

Strong one-neutron emission from two-neutron unbound states in β decays of r -process nuclei, $^{86,87}\text{Ga}$

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β -delayed one-neutron and two-neutron branching ratios (P_{1n} and P_{2n}) have been measured in the decay of $A = 84$ to 87 Ga isotopes at the RI-beam Factory at the RIKEN Nishina Center using a high efficiency array of ^3He neutron counters (BRIKEN). Two-neutron emission was observed in the decay of $^{84,85,87}\text{Ga}$ for the first time and the branching ratios were measured to be $P_{2n} = 1.6(2)\%$, $1.3(2)\%$, and $16.2(9)_{\text{stat}}(6)_{\text{sys}}\%$, respectively. One-neutron branching ratio of ^{87}Ga ($P_{1n} = 81(9)_{\text{stat}}(8)_{\text{sys}}\%$) and half-life of $29(4)$ ms were measured for the first time. The branching ratios of ^{86}Ga were also measured to be $P_{1n} = 74(2)_{\text{stat}}(8)_{\text{sys}}\%$ and $P_{2n} = 10.2(28)_{\text{stat}}(5)_{\text{sys}}\%$ with better precision than previous study.

The observation that $P_{1n} > P_{2n}$ for both $^{86,87}\text{Ga}$ was unexpected and is interpreted as a signature of dominating one-neutron emission from the two-neutron unbound excited states in $^{86,87}\text{Ge}$. In order to interpret the experimental results, shell-model and Hauser-Feshbach statistical model calculations of delayed particle and γ -ray emission probabilities were performed. This model framework reproduces the experimental results. The shell model alone predicts P_{2n} significantly larger than P_{1n} for the ^{87}Ga decay, and it is necessary to invoke a statistical description to successfully explain the observation that $P_{1n} > P_{2n}$.

Our new results demonstrate the relevance and importance of a statistical description of neutron emission for the prediction of the decay properties of multi-neutron emitters and that it must be included in the r -process modeling.

Delayed neutron emission after β decays is found in neutron-rich nuclei where the energy window of the β^- decay (Q_β) is high enough to populate excited states of the daughter nucleus above the neutron separation energy (S_n). It was first observed in 1939 [1]. The rare process of β -delayed two-neutron ($\beta 2n$) emission was first observed in 1979 [2] in ^{11}Li . In very neutron-rich nuclei where the Q_β is larger than the two-neutron separation energy (S_{2n}), delayed multi-neutron emissions may occur.

Beta-delayed neutron emission is expected to be a prevalent decay mode for thousands of neutron-rich nuclei, many of which will be accessible in new generation radioactive ion beam facilities. Therefore, studies of this decay mode will become a major focus of their experimental program. Quantitative understanding of the neutron-emission process is required for planning of future of experimental activities aimed to provide data for nuclear structure or astrophysics. In particular with increasing decay energies and decreasing neutron separation energies when neutron-rich nuclei become available, more complex multi-neutron emission is expected to dominate their decays based on simple phase space arguments. In this work, we show new data in the boundary region where $\beta 2n$ becomes important, compare them with the nuclear models, and achieve good agreement between the experiment and predictions. We discovered, that contrary to expectations, multi-neutron emission is a significant but not the main decay mode in decays of exotic isotopes of gallium. The observed effect can be explained in the framework of statistical model [3] which assumes that particle and γ -ray emission after β decay occurs from the compound nucleus.

All of the neutron-rich nuclei on the r -process path are either one or multi-neutron β -delayed precursors. Delayed neutron emission shapes the final abundance pattern due to the changes of the isotopic population by modifying the decay-path back to stability and by contributing significantly to the neutron flux after freeze-out. However, experimental data which enable the evaluation of the role of multi-neutron emission for the r -process nuclei, are almost non-existent. The only observation of strong two-neutron emission ($P_{2n} > 1\%$) in heavier nuclei, in the region relevant to the r -process nucleosynthesis, was achieved very recently for ^{86}Ga with $P_{2n} = 20(10)\%$ by Miernik *et al.* [4]. Even with the capabilities of the new generation radioactive beam facilities, the relevant multi-neutron emitters are very difficult to measure and the r -process modeling will continue to rely on predictions by nuclear theories. Therefore, new data points are of critical importance. The predictions for the

^{86}Ga [5–9] range between 21% and 56% for P_{1n} and 7% and 44% for P_{2n} and it is difficult to judge their reliability based on a single data point, particularly when $\beta 3n$ or $\beta 4n$ decay becomes significant.

