi_f\rangle = |\beta J M_f\rangle\), the transition amplitudes are proportional to the matrix elements. The Wigner–Eckart theorem can be used to express the matrix elements as a function of the reduced matrix element, \(|\langle \psi_f | \mathcal{M}(X\lambda) | \psi_i \rangle|\), which does not depend on the third component of the nuclear spin (the magnetic quantum number). To measure the total probability the reduced transition probability \(B(X\lambda)\) is defined as

\[
B(X\lambda; J_i \rightarrow J_f) = (2I_i + 1)^{-1} |\langle \psi_f | \mathcal{M}(X\lambda) | \psi_i \rangle|^2.
\]

It is obtained by averaging over the initial magnetic substates \(M_i\) and summing over the final state substates \(M_f\) and the third component \(\mu\) of the electromagnetic multipole \(\lambda\). The reduced transition probability has a main benefit compared to the reduced matrix element, namely that it can be directly measured experimentally and does not require the knowledge of the spin of the initial state. In fact the simple relationship below holds:

\[
B(X\lambda; J_i \rightarrow J_f) = \frac{2I_f + 1}{2I_i + 1} B(X\lambda; J_f \rightarrow J_i).
\]

Since the mathematical formalism of the emitted electromagnetic radiation caused by the oscillating electric and magnetic moments is well known, it is feasible to write the total transition rate for an electric or magnetic \((X = E \text{ or } M)\) transition of multipolarity \(\lambda\) from a nuclear state with spin \(J_i\) to a final nuclear state \(J_f\) as a function of the reduced transition probability as \[2\]:

\[
P_e(X\lambda; J_i \rightarrow J_f) = \frac{8\pi(\lambda + 1)}{\lambda!(2\lambda + 1)!} \frac{E_{\gamma}}{hc} \left(\frac{E_{\gamma}}{\hbar c}\right)^{2\lambda+1} B(X\lambda; J_i \rightarrow J_f).
\]

Therefore, for a defined magnetic or electric operator of a given multipolarity \(\lambda\) the measurement of the partial mean life \(\tau_\gamma\)—which is the inverse of the rate \(P_e\)—provides a direct and model independent access to the reduced transition probability via the relation

\[
B(X\lambda; J_i \rightarrow J_f) = \frac{\lambda!(2\lambda + 1)!}{8\pi(\lambda + 1)} \left(\frac{\hbar c}{E_{\gamma}}\right)^{2\lambda+1} \tau_\gamma^{-1}.
\]

Thus, the measurement of the mean life of excited nuclear state, \(\tau\), together with the gamma branching ratio, \(b_\gamma\), provides the partial mean life \(\tau_\gamma = \frac{\tau}{b_\gamma}\), from which electromagnetic transition rates are directly obtained. In addition to other transitions de-exciting the nuclear state, the internal conversion branches have to be taken into account to obtain \(b_\gamma\).

The \(B(X\lambda)\) values for the most probable transitions multipoles are compiled in table 1 as a function of energy and \(\tau\). In most cases the electromagnetic radiation has mixed contribution from only the two lowest multipoles. The mixing ratio, for example \(\delta(M1/E2)\), can often be determined in other experiments.

Reduced transition probabilities allow for comparison of rates for different transitions of the same type within the same nucleus. Single-particle Weisskopf estimates provide a (model-dependent) method to intercompare transition rates between different nuclei. When
expressing transition rates in Weisskopf units one obtains an indication of enhancement of a transition rate caused by collective nuclear effects, for example due to the quadrupole or octupole degrees of freedom. The Weisskopf estimates as a function of nuclear mass number \( A \) are compiled in table 1.

### 3. The advanced time delayed \( \beta\gamma\gamma(t) \) method—technical details

The experimentally accessible range of nuclear lifetimes spans forty-five orders of magnitude. Delayed coincidence methods using for instance HPGe detectors are employed to measure in the microsecond and nanosecond range [3], while the femtosecond and picosecond range is accessed by the recoil distance Doppler method [4] and the Doppler shift attenuation method [5]. The ATD \( \beta\gamma\gamma(t) \) method is the best suited to measure half-lives between a few picoseconds and several nanoseconds in \( \beta \)-decay.

The ATD method was invented by Henryk Mach in 1986 in the TRISTAN facility located at Brookhaven National Laboratory, and developed and published by Mach et al [6–8]. As other delayed coincidence methods, it is based on the time distribution between the population of an excited state and its de-excitation. It relies on triple coincidences between a \( \beta \)-particle and two \( \gamma \)-rays. A thin and very fast plastic detector is used to measure \( \beta \) particles following the \( \beta \)-decay of the parent nucleus, while a scintillator crystal with good timing resolution is used for the de-exciting \( \gamma \)-rays. The time distribution between them gives the half life of the excited level. The second \( \gamma \)-ray is detected by a HPGe detector, characterised by good energy resolution, to select a specific gamma cascade.

The ability to measure short lifetimes is defined directly proportional to the time resolution and inversely proportional to the statistics. Thus, the optimisation of the scintillator detector time resolution and the enhancement of the statistics are required to maximise the capability of measuring short lifetimes. When the level lifetime is long enough it will show as a slope on the delayed part of the time distribution. The exponential decay and the prompt time distribution, which can be approximated to a Gaussian function, can be deconvoluted from the time spectrum. In the case of shorter half-lives, down to 5–10 ps depending on the time resolution and the amount of statistics, the centroid shift technique is used. The centroid of the time distribution from a gamma de-exciting a level is shifted by the mean life of the level compared to a gamma of the same energy de-exciting a prompt level. This technique relies on precise calibrations of the timing detector for the \( \beta \), Compton and full-energy peak events, since each of them will give rise to different time response in the scintillators, as outlined below.
In order to optimise fast-timing measurements at ISOLDE, research and development on instrumentation have been done in several areas. Much of this work is based on the previous experience of the fast-timing collaboration, gathered over years of expertise.

3.1. Scintillation crystals

The Ultra Fast Timing method relies on the use of fast scintillation crystals coupled to fast photosensors for the detection of gamma radiation. When the method was established a new fast scintillator, BaF$_2$, had just become commercially available. The germanium-gated coincident method and the calibration procedures were developed based on the performance of the then new BaF$_2$ crystals, very well suited for this purpose. Breakthrough experiments were carried out at the TRISTAN on-line mass separator facility at Brookhaven National Laboratory using very small BaF$_2$ crystals in the shape of truncated cones with diameter of 25 mm at the base and 19 mm at the top, and height of 13 mm.

For the first series of experiments at the ISOLDE facility in the PS-Booster we used similar truncated cone BaF$_2$ crystals, but with height of 25 mm. The same type of scintillators had been tested at the ISOLDE-SC and were used for similar experiments at OSIRIS in Studsvik (Sweden), and afterwards at the IGISOL facility in Jyväskylä. At a later stage a new generation of crystals with customised shape and size was developed by Henryk Mach and produced by Scionix. The idea was to improve the detection efficiency for $\gamma$-rays up to 3 MeV by increasing the size, while maintaining the excellent time resolution. These detectors of the ‘Studsvik design’ consisted of a hybrid configuration, with a conical section and a cylindrical one. The cylindrical part had a diameter in the base of 40 mm, with a height of 17 mm (or 7 mm, depending on the design) and then a conical section with a base of 20 mm diameter at the entrance window. Longer crystals were not considered due to the deterioration of the crystal quality (self-absorption, internal scattering of light and internal activity) arising from larger crystal sizes.

The two main challenges for the production of such crystals are the fabrication of the complex geometries and the treatment of the surfaces. The best performing crystals were selected with a yield of only about 20%. The energy resolution for the 661.7 keV $^{137}$Cs line is 9.0% for the best crystals and about 10.0% for most of them. In spite of the steady development of inorganic scintillators for $\gamma$ detection, BaF$_2$ is still the fastest commercially produced crystal with time resolution of $\sim$80 ps FWHM for small $\sim$1–2 cm$^3$ crystals and 150 ps FWHM for crystals of our Studsvik design at $^{60}$Co energies.

