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Nuclear Spectroscopy with `Geant4`: Proton and Neutron Emission & Radioactivity.

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Abstract. With the aid of a novel combination of existing equipment – JYFLTRAP and the TASISpec decay station – it is possible to perform very clean quantum-state selective, high-resolution particle-γ decay spectroscopy. We intend to study the determination of the branching ratio of the $\ell = 9$ proton emission from the $I^\pi = 19/2^-$, 3174-keV isomer in the $N = Z - 1$ nucleus ^{53}Co. The study aims to initiate a series of similar experiments along the proton dripline, thereby providing unique insights into "open quantum systems". The technique has been pioneered in case studies using SHIPTRAP and TASISpec at GSI. Newly available radioactive decay modes in `Geant4` simulations are going to corroborate the anticipated experimental results.

INTRODUCTION

Over the years it has become increasingly more relevant to assist the experimental data analysis with detailed simulations. They have proven useful to characterize the detector's response to different experimental scenarios and even in the physics interpretation of experimental data [1]. For nuclear physics there are couple of simulation toolkits available to the public. Of particular popularity are `MCNP` [2], `FLUKA` [3], and `Geant4` [4] which are considered the *de facto* standards. The latter is often preferred by many application developers over the formers, due to its free software nature.

A central topic in contemporary nuclear structure physics is the investigation of exotic nuclear matter far from the line of β stability. Experimentally, one way of investigating the outskirts of long isotopic chains is by performing detailed and clean spectroscopy of ground or low-lying excited states in nuclei near or at the driplines. Of specific relevance is the preparation of an isotopically pure beam or source.

The purpose of this initiative is to start a series of studies aiming at high-resolution particle-γ decay spectroscopy in the vicinity of the proton dripline. The first study case concerns the branching ratio, thus spectroscopic factor, of the unique $\ell = 9$ proton decay of the $19/2^-$ isomer in ^{53}Co^m – more than 40 years after its discovery and marking proton radioactivity on the nuclear landscape [5, 6, 7]!

The hereby mentioned and potential future experiments along the $N \sim Z$ line between ^{56}Ni and ^{100}Sn rely on a novel combination of equipment: A Penning trap prepares the selected ground or isomeric state of an isotope near or at the proton dripline. Instead of measuring their masses, these isotopes are directly transferred into a high-resolution particle-γ coincidence set-up. Here, detailed decay characteristics of a specific nuclear quantum state are to be measured. This scheme, which is illustrated in Fig. 1(a), has been tested and commissioned at GSI [8, 9, 10] using SHIPTRAP [11] and TASISpec [12]. The latter was originally developed for superheavy-element spectroscopy behind the gas-filled separator TASCA at GSI Darmstadt [1]. In general, the development of this type of quantum-state selective decay spectroscopy is also relevant in the context of FAIR-NUSTAR [13], let it be behind MATS or MR-ToF systems.

In short, TASISpec covers five sides of a cube with pixelized double-sided silicon strip detectors: four 'box detectors' (16 × 16, 0.97 mm thick) and one in beam direction (32 × 32, 0.31, 0.52 or 0.97 mm thick). Each silicon detector is backed by a large composite Ge-detector of former EUROBALL Cluster or Clover type, see Fig. 1(b). Such a combination provides an unprecedented high-resolution particle-photon coincidence efficiency [12] and at the same time sufficient granularity for decay studies. Furthermore, TASISpec exists in virtual `Geant4` space [14, 15]. This allows for a self-consistent cross check of the decay schemes derived from the experimental spectra.

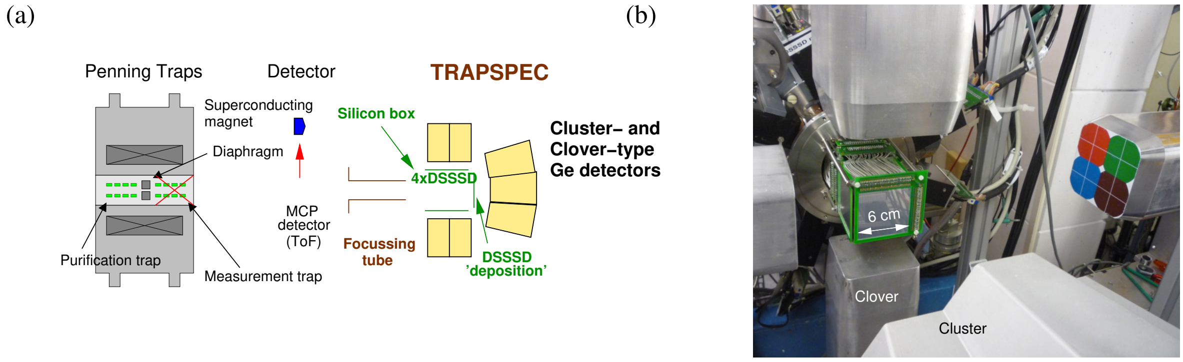

FIGURE 1. (a) Schematic drawing of the test set-up at GSI combining SHIPTRAP [11] purification with TASISpec [12] high-resolution particle-γ decay spectroscopy [8, 9, 10]. (b) Photograph of TASISpec behind the gas-filled separator TASCA at GSI.

