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Enhanced γ-Ray Emission from Neutron Unbound States Populated in β Decay

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Total absorption spectroscopy was used to investigate the β-decay intensity to states above the neutron separation energy followed by γ-ray emission in ^{87,88}Br and ^{94}Rb. Accurate results were obtained thanks to a careful control of systematic errors. An unexpectedly large γ intensity was observed in all three cases extending well beyond the excitation energy region where neutron penetration is hindered by low neutron energy. The γ branching as a function of excitation energy was compared to Hauser-Feshbach model calculations. For ^{87}Br and ^{88}Br the γ branching reaches 57% and 20% respectively, and could be explained as a nuclear structure effect. Some of the states populated in the daughter can only decay through the emission of a large orbital angular momentum neutron with a strongly reduced barrier penetrability. In the case of neutron-rich ^{94}Rb the observed 4.5% branching is much larger than the calculations performed with standard nuclear statistical model parameters, even after proper correction for fluctuation effects on individual transition widths. The difference can be reconciled introducing an enhancement of one order-of-magnitude in the photon strength to neutron strength ratio. An increase in the photon strength function of such magnitude for very neutron-rich nuclei, if it proved to be correct, leads to a similar increase in the (n,γ) cross section that would have an impact on r process abundance calculations.

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Neutron unbound states can be populated in the β decay of very neutron-rich nuclei, when the neutron separation energy S_n in the daughter nucleus is lower than the decay energy window Q_β. Given the relative strengths of strong and electromagnetic interactions these states decay preferentially by neutron emission. Beta delayed γ-ray emission from states above S_n was first observed in 1972 in the decay of ^{87}Br [1]. Since then it has been observed in a handful of cases: ^{137}I [2], ^{93}Rb [3], ^{85}As [4], ^{141}Cs [5], ^{95}Rb [6], ^{94}Rb [7], ^{77}Cu [8], and ^{75}Cu [9]. The paucity of information is related to the difficulty of detecting weak high-energy γ-ray cascades with the germanium detectors that are usually employed in β-decay studies. This problem has become known as the *Pandemonium* effect [10] and it also affects the accuracy of the data.

There is an analogy [11] between this decay process and neutron capture reactions which populate states in the compound nucleus that re-emit a neutron (elastic channel) or de-excite by γ rays (radiative capture). Indeed the reaction cross section is parametrized in terms of neutron and γ widths, Γ_n and Γ_γ respectively, which also determines the fraction of β intensity above S_n that proceeds by neutron or γ emission. Radiative capture (n,γ) cross sections for very neutron-rich nuclei are a key ingredient in reaction network calculations used to obtain the yield of elements heavier than iron in the rapid (r) neutron capture process occurring in explosive-like stellar events. It has been shown [12–14] that the abundance distributions in different astrophysical scenarios are sensitive to (n,γ) cross sections. In the classical "hot" r process late captures during freeze-out modify the final element abundance. In the "cold" r process the competition between neutron captures and β decays determines the formation path. Cross section values for these exotic nuclei are taken from Hauser-Feshbach model calculations [15], which are based on a few quantities describing average nuclear properties: nuclear level densities (NLD), photon strength functions (PSF) and neutron transmission coefficients (NTC). Since these quantities are adjusted to experiment close to β stability it is crucial to find means to verify the predictions for very neutron-rich nuclei.

The Total Absorption Gamma-ray Spectroscopy (TAGS) technique aims at detecting cascades rather than individual γ rays using large 4π scintillation detectors. The superiority of this method over high-resolution germanium spectroscopy to locate missing β intensity has been demonstrated before [16, 17]. However its application in the present case is very challenging, since the expected γ-branching is very small and located at rather high excitation energies. As a matter of fact previous attempts at LNPI [7] with a similar aim did not lead to clear conclusions. In this Letter we propose and demonstrate for the first time the use of the TAGS technique to study γ-ray emission above S_n in β-delayed neutron emitters and extract accurate information that can be used to improve (n,γ) cross section estimates far from β stability.

Neutron capture and transmission reactions have been extensively used [18] to determine neutron and γ widths (or related strength functions). An inspection of Ref. [18] shows that in general Γ_n is orders-of-magnitude larger than Γ_γ. In the decay of ^{87}Br, which is the best studied case [1, 19–21], a dozen states emitting single γ rays were identified within 250 keV above S_n collecting about 0.5% of the decay intensity to be compared with a neutron emission probability of 2.6%. The observation of such relatively high γ-ray intensity was explained as being due to a nuclear structure effect: some of the levels populated can only decay by emission of neutrons with large orbital angular momentum l, which is strongly hindered. In addition it has been pointed out [22] that a sizable γ-ray emission from neutron unbound states can be a manifestation of Porter-Thomas (PT) statistical fluctuations in the strength of individual transitions. The role and relative importance of both mechanisms should be investigated.