3.1.1. LaBr$_3$(Ce) crystals. In more recent times a new generation of inorganic scintillators has recast gamma-spectroscopy. They are mostly halide compounds, with a large effective atomic number, and therefore good stopping power, and they combine excellent timing properties with good energy resolution. The most prominent example is LaBr$_3$(Ce), commercially available for a decade. These crystals are characterised by a high photon yield of 63 000 photons MeV$^{-1}$, relatively high density of 5.3 g cm$^{-3}$ and a very sharp rise time (see figure 1).

LaBr$_3$(Ce) have a much better energy resolution than BaF$_2$ crystals of the same size, $\sim$3% FWHM at 662 keV, while maintaining an excellent time resolution, which is only 10% worse in comparison to BaF$_2$ crystals. It has been reported that the time resolution of LaBr$_3$(Ce) crystals depends on the Ce doping, improving with higher content of cerium. Standard crystals currently available have 5% Ce doping. The enhanced energy resolution of LaBr$_3$(Ce) entails a big advantage in fast timing measurements such as those performed at ISOLDE, when complex decay schemes, with a large number of transitions and with
overlapping energies, need to be disentangled. This is the case for decays of exotic nuclei with large beta decay $Q$-values, and for heavier nuclei in regions with large quadrupole deformation and with octupole collectivity. It is also specifically important to unravel level schemes of odd-nuclei. In addition, a better energy resolution provides a better ratio between the full-energy peak and the Compton continuum underneath, and thus results in smaller time corrections due to Compton background under the full-energy peaks [8].

The LaBr$_3$(Ce) crystals are commercially produced and encapsulated by Saint Gobain with a nominal Ce doping concentration of 5%. The housing has a thin aluminium window at the entrance, and is fitted with a glass light guide at the coupling side to the photosensor. Several inner layers of light reflector and shock absorbing material protect the crystal, assuring its stability and minimising photon losses [10]. In our experiments at ISOLDE the crystals are shielded as much as possible against Compton-scattered $\gamma$-rays from surrounding detectors, mainly from the HPGe detectors. As shielding we use Pb sheets of a few mm thickness, occasionally aided by 1 mm copper sheet next to the crystal.

The time resolution given as the full width at half maximum (FWHM) for an individual cylindrical crystal 1 inch in height and 1 inch in diameter has been measured by our collaboration to be below 100 ps [12] for $^{60}$Co peak-to-peak coincidences. This translates in a coincidence resolution time for a pair of similar crystals at the same energies of the order of 140 ps, thus providing very competitive results compared to BaF$_2$. In the experiments at ISOLDE we have employed two types of LaBr$_3$(Ce) cylindrical crystals, with the diameter and height of 1 in. $\times$ 1 in., and with dimensions 1.5 in. $\times$ 1.5 in. Other sizes and shapes are used in other facilities, and by the FATIMA collaboration for larger arrays [13, 14].

For the fast timing measurements performed over the last years at ISOLDE we have designed special geometries of LaBr$_3$(Ce) crystals aimed at enhancing the light collection and with the expectation of an improved time response. These new geometries make it possible to enhance the packing factor around the implantation point compared to cylindrical geometries, increasing the $\gamma$-ray detection efficiency. The first design is a truncated cone shape crystal, with 1 in. height and bases of 1.5 in. and 1 in. We use this type of crystals as the standard at ISOLDE, including the new set up at the ISOLDE decay station (IDS). A second design (shown in figure 1) is based on the ‘Studsvik’ design for BaF$_2$ crystals described above. It is a
tapered hybrid crystal with a cylindrical section of 1.5 in. in the base and 0.65 in. in height and, a 1.2 in. long conical section (i.e. with a total height of 1.85 in) with 0.75 in. in the small base. According to the experience with BaF$_2$, the conical shape should give about 20\% improvement in time resolution when compared to the cylindrical shape. However, the improvement depends on the details of the shape and crystal quality, which tends to be rather inhomogeneous. We have recently characterised the time and energy responses of the specially shaped crystals \cite{15}, and the conical shapes provide an excellent compromise between good time response (110 ps FWHM at $^{60}$Co energies), good energy resolution and enhanced geometrical efficiency for positioning the detectors in close geometry around the ISOLDE collection point.

3.1.2. CeBr$_3$ crystals. We have investigated several alternatives to LaBr$_3$(Ce). The recently developed CeBr$_3$ is a good candidate due to its fast rise time, a decay constant of 17 ns and high photon yield of about 68 000 photons MeV$^{-1}$ \cite{16}, a good energy resolution, of the order of 4\% at 662 keV has also been reported. A further advantage of this crystal over BaF$_2$ and LaBr$_3$(Ce) is that it does not possess internal activity. Moreover a competitive market price can be expected.

We have performed a detailed study of this type of crystal to assess whether it is a viable alternative to LaBr$_3$(Ce) \cite{9}. The time response of a CeBr$_3$ crystal of 1 in. in diameter and 1 in. in height, commercially available from Scionix, with a 2-inch fast-response R9779 Hamamatsu phototube is below 120 ps at $^{60}$Co energies, with a good energy resolution of 4.3\% at 662 keV. Nevertheless the energy (and also timing) performance of LaBr$_3$(Ce) is still superior to CeBr$_3$, and in addition special shapes and sizes have not been commercially available until very recently. For the experiments at ISOLDE we stick to LaBr$_3$(Ce) crystals as the basis for fast scintillator detectors.

3.1.3. Beta detector. A thin NE111A plastic scintillator, or the current commercial equivalent EJ-232, is used for the $\beta$ detector. The main purpose is giving a time response largely independent on the energies of the detected $\beta$-rays. The residual time dependence on the $\beta$-ray energy is determined and corrected for in the off-line analysis. A standard thickness is 3 mm since it provides a compromise between the requirement of a strong signal for good timing response and the need for the scintillator to be as thin and small as possible in order to minimise the $\gamma$ sensitivity. The ratio of coincident events when a $\gamma$-ray is recorded in the plastic scintillator and a $\beta$-ray deposits energy in the $\gamma$ scintillator is of the order of a percent compared to good $\beta$--$\gamma$ coincidences. The sensitivity of the $\beta$ detector to $\gamma$-rays of higher energies is much smaller. The time resolution of the $\beta$ detector is of the order of 50 ps FWHM \cite{17} and has a minor contribution to the time resolution of the detection system.

3.2. Choice of photosensors

In spite of recent advances of compact photosensors such as Silicon photomultipliers \cite{18}, the photodetectors of choice to couple fast scintillators of relatively medium volume are still photomultiplier tubes (PMTs).

The BaF$_2$ crystals were attached to Photonis XP2020URQ PMTs. They are 12-stage tubes equipped with a quartz window, and they were operated at 2100–2400 V, well below the design high voltage due to the excessive gain of these tubes for BaF$_2$ crystals. Fast 2-inch PMTs with lower number of dynodes only came to the market after 2005. Crystals are coupled to the PMTs using Viscasil silicon grease and wrapped into opaque tape in order to
avoid photon losses and maintain the detector isolated from light. The XP2020URQ photocathode is very fast, with a rise time of 1.4 ns and transit time spread (TTS) of 350 ps FWHM. For our measurements at ISOLDE only XP2020URQ PMTs with the highest sensitivity and the lowest noise were selected.