The neutron emission probability is proportional to the integrated population of states in the available energy window $Q_\beta - S_n$ and is related directly to the β -decay strength function for single neutron emitters when the competing γ decay is negligible. However, the neutron emission probability and decay strength decouple when two-neutron emission becomes energetically allowed; in such a case, competition between the $1n$ and $2n$ channels must be included. Very little is known about the role and sensitivity to nuclear structure for β -delayed neutron emitters. Often, predictions for the neutron emission probabilities are based on a simplified *cut-off* model that neglects γ -ray emission and assumes that only the higher multiplicity neutron emission prevails in the energy regions open to multiple neutron-emission channels [5–7]. In order to tackle the $1n/2n$ competition, Mumpower *et al.* [10] implemented Hauser-Feshbach (HF) statistical model of particle and γ -ray emission from compound nucleus [3] with QRPA (quasi-particle random-phase model) strength function.

This model (QRPA-HF) predicts P_{1n} or P_{2n} by following statistical decays of both the delayed- γ and neutron emission one by one until all the excitation energy is exhausted. The particle and γ emission process in the statistical model is not sensitive to the details of the nuclear structure of involved nuclei.

Substantial P_{1n} values are reported for neutron-rich $N > 50$ Ga ($Z = 31$) isotopes, 21.2(9)% in ^{82}Ga (weighted average of [11–13]), 62.8(25)% in ^{83}Ga [14], 74(14)% in ^{84}Ga [15] (superseded by 40(7)% [16] and 53(20)% [17]), 70(5)% for ^{85}Ga [18], and 60(10)% for ^{86}Ga [4]. The strong delayed-neutron emission branching ratios are due to their large β -decay energy window. A detailed neutron emission study was done by Madurga *et al.* [16] for $^{83,84}\text{Ga}$. Observation of high-energy neutrons emitted after β decay was interpreted as a signature of the shell structure effects dominating the β decay process [16]. Madurga *et al.* [16] compared existing data and calculations for half-lives and branching ratios of $^{82-87}\text{Ga}$ decays, based on the details of the β -strength distribution, but no statistical model treatment was included to make predictions for P_{xn} . Good agreement between the prediction by the shell model and experimental data was achieved for P_{1n} and half-lives by choosing a 50% quenching factor for the Gamow-Teller strength $B(\text{GT})$. This quenching was deduced from the experimental neutron spectrum and by adding a contribution from forbid-

den transitions based on experiments. The fact that the nuclear half-lives for Ga isotopes are relatively long despite the large Q_β values reflect the concentration of the B(GT) in highly excited neutron-emitting states in Ge isotopes. The model by Möller *et al.* [5, 9] uses QRPA calculations to make predictions of $P_{1n,2n}$ values and the model in principle reflects very similar shell-structure effects. The details of the model will result in a different strength distribution and delayed neutron emission probabilities. Most notably, the effects of deformation are included.

The focus of the present work is to study the delayed neutron emission for nuclei expected to be 2n precursors such as $^{84-87}\text{Ga}$. In the cases of $^{86,87}\text{Ga}$, both the shell model and QRPA predict that the majority of the B(GT) strength and resulting β -decay feeding within Q_β window is concentrated above the two-neutron separation energy. The predicted decay mode of ^{87}Ga based on the shell-model plus cut-off model results in two-neutron emission ($P_{2n}=69\%$) dominating over single-neutron emission. For the $^{84,85}\text{Ga}$ smaller but significant $P_{2n} \approx 10\%$ values are predicted by the cut-off models.

We studied neutron-rich Ga isotopes by means of β -neutron- γ spectroscopy at the RI Beam Factory (RIBF) at the RIKEN Nishina Center. Neutron-rich nuclei were produced by in-flight fission of a 345 MeV/nucleon $^{238}\text{U}^{86+}$ beam induced on a 4-mm-thick ^9Be production target. Fission fragments were separated and identified in the BigRIPS in-flight separator [19] on an event-by-event basis [20]. A total of 7×10^4 and 6×10^3 ions of ^{86}Ga and ^{87}Ga , respectively, were transported to the final focal plane for decay measurement.

The secondary ions of interest were implanted into active stoppers made of double-sided silicon-strip detectors (DSSSDs) which were capable of performing ion and β correlation measurements. AIDA (Advanced Implantation Detector Array) [21] was used in the first run while the DSSSD stack, WAS3ABi [22], and a YSO scintillator [23, 24] were employed for the second run. The typical total rate of the ion implantation in AIDA during the first run was ≈ 150 cps, while that in WAS3ABi during the second run was ≈ 60 cps.