We present here the results of measurements for three known neutron emitters, ^{87}Br [23], ^{88}Br [24] and ^{94}Rb [25] , using a newly developed TAGS spectrometer. The results for ^{93}Rb, also measured, will be presented later [26]. The measurements were performed at the IGISOL mass separator [27] of the University of Jyväskylä. The isotopes were produced by proton-induced fission of uranium and the mass-separated beam was cleaned from isobaric contamination using the JYFLTRAP Penning trap [28, 29]. The resulting beam was implanted at the centre of the spectrometer onto a movable tape which periodically removed the activity to minimize daughter contamination. Behind the tape was placed a 0.5 mm thick Si detector with a β-detection efficiency of about 30%. The Valencia-Surrey Total Absorption Spectrometer *Rocinante* is a cylindrical 12-fold segmented BaF_2 detector with a length and external diameter of 25 cm, and a longitudinal hole of 5 cm diameter. The separation between crystals is provided by a thin optical reflector. The total efficiency for detecting a single γ ray is larger than 80%. The spectrometer has a reduced neutron sensitivity in comparison to NaI(Tl) detectors, a key feature in the present application. It also allows the measurement of multiplicities which helps in the data analysis. In order to eliminate the detector intrinsic background and the ambient background we use β-gated TAGS spectra in the present analysis. Nevertheless other sources of spectrum contamination need to be characterized accurately.

In the first place the decay descendant contamination, was computed using the Geant4 simulation toolkit [30]. In the case of the daughter decay we use an event generator based on the well known decay level scheme [23–25]. The calculated normalization factor was adjusted to provide the best fit to the recorded spectrum. The measurement of ^{88}Br was accidentally contaminated by ^{94}Y, the long-lived grand-daughter of ^{94}Rb, and was treated in the same manner. The case of the contamination due to the β-delayed neutron branch is more challenging. The decay simulation must include the correct energy sequence β-neutron-γ. Neutrons interact with detector materials producing additional γ rays through inelastic and capture processes. An event generator was implemented which reproduces the known neutron energy distribution, taken from [31], and the known γ-ray intensity in the final nucleus, taken from [23–25]. The event generator requires the β intensity distribution followed by neutron emission $I_{\beta n}$ which was obtained from deconvolution of the neutron spectrum. Another issue is whether the interaction of neutrons with the detector can be simulated accurately. We have shown recently [32] that this is indeed the case provided that Geant4 is updated with the newest neutron data libraries and the original capture cascade generator is substituted by an improved one. The normalization factor of the β-delayed neutron decay contamination is fixed by the P_n value. Another important source of spectrum distortion is the summing-pileup of events. If more than one event arrives within the same ADC event gate, a signal with the wrong energy is stored in the spectrum. Apart from the electronic pulse pile-up effect for a single detector module [33] one must consider the summing of signals from different detector modules. A new Monte Carlo (MC) procedure to calculate their combined contribution has been developed. The procedure is based on the random superposition of two stored events within the ADC gate length. The normalization of the resulting summing-pileup spectrum is fixed by the event rate and the ADC gate length [33].

Several laboratory γ-ray sources were used to determine the energy and resolution calibration of the spectrometer. The highest calibration point was at 4.123 MeV. The measured singles spectra also served to verify the accuracy of the spectrometer response simulated with Geant4. The use of β-gated spectra in the analysis required additional verifications of the simula-

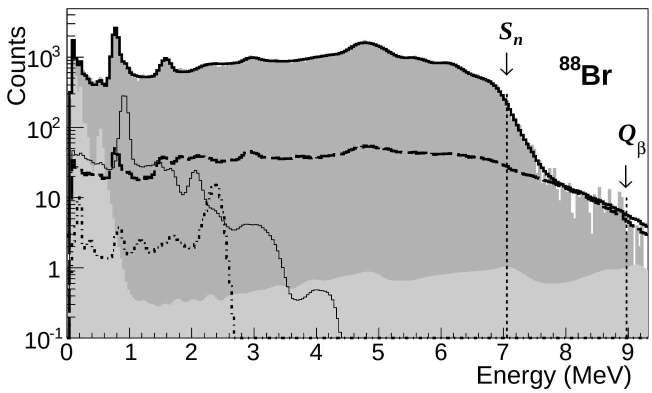

FIG. 1. Relevant histograms for ^{88}Br: parent decay (dark grey filled), daughter decay (dot-dashed line), summing-pileup (dashed line), β-delayed neutron decay (light grey filled), accidental contamination (thin continuous line), reconstructed spectrum (thick continuous line).