In the case of LaBr₃(Ce) crystals, with much higher photon yield, the matching photocathode designed by Photonis was the XP20D0, with 8 dynodes. Contrary to XP2020UR URQ PMTs, this model uses a simplified front end and was equipped with a double anode to optimise the time resolution. The XP20D0 has a rise time of 1.6 ns and a TTS of 520 ps FWHM [19], which are worse parameters than those for the XP2020UR tubes. The XP20D0 were operated at voltage just below 1000 V. Due to the short duration of the light flash in the crystal and the high photon yield, space charge effects cause energy nonlinearity, which can amount to 15% at this voltage. Photonis also started the development of 6-stage XP20E0 phototubes, with the expectation of better performance than the XP20D0 PMT for high-yield scintillators. However, in 2009 Photonis stopped the production of PMTs.

The Hamamatsu R9779 PMT is the 2-inch PMT designed by Hamamatsu for LaBr₃(Ce) crystals. This 8-stage tube has an accelerator ring at the front end to give similar timing characteristics if not slightly better than XP20D0, with an anode pulse rise time of 1.8 ns and a TTS of 250 ps. For ISOLDE experiments we use the R9779 in the base assembly model H10570 MOD, including modifications in the signal outputs, the length of the PMT and the voltage division tapering. Earlier tests with a NE111A plastic show that a FWHM time resolution of the order of 50 ps can be achieved [17], which is better than the value that can be obtained with the XP20D0 PMT, of the order of 75 ps. It should be also noted that the coupling of R9779 to LaBr₃(Ce) shows excellent performance in terms of time response and good linearity [20], compared to the nonlinear energy response of the XP20D0 PMT. In our investigations with LaBr₃(Ce) crystals coupled to Hamamatsu R9779 PMTs it is shown that at the normal operation voltages, where the best time resolution is achieved, the nonlinear effects at low energies can be neglected [12, 20]. The R9779 PMT shows a much better time resolution than detectors equipped with the same crystals but coupled to XP20D0 PMT [9, 17], and therefore it is the reference photomultiplier tube used in combination with LaBr₃(Ce) for ISOLDE experiments, with a time resolution down to 110 ps FWHM at ⁶⁰Co energies for conical LaBr₃(Ce) detectors [15].

3.3. Time calibrations

An important characteristic of the ATD method is the analysis procedure involving accurate calibrations of the time response of the system. This has special impact for the measurement of lifetimes of nuclear excited states below 5 ps accuracy when the centroid shift technique is used.

Two main calibrations are required for the γ scintillator detectors, one for the determination of the time response for prompt events where the full-energy of γ-rays has been absorbed in the crystal (full-energy-peak curve) and a second one due to Compton events, where part of the energy escapes the crystal as a Compton-scattered photon [8]. An example of a Compton curve for ²⁴Na is shown in figure 2. For high precision demanding experiments, a third correction (Compton correction) is applied to account for the time dependence of Compton events at a given energy originating from primary photons of different energy [8]. The good energy resolution of LaBr₃(Ce) crystals has made it possible to extend the technique and the calibration procedures for gamma–gamma lifetimes in large fast-timing arrays [14, 22]. Moreover, thanks to this property, it may be possible to apply the Compton walk correction using the data under analysis, without need of external calibrations.
For the calibration of the fast scintillator detectors standard sources were typically used at the ISOLDE facility. They are chosen due to the suitability of their emitted \( \gamma \)-rays that de-excite levels with well known lifetimes, together with their ground state decay lifetime and the easiness to be produced at ISOLDE for either online or offline use. The most widely used are \(^{24}\text{Na} \rightarrow ^{24}\text{Mg}\) (\( \gamma \)-rays at 1368.6 and 2754.0 keV), \(^{88}\text{Rb} \rightarrow ^{88}\text{Sr}\) (898.3 and 1836.0 keV), \(^{138}\text{Cs} \rightarrow ^{138}\text{Ba}\) (\( \gamma \)-rays at 138.1, 227.8, 409.0, 462.8, 547.0, 871.8, 1009.8, 1435.9, 2218.0 and 2639.6) and \(^{140}\text{Ba} \rightarrow ^{140}\text{La} \rightarrow ^{140}\text{Ce}\) (see table 2 for details). The combination of sources allows to calibrate the time response up to 2.7 MeV.

**Table 2.** Level half-lives in \(^{140}\text{La}\) and \(^{140}\text{Ce}\), from the \( A = 140 \) time calibration source. The level lifetime values are taken from measurements at OSIRIS \([23]\) and the literature. Some of the measured lifetimes correct the evaluated values reported in \([24]\).

| Nucleus | Level (keV) | \( J^\pi \) | \( T_{1/2} \) | References | Strong \( \gamma \)'s |
|---------|-------------|-------------|-----------|------------|----------------|
| \(^{140}\text{La}\) | 162.7       | \( 2^- \)   | \( \leq 10 \) ps | \([25]\) and \(-8.3(8.8)\) ps \([26]\) | 162.7           |
| \(^{140}\text{La}\) | 467.5       | \( 1^- \)   | \( \leq 7.7 \) ps | \([25]\) | 304.8, 423.7   |
| \(^{140}\text{Ce}\) | 1596.2      | \( 2^+ \)   | 0.090(25) ps | \([27]\) | 1596.2         |
| \(^{140}\text{Ce}\) | 2083.3      | \( 4^+ \)   | 3.47(10) ns  | \([23]\) | 487.0          |
| \(^{140}\text{Ce}\) | 2349.8      | \( 5^+ \)   | \( \leq 12 \) ps | \([23]\) | —              |
| \(^{140}\text{Ce}\) | 2412.0      | \( 3^+ \)   | 1.3(4) ps    | \([23]\) | 328.7, 815.8   |
| \(^{140}\text{Ce}\) | 2480.9      | \( 4^+ \)   | 22(7) ps     | \([23]\) | —              |
| \(^{140}\text{Ce}\) | 2515.8      | \( 3^+ \)   | \( \leq 2.5 \) ps | \([23]\) | 432.5, 919.6   |
| \(^{140}\text{Ce}\) | 2521.4      | \( 2^+ \)   | \( \leq 2.4 \) ps | \([23]\) | 925.2          |
| \(^{140}\text{Ce}\) | 2547.2      | \( 1^+ \)   | \( \leq 4.0 \) ps | \([23]\) | —              |

Figure 2. Left: ATD \( \beta \gamma(t) \) calibration for the time response to Compton events from a \(^{24}\text{Na}\) source. A gate was set in the 1369 keV peak in the HPGe and projected onto its LaBr\(_3\)(Ce) coincidences. The resulting projection is the spectrum with the 2754 keV photopeak and its long Compton component, which was divided up in gates of around 200 channels each, and projected onto the time spectrum (overlaid). Right: example of a \( \beta \) energy spectrum, taken in coincidences with the HPGe detectors for the \(^{65}\text{Mn}\) decay, shown in channels and in logarithmic scale. Only a fraction of the incident energy is deposited in the plastic. Additionally the digital processing of the signals affects the low and high energy parts. Below can be seen the time response of the plastic detector projected from triple \( bgg \) events before the \( \beta \)-walk correction is applied, and at the bottom the same response after the correction, reproduced with permission from \([21]\).
The plastic detector is compact and very thin with the intention of getting the same energy deposited in the detector independently of the incident $\beta$ energy, so that the time response depends as little as possible on the $\beta$ energy. But not all of the $\beta$ particles are impinging on the detector perpendicularly, so that the effective thickness can be much higher. Furthermore, once inside the plastic the $\beta$ particle may suffer substantial scatter. This is why there is still residual dependence on the deposited energy and the $Q_b$ of the $\beta$ particle, the $\beta$ walk. Due to the continuous nature of the $\beta$ energy distribution and the small time dependence, this is a second order contribution to the time response of the system, but still it is corrected in an event by event basis. An example is provided in figure 2.

3.4. The ISOLDE setup

At the ISOLDE facility we have measured lifetimes in the nanosecond and subnanosecond range of levels in neutron-rich nuclei populated in $\beta$ decay in several regions of the Segré chart. The experiments profit from the beam intensity of many rare isotopes and from the beam purity of ISOLDE beams.