The active stopper array was placed in the center of the high-density polyethylene moderator of the ^3He neutron counter array, BRIKEN [25]. The BRIKEN system is composed of 140 ^3He counters and two clover-type HPGe detectors [26] for high-resolution γ -ray detection. In this configuration, BRIKEN has 62(2)% neutron effective efficiency (ϵ_n) at ≈ 1 MeV of neutron energy [25].

The neutron-gated ion- β time spectra obtained in the second run are presented in Fig. 1. Neutron events are correlated with a β -decay event within a 200 μs time window after the β -ray detection. The half-lives and initial decay rates at $T_\beta - T_{ion} = 0$ for each neutron multiplicity (A_{0n}, A_{1n} , and A_{2n}) are obtained by binned maximum likelihood fitting to a convolution of contributions from

TABLE I. List of half-lives, P_{1n} , and P_{2n} obtained in this study. Q -values are adopted from [29].

nucl.	$T_{1/2}$ (ms)	branching ratio (%)		Q -values (MeV)		
		P_{1n}	P_{2n}	Q_β	$Q_{\beta 1n}$	$Q_{\beta 2n}$
^{84}Ga	97.6(12)	44(4) [†]	1.6(2) [†]	14.1(2)	8.3(2)	5.2(2)
^{85}Ga	95.3(10)	90(7) [†]	1.3(2) [†]	13.3(3)	10.2(3)	5.0(3)
^{86}Ga	49(2)	74(2) [*] (8) [†]	16.2(9) [*] (6) [†]	15.3(6)	11.0(4)	7.9(4)
^{87}Ga	29(4)	81(9) [*] (8) [†]	10.2(28) [*] (5) [†]	14.8(6)	12.1(7)	7.7(5)

^{*}statistical errors, [†]systematic errors

the decays of parent, daughter, $\beta 1n$, and $\beta 2n$ -daughter as well as a linear background, neglecting the small contribution of other descendants. The half-lives from 1n spectra are adopted since the 1n spectra have a smaller component from the decays of descendant nuclei than 0n spectra and the statistical error is smaller than in the 2n spectra, see Ref. [27, 28]. The $T_{1/2}$ and P_{xn} values obtained in this work are summarized in Table I. Because the neutron energy distribution is not known, the neutron detection efficiency makes the dominant contribution to the systematic error associated with our P_{xn} values. The P_{1n} and P_{2n} values obtained in this work for ^{86}Ga are consistent with the data by Miernik *et al.* [4]. The $T_{1/2}$ and P_{1n}, P_{2n} values of ^{87}Ga are obtained for the first time in this work. Our P_{2n} value for ^{85}Ga , 1.3(2) % is not consistent with the reported upper limit, < 0.1 % by Miernik *et al.* [18]. Our P_{2n} value is more reliable due to the fact that we have observed coincidence between two neutrons and the 247-keV γ -line from the $1/2_1^+ \rightarrow 5/2_{g.s.}^+$ transition in ^{83}Ge following the decay of ^{85}Ga . A more complete discussion of the coincidence spectroscopy results will be reported in a future publication. The P_{2n} discrepancy may be attributed to differences in detection thresholds for β particles.

Figure 2 (a) shows the comparison of the experimental neutron branching ratio with the shell model calculations with the cut-off model by Madurga *et al.* [16] and the same shell model calculations but with the Hauser-Feshback statistical model [30]. When comparing the new experimental results with the predictions by the shell model calculations, we notice a discrepancy between the cut-off model and experimental data for all the investigated $\beta 2n$ gallium precursors, most dramatically manifested in ^{87}Ga . The P_{2n} values measured here are much smaller than the cut-off model predictions.