THE PHYSICS CASE: PROTON-DECAY BRANCH OF ^{53}Co^{m}

The observation of a weak proton-decay branch in the decay of the $I^{\pi} = (19/2^{-})$ ^{53}Co^{m} isomeric state marked the discovery of proton radioactivity in 1970 [5, 6]. At the time, the proton energy was measured to 1.53(4) MeV [5], 1.57(3) MeV [6], and later-on 1.59(3) MeV [7]. Recent mass measurements of the relevant nuclear states with JYFLTRAP yield 1.530(7) MeV [16]. In Ref. [7], a branching ratio of $b_p \sim 1.5$ % is *estimated*, based on model-dependent comparisons of anticipated peak cross-sections of different reaction products of the reaction p+^{54}Fe. References [7, 17] provide excitation functions for the reactions relevant for the present study. These have been probed recently at IGISOL as well [16].

On the theoretical side, the explanation of proton-decaying states requires a description of the wave functions of initial and final state, as well as a static or, more advanced, time-dependent approach of the quantum tunneling process. Model calculations typically infer experimental values for the decay energies to derive (partial) half-lives or spectroscopic factors to be compared with experiment. Hence, for a complete theoretical assessment of a given case, decay energy Q_p, (cf. Ref. [16]), half-life $T_{1/2}$ [18], and the branching ratio b_p are the required experimental quantities.

Until now, an experimental measure of the proton-decay branching ratio was not available for ^{53}Co^{m} – more than 40 years after discovery [19]. The reason for that becomes clear when looking at the decay characteristics of ^{53}Co^{m} compared with those of ^{53}Co, as illustrated in Fig. 2: They are merely identical! The half-life of the decays can hardly be distinguished, and the dominating β^{+} branches have practically the same energetics, simply because the two states in ^{53}Co decay primarily into the respective isobaric analogue states (IAS) in the daughter ^{53}Fe. Hence, unless one is able to separate the two sources from each other, namely ^{53}Co^{m} and the ground state of ^{53}Co, one cannot measure b_p of ^{53}Co^{m}.

The separation of two nuclear states with an energy difference of more than 3 MeV can be easily done with a Penning trap like JYFLTRAP as shown in, for instance, Ref. [16].

As sketched in Fig. 1(a), once the mass-measurement cycle is established, the nominal MCP-ToF detector can be lifted up from the beam or lowered back to the beam without breaking the vacuum. Thus, the number of ions in the bunch can be estimated by measuring it every now and then but regularly, thereby monitoring the cyclotron beam intensity as a function of time. The ions of interest are then transferred to the particle-γ decay station, a concept proven for the combination SHIPTRAP and TASISpec [10, 19]. This includes a transfer efficiency close to 100 %, with the ions rather being *deposited* on, but not implanted into the central DSSSD in front of the Cluster detector. Particle decays shortly ($\Delta t < 0.25$ s to 0.50 s) after the transfer as well as subsequent prompt coincidences between β^{+} emission, 511-keV annihilation radiation, and/or γ(-γ) events can count the decays of ^{53}Co^{m} along the mass $A = 52$ and $A = 53$ decay chains, respectively (see Fig. 2). Brief reference measurements of the decays of ^{53}Co, ^{53}Fe^{m}, and ^{52}Fe are to be confronted with and thus cross-calibrate `Geant4` simulations in a self-consistent fashion. Though the weak ($b_\gamma < 2$ %) $E4$, 378-keV γ branch into the 6-day ground-state activity of ^{52}Mn may be considered negligible in the context of the ^{53}Co^{m}, even its influence is going to be part of the ^{52}Fe reference measurement. The goal is to determine b_p with a relative precision of 10 to 20 %, depending on its actual size, i.e. $b_p = 0.5(1)$ % to $b_p = 3.0(3)$ %, around the previously estimated value $b_p \sim 1.5$ %.