The ISOLDE separator offers excellent beam quality for fast timing studies. The beam implantation spot is very small with an area of a few square millimetres. This allows the arrangement of a few detectors in close geometry, and to place detectors so that the source illuminates the fast $\gamma$ scintillator along its axis of symmetry, which is when the best time response is achieved. We use a compact setup around the beam implantation point, and in order to increase the efficiency we employ two Ge and two fast $\gamma$ detectors, as shown in figure 3. Moreover, the excellent beam emittance makes it possible to use a lead or tungsten collimator 15 cm upstream the implantation point, which is very efficient to suppress
unfocussed beam. Only occasionally drifts of the beam position have been observed. The multiplicity for $\beta$-decay studies is normally not an issue, since typical gamma cascades are composed of two to four $\gamma$-rays.

In order to maximise the count rate for exotic species, many measurements were made with the radioactive source in saturation, that is, with continuous implantation of beam. This allows the study of isobars populated in a given mass decay chain, and has the advantage of a large solid angle efficiency for the detector array, as that used in the LA1 beam line shown in the left-hand side of figure 3. A tape transport system to remove the implanted beam has been used to suppress long-lived daughter activities and contaminants for the investigation of the most exotic nuclei, at the expense of smaller solid angle and losses due to the duty cycle and the process of activity build up and decay. The new IDS setup (right-hand side of figure 3) provides a low background environment, with high-efficiency segmented HPGe detectors that minimise gamma summing effects, and the possibility of installing a tape system where the fast-timing detectors (from a minimum of two to six) can be positioned close to the implantation point.

The fast timing detectors cannot accept count rates higher than $\sim 10$ kcps for the $\gamma$ detector and $\sim 25$ kcps for the $\beta$ detector. This limits the acceptable activity, mostly depending on the efficiency of the $\beta$ detector, which is of the order of 25%–40%. The saturated source activity was typically between 15 and 70 kcps during the experiments at ISOLDE and down to 10 cps for the most exotic species. Due to the pulsed nature of the beam, large instantaneous fluctuations in the intensity can be encountered. This may cause a deterioration of the energy and time resolutions of the detectors because of the response of the PMTs.

ISOLDE can provide selectively ionised mass separated beams very well suited for fast-timing studies, mostly profiting from the ISOLDE RILIS. The main concern is the isobaric contamination that may arise from surface-ionised isobars that cannot be suppressed. Occasionally multiply charged ions with the same mass over charge ratio have been transported to the experimental station together with the main beam. The possibility of contamination is monitored on-line and the portion of contaminated data can be removed if required during the offline analysis.

The electronic setup for the fast scintillator detectors take the last dynode signal from the PMT for energy measurements and the anode signal for time measurements. The energy signal is processed and preamplified if needed, before being sent to the data acquisition (DAQ) system. The anode signal has gone through an additional dynode step, so it is more amplified, and it is negative. It is processed via a NIM-standard ORTEC 935 Quad constant fraction discrimination (CFD), where the discrimination is achieved via an external delay. For the ISOLDE experiments the CFD parameters are optimised for each detector in order to enhance time resolution, which is the key ingredient to achieve a good sensitivity, and not to minimise the time walk [28], since the latter can be taken into account provided it has a smooth behaviour. Analogue time-delayed $\beta\gamma(t)$ coincidences are set up using ORTEC 567 time to amplitude converter (TAC) modules, each started by a signal from the $\beta$-detector and stopped by a fast signal from one of the $\gamma$ detectors. The time range is 50 ns for coincidences between the scintillator detectors. The output from the analogue TACs are fed into the DAQ.

In the early experiments data were collected in the multiparameter (MP) FAST ComTec list-mode multichannel DAQ system, able to handle eight 8192-channel ADCs. In more recent times data are collected using a digital DAQ system. The most widely used consists of four digital gamma finder Pixie-4 modules Revision C by XIA [29]. It is a MP system based on PXI, running at 75 MHz where pulse heights are measured with up to 16 bits accuracy (65536 channels). The energies from the five individual detectors, the four time differences between $\beta$ and each $\gamma$ detector signals and the time difference between the two fast $\gamma$ scintillators are recorded in an ungated triggerless mode. In addition, the time of arrival of the proton pulse on the ISOLDE target, the start and stop of tape transport, and the beam gate
structure are also recorded. For each parameter a Pixie time stamp is included. Coincident events between detectors are sorted off-line using custom software for data analysis.

4. The physics case

The ISOLDE facility provides unrivalled opportunities to study many regions of the nuclide chart with intensities and beam quality not matched in any other facility worldwide. The ATD method has been used to study nuclei populated in $\beta$ decay. Beta decay is an excellent spectroscopic tool due to the inherent selectivity of the decay process: the selection rules for the direct $\beta$ population of states in the decay daughter constrain allowed transitions to proceed from an initial state in the parent to final states in the daughter with the same parity and $\Delta J = 0, 1$.

A summary of the regions studied by the ATD method is illustrated in figure 4. The physics case encompasses topics of interest across the nuclide chart, including the evolution of shell structure around neutron shell and subshell closures such as $N = 20$, $N = 40$ and $N = 50$, shape coexistence, and the study of octupole correlations in heavy actinide nuclei.

4.1. The $N = 20$ island of inversion

The region of the nuclear chart of neutron-rich nuclei centred in $^{32}$Mg ($N = 20$), shows strong modifications of the expected nuclear properties associated with a neutron shell closure. Intruder configurations with $n$ particles in the $fp$ shell and $n$ holes in the $sd$ shell ($np$–$nh$)
dominate the wave function of low-lying states, becoming eventually the ground states. The extra correlation energy of these configurations is the signature of the disappearance of the \( N = 20 \) closure for nuclei in the region, specifically in the case of Mg isotopes \((Z = 12)\). Its origin is linked to modifications of the monopole term of the residual nuclear interaction between valence nucleons and the effect of the quadrupole interaction. For even–even nuclei such as \( ^{30,32}\text{Mg} \), spherical and deformed \( 0^+ \) states coexist a low energies.

The island of inversion has been experimentally studied with many complementary techniques (see contributions to this volume). Fast-timing experiments were performed at ISOLDE in the region in the years 2003–06, with the aim of addressing the coexistence of spherical and intruder configurations.

Magnesium isotopes and their descendants were populated following the \( \beta^- \) decay of \(^{28–32}\text{Na} \) produced by protons impinging on a UC\(_t\) target and surface ionised. The \( \beta^- \) decay populates long decay chains down to the stable isobars. A complete mapping of the Mg nuclei in the island of inversion from its low mass limits up to \(^{35}\text{Mg} \), is possible. For instance, neutron-rich aluminium nuclei are placed at an interesting merging point at the border of the island of inversion, and they can serve to test shell-model calculations using \( sd \) configurations, and including the intruder \( fp \) shell intruders is required. A recent study on \(^{30}\text{Al} \)\(^{30}\) confirms that it is outside the island of inversion and that no sizeable occupation of the \( fp \) orbits is needed to reproduce either the low energy positive parity states, or the measured \( B(M1) \) transition rates.

The Mg isotopic chain is ideally placed to tackle the transition to the island of inversion since it is possible to identify low-lying states and their assignment to normal or intruder configurations based on the lifetimes and transition rates. The nucleus \(^{28}\text{Mg} \) should be located outside the island of inversion. In a recent analysis [31] using the ATD \( \beta\gamma\gamma(t) \) method lifetimes in the nanosecond range were measured for the 54- and 1430-keV states. Together with the existing spin-parity assignments by Shimoda et al (using \( \beta^- \)-decay of spin-polarised radioactive nuclei) [32], the obtained transition rates confirm the intruder structures in \(^{29}\text{Mg} \).

This is in agreement with spectroscopic factors obtained in neutron knock-out reactions [33].