The model we adopt to estimate the GT decay strength distribution for Ga isotopes [16] was based on a shell-model calculation using the NuShellX code [31] with hybrid interactions and the truncation described previously in [16, 32, 33]. In this model, the β -decay properties are dominated by the Gamow-Teller decay of the ^{78}Ni -core states, leaving the nucleus in the highly excited state because of the $N = 50$ shell gap. The coupling of va-

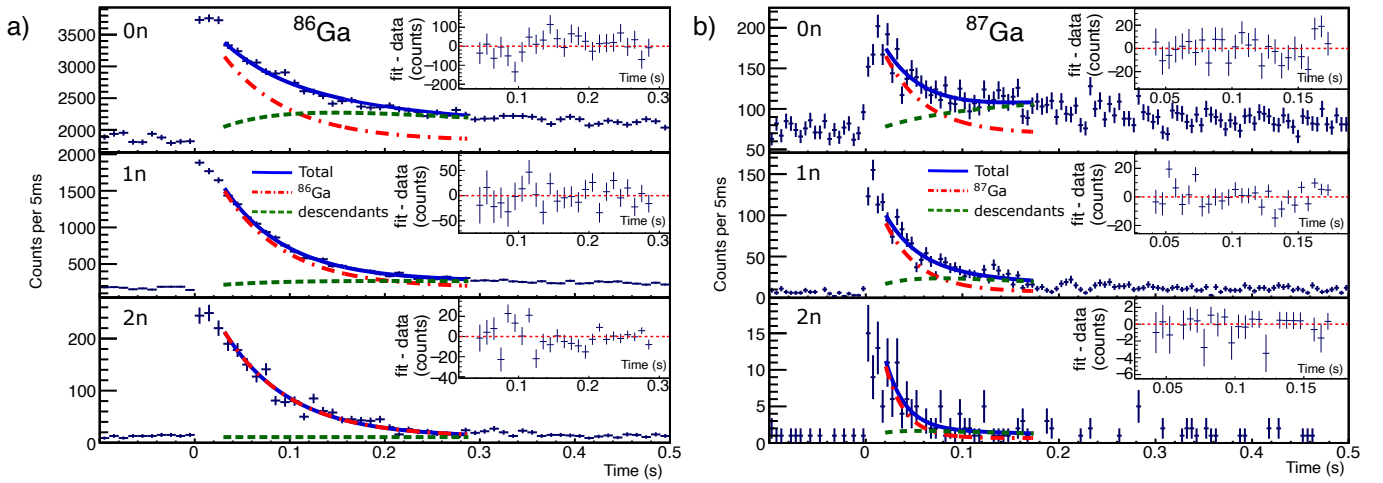


FIG. 1. The decay curves and the residuals gated by neutron multiplicity 0, 1, and 2 for (a) ^{86}Ga and (b) ^{87}Ga obtained in this work.

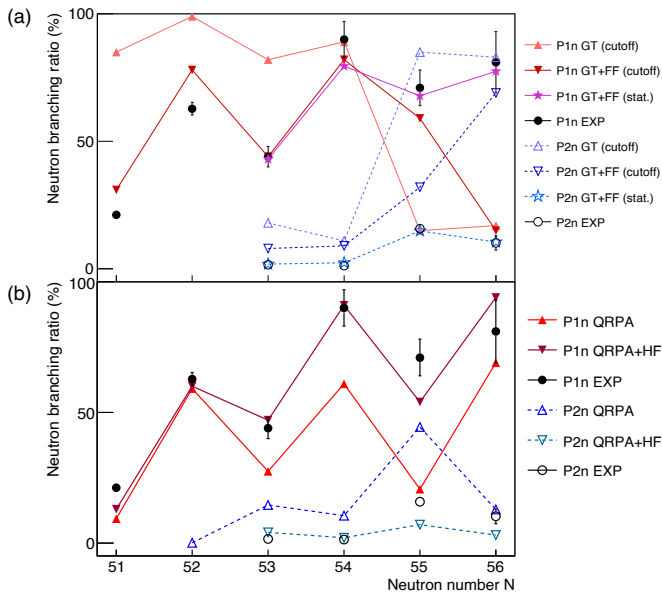


FIG. 2. Comparison of experimental P_{1n} and P_{2n} values reported for Ga precursors with calculations. (a) Comparison with shell model calculations by Madurga *et al.* [16]. “GT” and “GT+FF” in the legend show the shell model calculations with pure Gamow-Teller and GT + first forbidden transitions, respectively. Each calculation is coupled with the cut-off model (cutoff) and the statistical model (stat.). (b) Comparison with QRPA calculations [5] and those with the Hauser-Feshbach statistical model (QRPA+HF) [9]. Experimental values at $N = 53-56$ are obtained in this work. $N = 51$ and 52 are from the references shown in the text.

lence neutrons and protons to the excitations of the ^{78}Ni core produces a high density of β -populated states. For $^{86,87}\text{Ga}$ the B(GT) threshold is close to the two-neutron separation energy in the daughter. In both cases, the majority of the β decay feeds states with $E > S_{2n}$. The calculation in Madurga *et al.* [16] shows a strong odd-

even effect in the P_{1n} systematics. This apparent regularity at $^{82-85}\text{Ga}$ may break down when the two-neutron emission channel opens up.