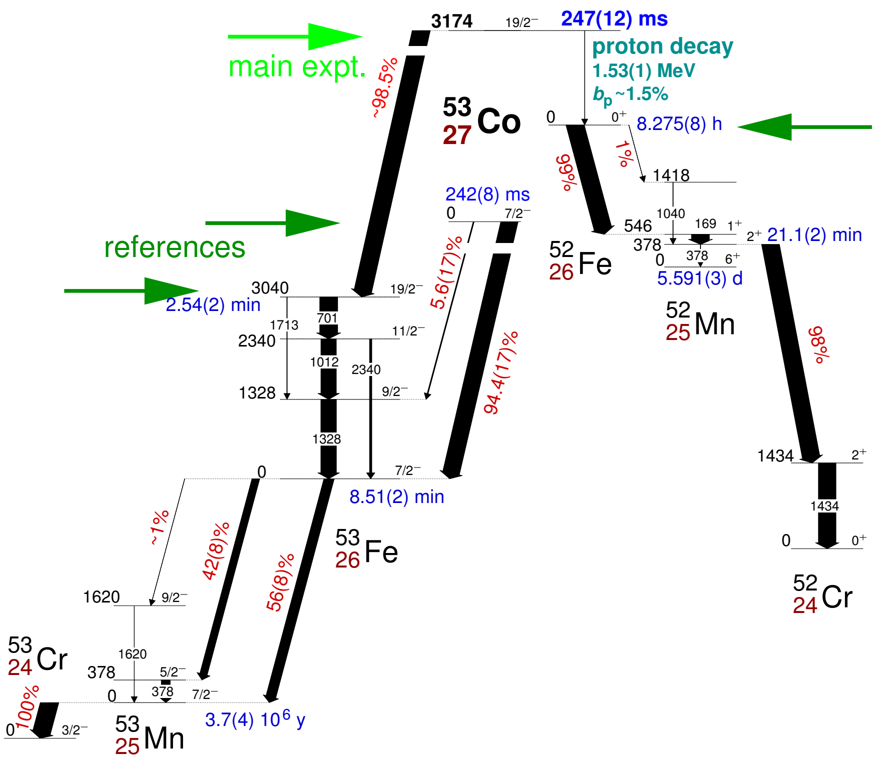

FIGURE 2. Main decay sequences of ^{53}Co and ^{53}Co^m. Level energies are in keV and with respect to the ground state of a given isotope. Gamma-ray energies are given in keV. Half-lives of isomeric and ground states are denoted in blue, β^+/ϵ branching ratios in red. See text for details.

To establish a precise value of Q_p for ^{53}Co^m, the mass excess of the ^{52}Fe ground state should also be measured . Its value, ME=-48332(7) keV [20], is the main contributor to the uncertainty in the proton separation energy of ^{53}Co, S_p = 1615(7) keV [20]. A precise proton separation energy fixes the energy scale of the proton decay. It could also be used for a proper calibration of the silicon detectors.

Although `Geant4` already includes radioactive decay physics since long time ago [21], the decay modes were restricted to α, β^- and β^+/EC. With the aim to cover the physics cases outlined here, the `Geant4` Radioactive Decay Mode has been extended to include the emission of protons in a new class `G4ProtonDecay` which is available since the version 10.2 released on December 4, 2015, which is to be used in conjuction with `G4RADIOACTIVEDATA` v4.3. It should be noted that although in the same release the neutron emission has also been included with a new `G4NeutronDecay` class this decay mode has not been equally verified as the proton decay.

`GEANT4` SIMULATIONS

A virtual `Geant4` version of TASISpec was prepared in connection with a self-consistency check of the interpretation of superheavy element 115 data [1, 14, 15]. On the detector side, details such as individual pixel-by-pixel deadlayers were measured and implemented in the simulations [22]. Physicswise, the `Geant4` package was upgraded towards realistic element $Z \geq 100$ simulations [15] and, for the present study, proton radioactivity.

Figure 3 provides some simulated spectra of the 'deposition DSSSD', probing some detector parameters as well as indicating some analysis methods to highlight the ^{53}Co^m proton-decay branch in the anticipated real experimental data [19]. The spectra are based on 100000 ions, deposited onto and distributed over the detector with a beam of Gaussian shape with 10 mm FWHM in x and y. Typically, all decays happening within 24 h along the A = 52 and A = 53 decay paths shown in Fig. 2 are recorded, i.e. the spectra shown here reflect also the expected long-term particle background.