tion. Due to the existence of an electronic threshold in the Si detector (100 keV) the β-detection efficiency has a strong dependence with β-endpoint energy up to about 2 MeV. This affects the region of interest (see Fig. 1). To verify that the MC simulation reproduces this energy dependence we use the information from a separate experiment [34] measuring P_n values with the neutron counter BELEN and the same β detector. Several isotopes with different neutron emission windows $Q_\beta - S_n$ were measured, resulting in variations of the neutron-gated β efficiency as large as 25%. Geant4 simulations using the above mentioned β-delayed neutron decay generator are able to reproduce the isotope-dependent efficiency within better than 4%.

Figure 1 shows the β-gated TAGS spectrum measured during the implantation of ^{88}Br ions. Also shown is the contribution of the daughter ^{88}Kr decay, the neutron decay branch populating ^{87}Kr, the summing-pileup contribution and the accidental contamination of ^{94}Y. About 30% of the emitted neutrons produce a signal (light grey filled histogram). Most of the signals, concentrated below 1 MeV, are due to inelastic scattering. Only 1.5% of the neutrons undergo capture depositing energy up to 10 MeV. Notice the presence of net counts beyond the neutron separation energy, which can only be attributed to the decay feeding excited states above S_n which deexcite by γ-ray emission. In this region the major background contribution comes from summing-pileup which is well reproduced by the calculation as can be observed. Similar pictures were obtained for the decay of ^{87}Br and ^{94}Rb.

The analysis of the β-gated spectra follows the method developed by the Valencia group [35, 36]. The intensity distribution $I_{\beta\gamma}$ is obtained by deconvolution of the TAGS spectrum with the calculated spectrometer response to the decay. The response to electromagnetic cascades is calculated from a set of branching ratios (BR) and the MC calculated response to individual γ rays. Branching ratios are taken from [23–25] for the low energy part of the decay level scheme. The excitation energy range above the last discrete level is treated as a continuum divided into 40 keV bins. Average BR for each bin are calculated from NLD and PSF as prescribed by the Hauser-Feshbach model. We use NLD from Ref. [37] as tabulated in the RIPL-3 library [38]. The PSF is obtained from Generalized Lorentzian (E1) or Lorentzian (M1, E2) functions using the parameters recommended in Ref. [38]. The electromagnetic response is then convoluted with the simulated response to the β continuum. The spin-parity of some of the discrete states at low excitation energy in the daughter nucleus is uncertain. They are however required to calculate the BR from the states in the continuum. The unknown spin-parities were varied and those values giving the best reproduction of the spectrum were adopted. There is also ambiguity in the spin-parity of the parent nucleus which determines the spin-parity of the levels populated in the continuum. Here we assume that allowed Gamow-Teller (GT) selection rules apply. Our choices, $3/2^-$ for ^{87}Br, 1^- for ^{88}Br and 3^- for ^{94}Rb, are also based on which values best reproduce the spectrum.

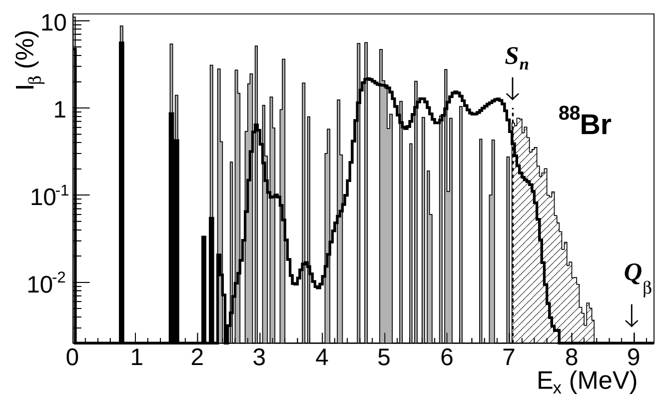

FIG. 2. Beta intensity distributions for ^{88}Br: TAGS result (continuous line), high-resolution γ spectroscopy (light grey filled histogram), from β-delayed neutron (hatched area).