The \( 0^+_1 \) intruder state in \(^{30}\text{Mg} \) was suggested by Mach et al [34] to be located at 1788 keV, based on the rearrangement of the level scheme and the measured \( T_{1/2} \) of 3.9(4) ns half life. Recently the lifetime of the 1788 keV \( 0^+ \) level in \(^{30}\text{Mg} \), populated in the \( \beta^- \)-decay of \(^{30}\text{Na} \) has been obtained with higher precision [31], and limits have been established for the 1482 and 2467 keV states. The excited \( 0^+ \) state at 1788 keV had been confirmed by the direct measurement of the E0 transition [35] together with its monopole strength. In addition, several (not always consistent) measurements of the \( B(E2; 0^+ \to 2^+) \) transition rate exist in the literature [27]. They are supportive of \(^{30}\text{Mg} \) being located outside the island of inversion.

The isotope \(^{31}\text{Mg} \) is an important nucleus located at the border of the island of inversion. Fast-timing measurements on \(^{31}\text{Mg} \) clarified the level scheme [34] including the location of the \( 1p–1h \) 11/2\(^-\) state, and the \( 1p–1h \) 7/2\(^-\) -461 keV level with a long half-life, \( T_{1/2} = 10.5(8) \) ns, de-excited by a collective E2 transition to the 3/2\(^-\) state. The reduced transition rate is very close to the enhanced \( B(E2) \) value for \(^{32}\text{Mg} \), and allows to interpret the states as \( 1p–1h \) configurations, while the ground and first excited state would correspond to \( 2p–2h \) structures. The contribution of intruders to excited states was later investigated in Coulomb excitation [36], and the positive parity states were identified.

In the case of \(^{32}\text{Mg} \), the low energy of the \( 2^+_1 \) state at 885 keV points towards a diminished \( N = 20 \) gap. The \( B(E2) \) reduced transition probability has been measured in inelastic scattering experiments and by Coulomb excitation techniques yielding discrepant values [27]. Therefore, the direct measurement of the lifetime of the \( 2^+_1 \) offered the
opportunity of an independent determination of the B(E2) value. A partial analysis by centroid shift method is reported in [34] and yields \( T_{1/2} = 23(6) \) ps giving B(E2; 0\(^+\) \rightarrow 2\(^+\)) = 327(87) \( e^2 fm^4 \), which is among the lowest values reported to date. The analysis of the complete data set is presently underway. It is worth mentioning that a two-neutron transfer reaction in inverse kinematics with a \(^{30}\text{Mg}\) beam performed at REX-ISOLDE [37] made it possible to identify the shape coexisting 0\(^+\) state in \(^{32}\text{Mg}\) mostly based on normal 0p–0h configuration, and with an estimated lifetime above 10 ns. This level at 1058(2) keV should be populated and measurable in the \( \beta \)-delayed neutron decay of \(^{33}\text{Na}\).

4.2. Shell evolution from \( N = 40 \) to \( N = 50 \)

The nucleus \(^{68}\text{Ni}\) with \( Z = 28 \) and \( N = 40 \) is an anchor point in the study of shell evolution. On one side, the growth of deformation can be studied as \( Z \) increases from the \( Z = 28 \) proton shell closure to the strongly deformed region at \(^{80}\text{Zr} \ (Z = 40) \) [38]. On the other side, the evolution of shell structure in the Ni isotopic chain can be investigated when neutrons are added up to the doubly magic \(^{78}\text{Ni}\) with \( N = 50 \).

It is known that \(^{68}\text{Ni}\) has some of the features of a doubly magic nucleus, such as a high energy of the first excited 2\(^+\) (the highest in the Ni isotopic chain), a low B(E2) transition rate to the ground state, and a first excited 0\(^2\)\(^+\) located below the 2\(^+\)\(^\ast\). Nonetheless, the systematics of two-neutron separation energies [39] do not show evidence for an enhanced \( N = 40 \) harmonic oscillator shell gap. In addition strong collectivity emerges for \(^{68}\text{Fe}\) [40] and \(^{64}\text{Cr}\) [41], with just two and four protons less than \(^{68}\text{Ni}\), as revealed by the low \( E(2\,^1_\pi) \) values and the enhanced B(E2; \( 2\,^1_\pi \rightarrow 0\,^1_\pi \)) rates. This shows that complementary measurements are required to establish new magic numbers and the evolution of the shell structure in exotic nuclei. The particularity of \(^{68}\text{Ni}\) may be explained by the fact that 1p–1h configurations across the \( N = 40 \) subshell will lead to excited negative parity states, while forming a 2\(^+\) state requires a neutron pair promoted from the fp to the \( g_{9/2} \) shell, which is less favoured. In addition \(^{68}\text{Ni}\) provides a very rich case for shape coexistence, because several 0\(^+\) states have been observed below 3 MeV and they seem to originate from excitations across the \( Z = 28 \) and the \( N = 40 \) gaps [42]. The neighbouring odd-A nuclei show a significant polarisation of the \(^{68}\text{Ni}\) core when nucleons are coupled, which implies also a reduction of the \( N = 40 \) subshell [43].

Fast-timing studies at ISOLDE have been focussed mainly on neutron-rich Fe nuclei and on \(^{68}\text{Ni}\) itself. These nuclides were populated in the \( \beta \)-decay of Mn isotopes, strongly produced at ISOLDE on a UC\(_3\) target, and selectively ionised by the resonant ionisation laser ion source (RILIS) yielding high purity beams. Many nuclei populated in the \( \beta \)-decay chains of \(^{59}–^{67}\text{Mn}\), including the \( \beta \)-delayed neutron branches, can be accessed for fast timing studies. The experiments are complementary to other techniques, such as Coulomb excitation and transfer reactions, used at ISOLDE and described in other chapters of this special issue. The lifetimes in this context can aid the interpretation of Coulomb excitation experiments, since the combination of CoulEx cross sections and level lifetimes allows us to measure spectroscopic quadrupole moments, see for instance [44].

The even–even \(^{66}\text{Fe}\) has been subject of investigation since the erosion of the \( N = 40 \) subshell gap and the development of collectivity is observed already when two protons are removed from \(^{68}\text{Ni}\). For \(^{66}\text{Fe}\), the recoil distance Doppler shift method had been used to measure the \( \tau = 39.4(40) \) ps lifetime of the \( 2\,^1_\pi \) state [45], yielding B(E2; 0\(^+\) \rightarrow 2\(^+\)) = 21(2) \( W_u \) in accordance with the Coulomb excitation value of 18(2) \( W_u \) [46]. Our fast-timing measurement [47] expands the level scheme, and cross-checks the half life of the \( 2\,^1_\pi \) state. In addition four other state lifetimes in \(^{66}\text{Fe}\) have been measured, which allows to tentatively establish their
spin-parity. These results are compared with large-scale shell-model calculations based on the Lenz–Nowacki–Poves–Sieja interaction [48].

Odd-A nuclei are specially suited for ATD $\beta\gamma(t)$ studies, which has prompted the study of Fe isotopes to be extended to $^{61,63,65,67}$Fe. The low energy structure of $^{65}$Fe [49, 50] and $^{65}$Mn [51] has been studied in detail by means of $\gamma$- and fast-timing spectroscopy. In [50] the level scheme of $^{65}$Fe populated following the $\beta^-$ decay of $^{65}$Mn was established for the first time, including the determination of the excitation energy of the $\beta^-$-decaying isomer at 393.7(2) keV. This level is based on the $g_9/2$ intruder orbital and provides information on the systematics of positive parity intruders into the negative $pf$ shell. Four lifetimes and five lifetime limits in the subnanosecond range were measured, allowing tentative spin and parity assignments to be established and compared to calculations and to the systematics of the region. Spin-parity assignments for the low-lying negative parity $1/2^-$, $3/2^-$, and $5/2^-$ states can be made based on the measured transition rates, and the positive parity levels are also identified. The structure is very similar to that observed in $^{63}$Fe, where the half-lives of 805(30) and 125(20) ps for the 451.1 and 357.3 keV levels, respectively [21, 49, 50], make it possible to establish the E2 character for the 451.1 keV transition, and the M1 multipolarity for the 93.8- and 375.3 keV transitions. With this information the sequence of the low-lying negative-parity states is $1/2^- \rightarrow 3/2^- \rightarrow 5/2^-$, as illustrated in figure 5.