Panel (a) shows the results for the three available thicknesses of the detector, D=0.31, 0.52, or 0.97 mm. Here,

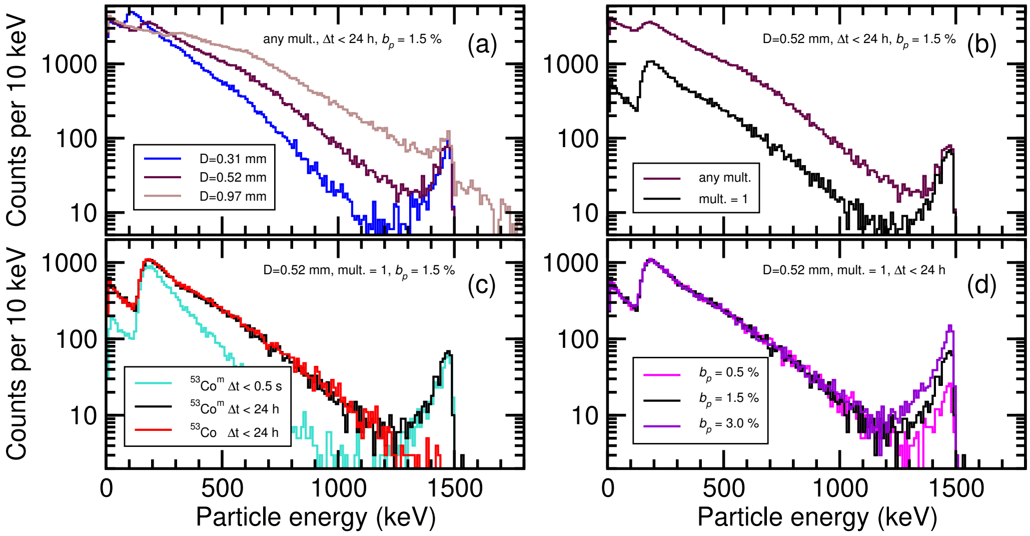

FIGURE 3. Simulated particle energy spectra of the central 'deposition DSSSD' probing various experimental parameters concerning the main ^{53}Co^m decay: (a) Different detector thicknesses (D=0.31, 0.52, and 0.97 mm); (b) Any multiplicity of pixels per decay event or exactly one; (c) Short $\Delta t < 0.5$ s correlation with Penning-trap outlet or practically none (here: $\Delta t < 24$ h); (d) Assuming different proton-decay branching ratios, b_p. See text for details.

the spectra are incremented pixel-by-pixel, i.e. no 'add-back' for neighbouring strips was performed. The reason is that a 1.53 MeV proton is expected to be always stopped in exactly one pixel because of its limited range, while β^+ particles can easily pass through several pixels, depending on emission angle and energy. Note also the triangular shape (in logarithmic scale) of the proton peak. This is due to the fact that the proton first has to penetrate through the $\sim 2\ \mu$m thick deadlayer of the DSSSD, which costs about 25 keV per micron and is angle dependent. Clearly and as expected, the thinner the detector the better the separation of the proton line from β^+ background events. The 0.97 mm thickness was excluded from being useful for the experiment.

Panel (b) illustrates the multiplicity effect using the example of the D=0.52 mm thick DSSSD. Requiring multiplicity = 1, a large amount of β^+ background can be eliminated, and the valley between the proton line and background becomes comparable with the blue spectrum in Fig. 3(a) (D=0.31 mm). Panel (c) dwells on two additional options to separate the proton peak. First of all, the β^+ decay of the ^{53}Co ground state is merely identical in Q-value and decay time to that of ^{53}Co^m. Thus, a properly normalized reference spectrum of ^{53}Co (red) can simply be subtracted from that of ^{53}Co^m (black) to have a plain proton-decay spectrum remaining. Secondly, it is only those two decays which are fast, i.e. which can be correlated with the time of the release from the Penning trap. The *ideal* case is the third spectrum in Fig. 3(c) (cyan). The gap increases, because the high Q-value decays of ^{53}Co and ^{53}Co^m emit on average higher energy β^+ particles, which in turn leave on average less energy in just one pixel of the 'deposition DSSSD'. In reality, the ideal time-correlated spectrum (cyan) will be mixed with grown-in background (red), while a spectrum correlated with 0.5 s < Δt < 1.0 s (for a 1.0-s cycle) will contain primarily this grown-in background, and a substraction of the two may once more provide a clean proton spectrum. (In case a 0.50-s cycle is used, the same method can be applied for spectra taken at Δt < 0.25 s and 0.25 s < Δt < 0.50 s, respectively.)