As an example of the results of the analysis we show in Fig. 2 the $I_{\beta\gamma}$ intensity obtained for ^{88}Br. The spectrum reconstructed with this intensity distribution reproduces well the measured spectrum (see Fig. 1). The analysis for the other two isotopes shows similar quality in the reproduction of the spectra. We also include in Fig. 2 the intensity obtained from high-resolution measurements [24], showing a strong *Pandemonium* effect. The *Pandemonium* effect is even stronger in the case of ^{94}Rb and somewhat less for ^{87}Br. The complete $I_{\beta\gamma}$ and its impact on reactor decay heat [39] and antineutrino spectrum [40] summation calculations will be discussed elsewhere [41]. Here we concentrate on the portion of that intensity located in the neutron unbound region. A sizable TAGS intensity is observed above S_n extending well beyond the first few hundred keV where the low neutron penetrability makes γ-ray emission competitive. For comparison Fig. 2 also shows $I_{\beta n}$ deduced from the neutron spectrum [31] as explained above. The $I_{\beta\gamma}$ above S_n adds up to $\sum I_{\beta\gamma} = 1.6(3)\%$, to be compared with the integrated $I_{\beta n}$ (or P_n) of 6.4(6)%. From the TAGS analysis for the other two isotopes we find a $\sum I_{\beta\gamma}$ of 3.5(5)% (^{87}Br) and 0.53(16)% (^{94}Rb) to be compared with P_n-values of 2.60(4)% and 10.18(24)% respectively. In the case of ^{87}Br we find 7 times more intensity than the high-resolution measurement [21]. The uncertainty quoted on $\sum I_{\beta\gamma}$ is dominated by systematic uncertainties. We did a careful evaluation of possible sources of systematic effects for each isotope. The uncertainty coming from assumptions in the BR varies from 1% to 5% (relative value) depending on the isotope. The impact of the use of different deconvolution algorithms [36] is in the range of 2% to 10%. The uncertainty in the energy dependence of the β efficiency contributes with 4%. The contribution of uncertainties in the width calibration ranges from 2% to 6%. A major source of uncertainty comes from the normalization of the background contribution, which at the energies of interest is dominated by the summing-pileup. We estimated that reproduction of spectra could accommodate at most a $\pm 15\%$ variation from the nominal value, which translates into uncertainties of 6% to 22%. The integral value $\sum I_{\beta\gamma}$ is affected also by the uncertainty in the integration range. The S_n value is known to better than 8 keV for all three isotopes and we estimate that the energy calibration in this region is correct to about one energy bin. This represents an additional uncertainty ranging from 11% to 15%.

Figure 3 shows the ratio $I_{\beta\gamma}/(I_{\beta\gamma}+I_{\beta n})$ in the range of energies analyzed with TAGS for all three cases. This ratio is identical to the average ratio $\langle\Gamma_\gamma/(\Gamma_\gamma+\Gamma_n)\rangle$ over all levels populated in the decay. The shaded area around the experimental value in Fig. 3 serves to indicate the sensitivity of the TAGS results to background normalization as indicated above. The average width ratio was calculated using the Hauser-Feshbach model. The results for the three spin-parity groups populated in GT decay are shown. The NLD and PSF values used in these calculations are the same as those used in the TAGS analysis. The new ingredient needed is the NTC, which is obtained from the Optical Model (OM) with the TALYS-1.4 software package [42]. OM parameters are taken from the so-called local parametrization of Ref. [43]. Neutron transmission is calculated for known final levels populated in the decay [23–25]. In order to compute the average width ratio we need to include the effect of statistical fluctuations in the individual widths [22]. We use the MC method to obtain the average of width ratios. The sampling procedure is analogous to that described in Ref. [35]. Level energies for each spin-parity are generated according to a Wigner distribution and their corresponding Γ_γ and Γ_n to individual final states are sampled from PT distributions. The total γ and neutron widths are obtained by summation over all possible final states and the ratio computed. The ratio is averaged for all levels lying within each energy bin. In order to suppress fluctuations in the calculated average, the sampling procedure is repeated between 5 and 1000 times depending on level density. Very large average enhancement factors were obtained, reaching two orders-of-magnitude when the neutron emission is dominated by the transition to a single final state.