An important element of the decay process description implemented in this framework is the contribution from the first forbidden transitions to the low excited states in Ge daughters. Despite small matrix elements, their intensities are amplified by the large phase space factor and result in a significant population of the neutron-bound states decreasing the P_{1n} as can be observed in Fig. 2 (a) up to ^{85}Ga . The Q_β dependence further enhances the observed odd-even effect.

The inherent uncertainties of the B(GT) strength calculations as well as decay energies and neutron separation energies are expected to be strongly coupled with half-lives and P_{xn} . In order to investigate the consequences of B(GT) strength uncertainty, we varied the relative position of the B(GT) distribution to match the experimental data on $T_{1/2}$ and P_{xn} . We assumed 50% quenching of the strength as in Ref. [16]. The FF contribution is constrained by the P_{0n} and half-lives.

Q_β and neutron separation energies are taken from the recent mass evaluations [29]. This procedure allows us to determine the best parameters for each isotope to describe P_{0n} and $T_{1/2}$, but as shown in Fig. 3, the cut-off model is not able to reproduce the experimental P_{1n} and P_{2n} . The same scheme to vary the strength distribution was repeated including statistical particle evaporation and γ emission [10, 30]. The results are plotted in the right panel of Fig. 3. In this case for the $A = 84-87$ isotopes of gallium, we find very good agreement with experiment without major modification of the B(GT) distribution positions (by only -0.5 MeV) and the adjusted value of the FF contribution: a factor 0.2 for ^{86}Ga and 1.0 for ^{87}Ga compared to that of ^{83}Ga and ^{84}Ga . We consider these empirical adjustments to be within the model

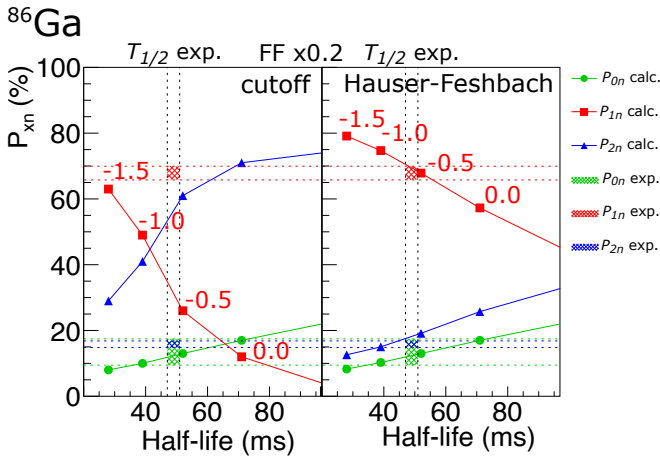


FIG. 3. Shell model predictions for P_{xn} versus $T_{1/2}$ for cutoff and statistical model for the decay of ^{86}Ga . P_{xn} values are plotted as a function of shifting the B(GT) distribution from -1.5 to 0.0 MeV and fixed FF contribution. The horizontal and vertical dashed lines show the range of the experimental $T_{1/2}$ and P_{xn} values with errors. For the experimental values of ^{86}Ga , overlaps between Miernik *et al.* [4] are plotted. The calculated P_{xn} with B(GT) shift up to +1.5 MeV are even further away from the experimental values both for the cutoff and the statistical model. The plot demonstrates that even if there were uncertainties in the shell model as large as ± 1.5 MeV, it is impossible to reproduce the experimental data within the cutoff model. In contrast, when using a statistical model, calculated P_{xn} values stay around the experimental values and are relatively insensitive to the amount of B(GT) shift.

uncertainties.

In contrast to the cut-off model, the inclusion of the statistical model correctly reproduces the dominating role of one-neutron emission from two-neutron unbound states. The same conclusion, on the necessity of adding statistical model can be drawn from QRPA model, see Figure 2 (b), which compares the predictions for P_{1n} and P_{2n} with [5] and without [9] HF. Here, however a very strong odd-even effect is due to combined effects of deformation [34] or forbidden transitions which persists in ^{86}Ga and ^{87}Ga and results in worse agreement between data and the prediction.