Panel (d) repeats the black spectrum of panel (c) and compares it to two simulations using either a smaller (b_p = 0.5 %) or larger (b_p = 3.0 %) proton-decay branching ratio. A simplistic analysis of the three spectra, i.e. plain integration [1.38-1.55] MeV vs. [0.10-1.60] MeV returns b_p = 0.59(5), 1.67(8), and 3.06(11) % with uncertainties being purely statistical.

In case the proton or the first ^{53}Co or ^{53}Co^m β^+ is detected in one of the four box DSSSDs, the daughter nuclei are pushed further into the 'deposition DSSSD'. Furthermore, the low-energy β^+ particles from ^{52}Fe cannot counteract the implantation momentum of the protons, i.e. in this case also ^{52}Mn residues will remain at the deposition spot. This allows for subsequent detection and counting of β^+-γ coincidences, sum-energy of the 'deposition DSSSD' vs. Ge-detector energy. There are three interesting cases: for the $A = 53$ β^+ decay branch, there is the β^+-378-keV coincidence in the decay of ^{53}Fe ($E_{\beta,\max}$ = 2.34 MeV). For the $A = 52$ branch populated by the proton decay, both β^+-169-keV

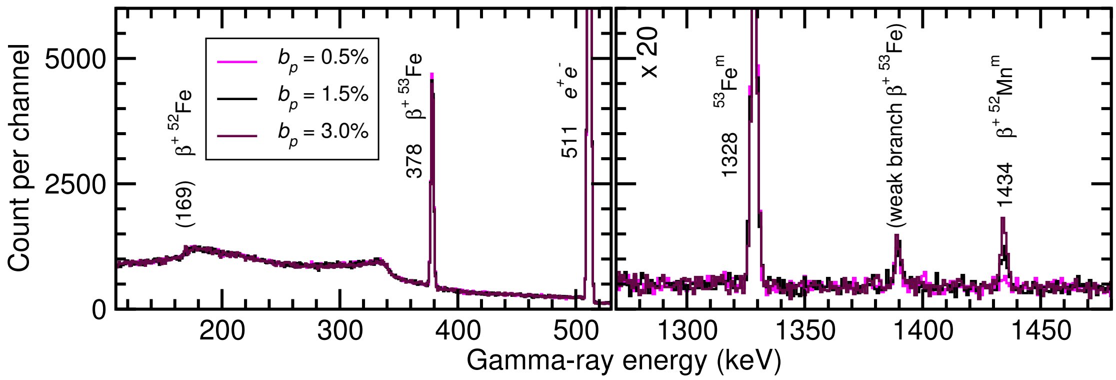

FIGURE 4. Simulated γ-ray energy spectra in prompt (Δt = 300 ns) coincidence with a 0.1-3.0-MeV particle dectected in the central 'deposition DSSSD' assuming different proton-decay branching ratios, b_p. Energy labels are in keV.

(^{52}Fe decay, $E_{\beta,\max}$ = 0.80 MeV) and β^+-1434-keV (^{52}Mn^m decay, $E_{\beta,\max}$ = 2.64 MeV) can be considered.

Figure 4 shows the total γ-ray projection of the particle-photon matrix described above, simulated for 500000 decays of ^{53}Co^m and proton-decay branching ratios of 0.5, 1.5, and 3.0 %. First of all, the 169-keV line can hardly be seen because of a relative small peak-to-background ratio. In turn, the ratio of β^+-γ coincidence yield of 378 keV vs. 1434 keV γ rays carries independent information on the proton branching ratio of ^{53}Co^m; performing standard least-square peak fits in the spectra of Fig. 4, one obtains R = 0.0027(10), 0.0102(13), and 0.0182(16), reflecting the change in b_p (taking into account different γ-ray detection efficiencies at 378 and 1434 keV, respectively). The 701-1012-1328-keV cascade in ^{53}Fe marks another possible 'γ counter' for the β^+ decay branch of ^{53}Co^m. In any case, high-statistics reference measurements of the decays of ^{52}Fe, ^{53}Fe^m, and ^{53}Co are required to properly establish the reference or normalisation yields for this method.

Note finally that the (coarse) ratios of yields extracted from particle-singles spectra (see Fig. 3) or projected γ-ray spectra can be complemented if not superseded by least-square fitting of simulated spectra to experimental ones. Here, alike in Refs. [1, 15], `Geant4`-generated listmode data is going to pass through a data replay identical to that used for the real data. This method allows for a one-to-one comparision of real and `Geant4` listmode data sets, and it is expected to lead to further reduction of uncertainties, both for the reference cases, but primarily for the proton-decay branch.

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