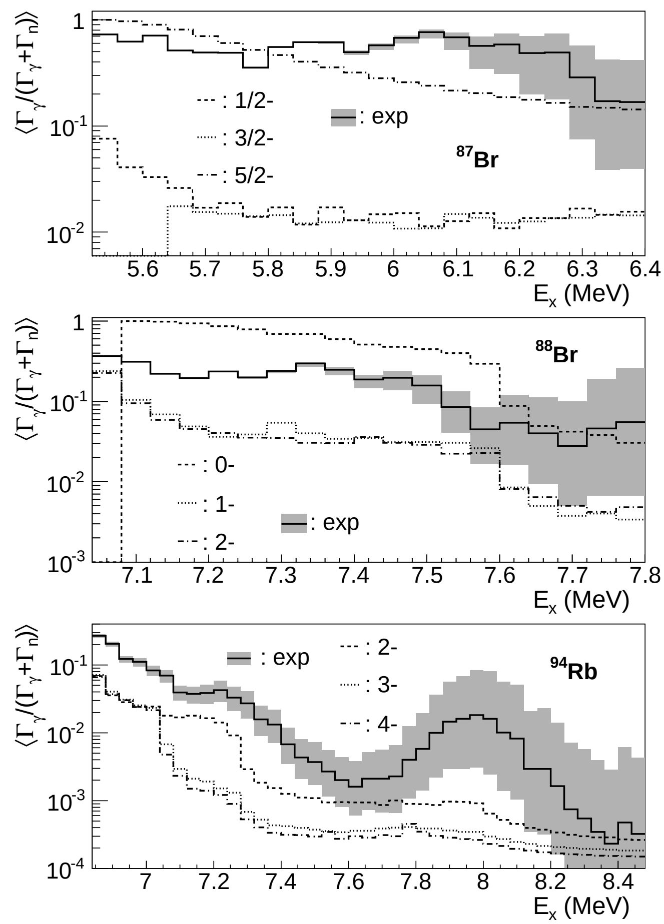

FIG. 3. Average gamma to total width ratio from experiment and calculated for the three spin-parity groups populated in allowed decays. The shaded area around the experimental value indicates the sensitivity to the background normalization (see text).

In the case of ^{87}Br $3/2^-$ decay one can see in Fig. 3 that the strong γ-ray emission above S_n can be explained as a consequence of the large hindrance of $l = 3$ neutron emission from $5/2^-$ states in ^{87}Kr to the 0^+ g.s. of ^{86}Kr, as pointed out in Ref. [1]. In the case of ^{88}Br 1^- decay a similar situation occurs for 0^- states in ^{88}Kr below the first excited state in ^{87}Kr at 532 keV, which require $l = 3$

to populate the $5/2^+$ g.s. in ^{87}Kr. For a more quantitative assessment one should know the distribution of β intensity between the three spin groups, which could be obtained from β-strength theoretical calculations. The case of ^{94}Rb 3^- decay is the most interesting. The final nucleus ^{93}Sr is five neutrons away from β stability. The γ intensity although strongly reduced, only 5% of the neutron intensity, is detectable up to 1.5 MeV beyond S_n. The structure observed in the average width ratio, is associated with the opening of βn channels to different excited states. Note that the structure is reproduced by the calculation, which confirms the energy calibration at high excitation energies. In any case the calculated average gamma-to-total ratio is well below the experiment. In order to bring the calculation to the experimental value one would need to enhance the PSF, or suppress the NTC, or any suitable combination of the two, by a very large factor. For instance we verified that a twenty-fold increase of the E1 PSF would reproduce the measurement assuming a β-intensity spin distribution proportional to $2J + 1$. An enhancement of such magnitude for neutron-rich nuclei, leading to a similar enhancement of (n, γ) cross sections, will likely have an impact on r-process abundance calculations. Therefore it will be important to investigate the magnitude of possible variations of the NTC.

In conclusion, we have confirmed the suitability of the TAGS technique to obtain accurate information on γ-ray emission from neutron unbound states and applied it to three known β-delayed neutron emitters. A surprisingly large γ-ray branching of 57% and 20% was observed for ^{87}Br and ^{88}Br respectively, which can be explained as a nuclear structure effect. In the case of ^{87}Br we observe 7 times more intensity than previously detected with high resolution γ-ray spectroscopy, which confirms the need of the TAGS technique for such studies. In the case of the more neutron-rich ^{94}Rb the measured branching is only 4.5% but still much larger than the results of Hauser-Feshbach statistical calculations, after proper correction for individual width fluctuations. The large difference between experiment and calculation can be reconciled by an enhancement of standard PSF of over one order-of-magnitude. To draw more general conclusions it will be necessary to extend this type of study to other neutron-rich β-delayed neutron emitters. Such measurements using the TAGS technique are already underway and additional ones are planned.

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