This result is the first demonstrated case in medium/heavy nuclei where the effects of statistical emission must be considered in order to model β -delayed multi-neutron emission. We have also examined the influence of the inclusion of the statistical model on the isotopic distribution of the r -process abundances. As pointed out by Mumpower *et al.* in their theoretical evaluation [10], this is particularly important for the r -process modeling in scenarios where the majority of the nuclei are β -delayed multi-neutron emitters, such as in the recently discovered neutron star merger [35]. We have modeled the final isotopic distribution resulting from the decay of a single r -process isotope following up every pos-

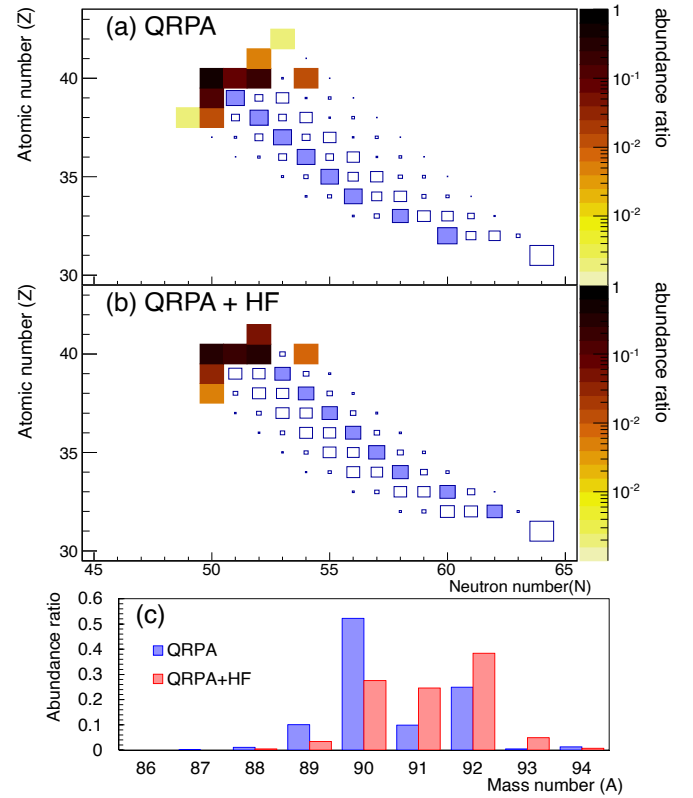


FIG. 4. Decay paths and final abundances of ^{95}Ga nuclei simulated from P_{xn} and $T_{1/2}$ values by (a) QRPA and (b) QRPA+HF calculation. The area of the blue boxes is proportional to its maximum population during the decay process. The filled boxes show the most abundant nuclei in each isotope. Mass (A) projection of the final abundances (c). The abundances are plotted as ratios relative to the initial number of ^{95}Ga nuclei.

sible decay path on the way to stability. As an example, we have chosen ^{95}Ga , which was identified to be one of the most exotic abundantly populated gallium isotopes at the freezeout [36] in the low-entropy scenario. The results are shown in Figure 4, where all the isotopes populated in the decay of ^{95}Ga are drawn, the lifetimes and branching ratios from both Möller QRPA models were used [5, 9]. Among $N > 50$ isotopes populated in the chain of ^{95}Ga decay, more than half of them are populated in the first second after formation of ^{95}Ga and all of them are 1n, 2n or 3n emitters. Their respective P_{xn} determine the final isotopic distribution. The inclusion of HF increases the population of heavier zirconiums significantly.

In summary, we discovered new β -delayed two-neutron emitters $^{84,85,87}\text{Ga}$, and measured their two-neutron branching ratio for the first time. For ^{87}Ga , its P_{1n} and $T_{1/2}$ values are measured also for the first time. The P_{1n} and $T_{1/2}$ of $^{84,85,86}\text{Ga}$ are measured with better precision than previous studies. In all of the nuclei, the shell model and QRPA calculations could reproduce the experimental neutron branching ratios and the half-lives only

if the statistical model is incorporated. The conventional cut-off model cannot describe the experimental data using previously established model parameters. The results show that the measurements of β -delayed neutron emission branching ratios cannot be used in a straightforward way to deduce the strength distribution, but a model of competing particle and γ -ray emission must be included. These results suggest that decays via one neutron emission dominate even from states which are two-neutron unbound and that it is critical to consider the competition between one- and two-neutron emission in β -delayed neutron emission models, which is of particular importance for r -process modeling. We have demonstrated the sensitivity of the final isotopic distribution to the inclusion of the statistical model. The statistical model approach, which is insensitive to the details of the nuclear structure provides a simple prescription of the β -neutron modeling. Further studies are needed to prove if it is universal.

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