For the study of lifetimes in Ni isotopes we take advantage of their population in the long $\beta$-decay chain. The investigation addresses shape coexistence in these nuclei via the identification of the $\gamma$-decay branches and the measurement of transition rates, specially from the excited, shape coexisting, $0^+$ states. Complementary measurements of E0 monopoles are required to characterise the $0^+ \rightarrow 0^+$ strength. The coexistence in $^{66}$Ni $N = 38$ can be investigated in this way. Its structure is known from $\beta$-decay [52] and from deep-inelastic scattering [53], which shows a large population to the $0_1^+$ state at 2671 keV, above the second $0_2^+$ state at 2443 keV and the $2_1^+$ level at 1425 keV. The lifetime of the $0_1^+$ state yields the transition rate to the $2_1^+$ state [54] and allows comparison to other transition probabilities in this nucleus to help elucidate the deformation of this state.

In the case of $^{68}$Ni a recent fast-timing experiment [55] was focussed on measuring the half-life of the $0_1^+$ state [56] to provide information on the shape coexistence in this nucleus. The low-spin states in $^{68}$Ni were populated in $\beta$ decay of the low-spin isomer in $^{68}$Co, which is selectively fed in the $\beta$-decay of the $^{66}$Fe $0^+$ ground state. The half-life of the 2511-keV $0^+$ state, estimated to be in the range of nanoseconds, can be measured using coincidences between the populating 1515 keV and de-populating 478 keV transitions.

4.2.1. Simple nuclei around $N = 50$. The strong spin–orbit interaction should make the $N = 50$ spin–orbit shell gap much more favoured than $N = 40$ harmonic oscillator magic number. At ISOLDE we have used the ATD $\beta\gamma(t)$ method to study nuclei around the double shell closure at $^{78}$Ni. They were populated in the decay of $^{77–82}$Zn, which were produced by neutron-induced fission caused by protons from the CERN PS-Booster impinging on the neutron converter of a UC$_3$ target. The target was equipped with a temperature-controlled quartz glass transfer line to suppress contaminants [57]. Selective ionisation was performed by the ISOLDE RILIS.

The $N = 50$ nucleus $^{81}$Ga was investigated after the $\beta$-decay of $^{81}$Zn. This should in principle be a simple nuclear system with three particles outside the doubly magic $^{78}$Ni core that should be well described in a shell-model picture. The lifetime measurements will constrain the model parameters and serve to verify its predictions. In our measurement [58, 59] we have significantly extended the available decay scheme and measured the half-life
of the first excited state in $^{81}$Ga to be $T_{1/2} = 60(10)$ ps. This value indicates an $l$-forbidden $M1$ transition of 351 keV to the $5/2^-$ ground state that should happen between states based on the proton $p_{3/2}$ and $f_{5/2}$ configurations. This is supported by shell-model calculations and by the systematics of $N = 50$ isotones. For the second excited state the half-life of $\sim 25$ ps allows to propose a $f_{5/2}$ configuration and a $3/2^-$ spin-parity assignment.

For the $N = 49$ nucleus $^{80}$Ga, with three proton particles and one neutron hole outside the doubly magic core, the forecast configurations arise from the coupling of a $\pi f_{5/2}$ particle and a $\nu g_{9/2}$ hole. In a recent investigation by $\gamma$ and fast-timing spectroscopy [60], the spin-parity of the ground state was firmly assigned to be $6^-$, and the previously unknown excitation energy of the low-lying isomer observed by collinear laser spectroscopy [61] was established at 22.4 keV, in agreement with shell-model calculations. Candidates for the $\pi f_{5/2} - \nu g_{9/2}$ multiplet members were found. A new isomeric level with the half-life of $18.3(5)$ ns was observed at 707.8 keV, and the de-exciting 685.4-keV transition was assigned $M2$ multipolarity. The measured spectroscopic observables agree with shell-model calculations using the $jj44bpm$ and $JUN45$ interactions.

Nuclei down the $\beta$-decay chains of the Zn parents grant access to interesting nuclei in the surroundings of $^{78}$Ni. The odd-mass $N = 49$ isotope $^{81}$Ge, with $Z = 32$ is populated in the

![Figure 5. An illustration of the measurement of lifetimes for the first and second excited levels in $^{63}$Mn [49, 50]. The lifetimes are long enough to be measured from the slope of the delayed part of the time distribution. On the right-hand side, the slope in the upper panel is due to the lifetime of the 451 keV level, while the slope in the lower panel has two components originating from the lifetimes of the 451 and 357 keV levels, reproduced with permission from [21].](image-url)
$^{81}\text{Zn} \rightarrow ^{81}\text{Ga} \rightarrow ^{81}\text{Ge}$ $\beta$-decay and allows to explore the neutron configurations. The long-lived $1/2^-$ state, based on a single-particle $p_{1/2}$ configuration, changes its behaviour compared to the systematics of the isotones. A pair of $1p$–$2h$ intruder states based on two $g_{9/2}$ holes and an excited neutron gives levels with $1/2^+$ and $5/2^+$ spin-parity. We have measured the half-life of the 711 keV state in $^{81}\text{Ge}$ and confirmed its intruder $5/2^+$ nature. The half-lives of the 895.5, 1724 and 1832 keV levels have been measured as well. The $B(X\lambda)$ values of the de-exciting $\gamma$ transitions allow to test the $1/2^+$ assignment for the 679-keV isomer.

4.3. An excursion to the neutron-deficient region

ISOLDE also offers excellent capabilities to investigate neutron-deficient nuclei. Such is the case of $^{76}\text{Kr}$, which was studied after the $\beta$-decay of $^{76}\text{Rb}$ combining fast timing measurements, conversion electron and $\gamma$-ray spectroscopy [63]. The study was aimed at understanding shape coexistence in this nucleus via the identification of excited $0^+$ states and the measurement of the E0 and E2 transition strengths of the de-exciting transitions. The excited structure was populated in the decay of $^{76}\text{Rb}$. Several new levels and transitions were identified. The lifetimes of many of them in the tens of picoseconds were measured by means of the ATD $\beta\gamma\gamma(t)$ method. Negative parity states were unambiguously identified at surprisingly low excitation energy for an even–even nucleus, below 2.6 MeV. The third $0^+$ state was identified and the decay properties of the $0^+_2$ and $0^+_3$ states were studied. An experimental value of $\rho^2(\text{E0}; 0^+_2 \rightarrow 0^+_1)$ was obtained and compared to calculations in a simple two-states mixing model. No simple description is able to account for the complete experimental data set, pointing towards a complex structure of the $0^+_2$ state, which can only understood if both single-particle and collective degrees of freedom are considered.

Following the observation of low-lying $0^+$ states, Coulomb Excitation studies [64] showed that the spectroscopic quadrupole moments for the bands built on the ground state and on the excited $0^+_2$ state have opposite signs. The triaxial degree of freedom is claimed to be required to describe the excited $0^+$ state bands, and shape coexistence in the light Krypton isotopes. This analysis requires the combination of nuclear lifetime data and Coulomb Excitation cross sections, and underlines the need for complementary techniques to achieve a complete description of complex nuclei.

Experimental fast-timing investigations of this kind can be extended to other neutron-deficient nuclides where the fast timing $\beta\gamma\gamma(t)$ method provides access to lifetimes of excited states and allows to characterise the de-exciting transitions. One of such regions lies around Hg ($Z = 80$) and Pb ($Z = 82$) and here lifetime measurements may complement the ongoing Coulomb excitation [65] and total absorption spectrometry [66] measurements.

4.4. Studies around the doubly magic $^{132}\text{Sn}$

The region of neutron-rich exotic nuclei around $^{132}\text{Sn}$ with $Z = 50$ and $N = 82$ attracts strong experimental and theoretical efforts with the aim of understanding the evolution of the shell structure when moving away from nuclear stability. Transition probabilities play an important role in helping identify the configurations, studying the evolution of collectivity for these nuclei and constraining the shell-model calculations. The main configurations around $^{132}\text{Sn}$ are schematically shown in figure 6.

Antimony nuclei have been studied at ISOLDE via the ATD $\beta\gamma\gamma(t)$ method populated in the $\beta$-decay of Sn produced on a standard UC$_s$ target. To improve the selectivity and avoid the presence of surface-ionised contaminants, the Sn beam was extracted in the form of a
molecular side band by injection of a controlled leak of $^{34}\text{S}$, isotopically enriched to 99.9%. Molecular beams of $^{134}\text{Sn}^{34}\text{S}^+$, $^{135}\text{Sn}^{34}\text{S}^+$, and $^{136}\text{Sn}^{34}\text{S}^+$ with negligible contamination were produced, surface ionised, mass separated and employed for $\beta$-decay experiments.

The nucleus $^{135}\text{Sb}$ is one of the most exotic nuclei north–east of $^{132}\text{Sn}$ where one can obtain detailed spectroscopic information. With two neutrons and one proton above the double shell closure it should provide information on the proton single particle orbitals. A very low-lying first excited state at 282 keV is known with a very weak non-collective E2 and a strongly retarded M1 strength for the 282 keV transition to the ground state [67]. Our fast timing measurements were able to identify the $\gamma$ transitions and levels in $^{135}\text{Sb}$ populated in the beta-delayed neutron emission of $^{136}\text{Sn}$ [68], including the missing $1/2^+$ state at 523 keV excitation, arising from coupling of the $d_{5/2}$ proton orbital to the collective core. Its half life of $T_{1/2} = 1.24(6)$ ns entails a very collective transition with $B(\text{E2}; 1/2^+ \rightarrow 5/2^+) = 12.7(6)$ Wu, well above the limits expected from the neighbouring even–even nuclei.

The low-lying structure of the nucleus $^{134}\text{Sb}$, arises from the coupling of a $g_{7/2}$ proton and a $f_{7/2}$ neutron to $^{132}\text{Sn}$. The lifetime of the $3^-$ member of the multiplet at 383 keV is reported in [68]. The reduced probably for the $3^- \rightarrow 2^-$ transition is $B(\text{M1}) = 2.0(4) \mu_N^2$, making this M1 transition one of the fastest in all known nuclei at excitation energy below 3 MeV. Shell-model calculations performed by Brown and by Covello and Gargano [69] agree with this experimental value.

The systematics of the B(E2; $0^- \rightarrow 2^+$) transition probabilities for even–even nuclei has been subject of scrutiny after the publication of the very low value of this transition rate in $^{136}\text{Te}$ [70]. With the ATD $\beta\gamma$() method we were able to independently measure the rate from the the mean life of the $2^+$ state, yielding a preliminary value of $B(\text{E2}; 0^- \rightarrow 2^+) = 122(24) \ e^2f m^2$ [71]. This value is 20% faster than the previously reported one, but still does not completely explain the departure from systematics for $^{136}\text{Te}$.

4.4.1. The Sn isotopes. Realistic shell-model calculations using an effective interaction derived from the CD-Bonn nucleon–nucleon potential lead to a good description of the structure around $^{132}\text{Sn}$ [72]. The model space includes the neutron-particle orbitals $0h_{9/2}, 1f_{7/2}, 1f_{5/2}, 2p_{3/2}, 2p_{1/2}, 0h_{11/2}$ for Sn isotopes with $N > 82$ and the neutron-hole orbitals $0g_{7/2}, 1d_{5/2}, 1d_{3/2}, 2s_{1/2}, 0h_{11/2}$ for $N < 82$. At the ISOLDE we have investigated the low-lying structure of $^{129}\text{Sn}$ populated in the decay of the $^{129}\text{In}$ low-spin isomers and

![Figure 6. Sketch of proton and neutron configurations that play a role in nuclei around $^{132}\text{Sn}$.](image-url)
measured the half-life of the lowest $1/2^+$ state in $^{129}$Sn populated in the $\beta^-$ decay of $^{129}$In \[73\]. The measurement was carried out in the new IDS. Although the $315.3$ keV $12^+_1$ state, expected to be dominated by the $\nu d_{3/2} s$ single particle configuration, and the $3^2_2$ ground state, based on the $\nu d_{3/2}$ configuration, should be connected by a retarded $l$-forbidden M1 transition, the measurement yields $T_{1/2} = 19(10)$ ps, implying a enhanced transition rate of $B(M1) = 0.036(19)$ Wu. The shell-model calculations with standard $g$-factors and effective charges will give a 4 ns half-life for the $1/2^+$ level, and only when an effective M1 operator consistent with the derivation of the effective interaction is used a significantly smaller value, $T_{1/2} = 195$ ps, is obtained. Thus, the short lifetime of the $1/2^+$ level in $^{129}$Sn does not necessarily imply a change in the shell structure, but can be explained as arising from the renormalisation of the M1 operator \[73\].

It is of great interest extending the investigation to the next odd Sn isotope, $^{131}$Sn, the nearest neighbour below $^{132}$Sn and also to probe the interaction for neutron particles for $^{133}$Sn, across the $N = 82$ gap. The ISOLDE facility provides excellent beams to study $^{132}$Sn itself and the chain of Sn isotopes. They can be populated in the $\beta$-decay of In isomers, produced from a UC$_x$ target unit equipped with a neutron converter and ionised with RILIS. Moreover RILIS adds the capacity of selective isomer ionisation, specifically for the odd-A isotopes, where the large hyperfine splitting in the high-spin states makes it possible to use narrow band laser to select a particular isomer with an estimated ratio exceeding ten to one. With this in mind an experiment has been recently performed \[74\] at the IDS to investigate the doubly magic $^{132}$Sn and its four neighbours: the $2n$ hole $^{130}$Sn, the $1n$ hole $^{131}$Sn, the $1n$ particle $^{133}$Sn and the $2n$ particle $^{134}$Sn. Sample spectra for $^{132}$Sn are shown in figure 7.

### 4.5. Study of octupole correlations

The investigation of octupole correlations was the first application of the ATD $\beta\gamma(\gamma)$ method at ISOLDE, shortly after its move to the PS-Booster. Intense octupole correlations leading to asymmetric nuclear shapes are known to develop in certain regions of the nuclear chart, where octupole–octupole interactions contribute to the equilibrium energy (see \[75\] for a recent review). In a multipole expansion of the nuclear surface, the magnitude of the octupole correlations depend on the matrix elements of the $Y_{30}$ spherical harmonic between single
particle states that fulfil the condition $\Delta j = \Delta l = 3$. This occurs for particle numbers just above shell closures and approximately around 34 ($p_{1/2} \leftrightarrow g_{9/2}$), 56 ($d_{5/2} \leftrightarrow h_{11/2}$), 88 ($f_{7/2} \leftrightarrow p_{1/2}$), and 134 ($g_{9/2} \leftrightarrow j_{15/2}$). The strongest correlations are expected in the Ba region for $Z \sim 56$ and $N \sim 88$, and in the Ra region with $Z \sim 88$, $N \sim 134$.

Fast-timing measurements at ISOLDE were focussed on the island of octupole deformation around $A = 225$, and on the Sm/Ba region. Pioneering studies in the Ba region were aimed at complementing the information on $^{142,144,146}$Ba studied in OSIRIS at Studsvik and TRISTAN at Brookhaven National Laboratory [76]. Barium nuclei were populated in the $\beta$-decay of Cs, and the investigation was addressed to the exotic $^{148}$Ba and $^{149}$Ba nuclides. The $^{149}$Ba level scheme was constructed and lifetimes of low-lying states were measured [77]. Complementary studies were performed at OSIRIS [78]. Recent results from a new experiment with enhanced capabilities performed at the IDS are reported in a contribution to this special issue [79]. Only recently direct evidence of an enhanced $B(E3; 3^- \rightarrow 0^+)$ transition rate in $^{144}$Ba, consistent with stable octupole deformation, has been reported after a Coulomb excitation measurement [80].

The heavy Ra region is probably the best suited to address octupole correlations in nuclei. In even–even isotopes, such as $^{222}$Rn, the energy spectrum is characterised by a negative-parity band of alternating spins built on the $1^-$ state close to the positive parity ground state $0^+$ band. These opposite parity states are connected by enhanced transitions, and specifically the $E3$ moment is the best suited observable to be correlated with intrinsic octupole deformation [75]. Recent Coulomb excitation studies at REX-ISOLDE probed $E2$ and $E3$ transitional matrix elements via excitation cross sections and indicate a moderate enhancement of collectivity for $^{224}$Ra and $^{226}$Ra [81, 82]. One of the first fast-timing measurements performed at ISOLDE, combined with electron spectroscopy, investigated $^{228}$Ra [83]. It resulted in half-life limits of 7 and 6 ps for the $1^-$ and $3^-$ members of the $K^\pi = 0^-$ band that yielded very fast rates ($B(E1) \gtrsim 4 \times 10^{-8} e^2f m^2$) for the de-exciting transitions, which revealed octupole correlations in this nucleus.

A specific effort was put into the ATD $\beta\gamma\gamma(t)$ study of odd-A nuclei in the Ra region. In these nuclei the odd nucleon couples to the reflection-asymmetric even–even core leading to parity doublet bands, where in the limit the states with the same spin but with opposite parity are degenerate in energy. This is a region of evolution of the Ra, Ac and Th from the spherical $N = 126$ to the quadrupole-deformed $N = 142$ region, and it is ideally located to explore the interplay of the quadrupole and octupole degrees of freedom. In $\beta$-decay studies a wealth of states is populated in the daughter nucleus, and parity partner bands can be identified. Due to the small energy split, relatively long-lived states are expected, and fast-timing makes it possible to directly measure their lifetimes in the low picosecond to nanosecond regime. This provides a direct measurement of $B(E1)$ transition rates and $D_0$ dipole moments, which are sensitive to the configurations. The $E1$ moments can be compared with those at higher spin and excitation energies, where in principle the rates will be higher because of the stabilised rotation. Moreover, the quenching of $B(E1)$ rates in certain nuclei can be studied, and its dependence on specific intrinsic configurations of the parity doublet bands in the odd-nuclei investigated.

Several odd-A nuclei in the octupole Ra–Th region exhibit parity doublet bands separated by less than 100 keV, such as $^{223,225,227}$Ra and $^{231}$Ra, $^{223,225,227}$Ac and $^{227}$Th. At ISOLDE several experiments aimed at odd-Ra isotopes were performed in nuclei around the central point of octupole deformation in the region ($^{222}$Ra) where the isotopes of interest were populated in the spallation reactions induced by the PS-Booster 1 GeV proton beam on UC$_x$ targets. These include $^{223}$Ra [84], where electric dipole moments are below what it is...
expected for the centre of the octupole deformation, and the heavier $^{227}$Ra [85], $^{229}$Ra [86] and $^{231}$Ra [87]. For $^{228}$Ra the half-lives reveal $|D_{0}|$ values for the low-spin $K^\pi = 3/2^\pm$ parity partners that are much larger than those for $K^\pi = 1/2^\pm$ and $K^\pi = 5/2^\pm$, and also than those observed for the same $K^\pi = 3/2^\pm$ band for higher spin states.

The $^{227}$Ra investigation was the first complete ATD $\beta\gamma\gamma(t)$ study done at PSB-ISOLDE [85]. Several excited level lifetimes were measured for the first time unveiling strong E1 transitions connecting parity doublet bands. The transition rates for transitions connecting states within a parity doublet are similar to those for $^{223}$Ra, although some of them suffer an intense quenching. There is a strong dependence of the $|D_{0}|$ values on the configuration. The nuclei $^{228}$Ra and $^{231}$Ra were investigated at ISOLDE combining $\gamma$ and internal conversion electron spectroscopy with lifetime measurements of excited states [86, 87]. The reduced transition probabilities, in particular the B(E1) rates of the transitions connecting the lowest lying $K^\pi = 5/2^\pm$ and $1/2^\pm$ bands in both nuclei, show a gradual reduction of the $|D_{0}|$ moment when moving out from the centre of the island of octupole deformation, but nevertheless octupole correlations, stronger in $^{229}$Ra and weaker in $^{231}$Ra, still persist. The octupole effects seem to remain, although very weak, even in the transitional nucleus $^{231}$Ac [88].

The boundaries of the island of octupole deformation in this mass region were also explored with the fast timing $\beta\gamma\gamma(t)$ method. For $^{227}$Fr, populated in the $\beta$-decay of $^{227}$Rn, the observed E1 strength for transitions connecting the lowest $K^\pi = 3/2^\pm$ bands confirm the presence of octupole correlations and are consistent with the transitional character of $^{227}$Fr [89]. Enhanced E1 transitions for selected bands were still observed for the octupole-vibrational $^{229}$Th [90, 91] and $^{231}$Th [92], located at the north border of the octupole region of the actinides, where the interplay between reflection-symmetric and asymmetric configurations emerge. In this context it will be of interest to characterise the Rn nuclei located on the south border of the island of octupole deformation, which could be populated in the $\beta$-decay of negative At ions [93].

5. Summary and outlook

The measurement of nuclear state lifetimes via ATD $\beta\gamma\gamma(t)$ spectroscopy has been established at ISOLDE for nuclear structure studies of isotopes populated in $\beta$-decay. A compact setup optimised for the quality and size of the ISOLDE beam has been employed in experiments for the last 25 years, sometimes in combination with gamma and internal conversion electron spectroscopy. A wide span of the nuclide chart has been investigated, from the light neutron-rich magnesium isotopes up to the region of octupole correlations of the heavy radium. The physics themes range from the modification of shell structure for exotic nuclei in the vicinity of closed shells to the understanding of nuclear collectivity and the coexistence of nuclear shapes.

With the installation of the new IDS a dedicated setup with improved capabilities for fast timing $\beta\gamma\gamma(t)$ measurements is in place. This adds up to novel, high-performing scintillator crystals for $\gamma$-ray detection coming into the market, to improved methodology and calibration procedures [22] and to enhanced data processing including digital acquisition [94]. All in all, it will make precision measurements of lifetimes of excited states down to tens of picoseconds available at ISOLDE for extended regions of the nuclear charge. Moreover, the future High Intensity and Energy upgrade of ISOLDE (HIE-ISOLDE) is expected to provide even higher yields of exotic nuclei. Experiments are intended to explore, among others, the neutron deficient Hg–Po region, the Zn structure populated from the $\beta$-decay of Cu isotopes [95] and the structure of $n$-rich Ar nuclei [96]. The use of a tape transport system grants better access to
the study of exotic nuclei further away from stability. In the future, fast-timing trap-assisted spectroscopy, achieved by coupling a timing detection setup to a Penning trap system such as ISOLTRAP [97] or to a multi-reflection time-of-flight mass separator [98], will open huge possibilities for fast-timing experiments with isobarically or even isomerically pure radioactive beams at ISOLDE.

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