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New half-life measurements of the most neutron rich arsenic and germanium isotopes

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The β-decay properties of several neutron-rich germanium and arsenic isotopes were measured at the Holifield Radioactive Ion Beam Facility at Oak Ridge National Laboratory. The measurements of almost pure radioactive sources were enabled by the combination of ion-source chemistry and two-stage mass separation. The half-life of ^{86}Ge (226±21 ms) was determined for the first time, while those of ^{84,85}Ge and $^{84-87}$As were remeasured. The results are compared to theoretical predictions of gross theory of β decay, of the finite-range droplet model and of new calculations using the energy density functional DF3a with continuum QRPA (CQRPA). We confirm the robustness and good predictive power of the latter model for nuclei near closed shells. These DF3a+CQRPA calculations were used recently to analyze r-process isobaric abundances.

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The knowledge of gross properties, like mass, half-life and β-delayed neutron probability, of very neutron-rich nuclei is mandatory for an analysis of processes occurring in neutron-rich environments like nuclear reactors and the rapid-neutron-capture (r-) process. In particular, the r-process, which follows explosive stellar events, involves multiple captures of neutrons on iron; these captures continue until β decay results in the change of atomic number (element) allowing more neutron capture to occur. It is responsible for creation of about half of the nuclei heavier than iron [1–3]. The astrophysical site for the r-process has still to be determined. Nevertheless, it is certain that the r-process path includes neutron capture and β-decay of nuclei very far from stability. Since it is not possible to measure such properties in the laboratory for the thousands of nuclei involved in the process, most of which are still unknown or at present too exotic to reach experimentally, reliable theoretical predictions need to be employed. Removing nuclear physics uncertainties from the r-process model, will allow to better discriminate between various models which involve different physical conditions.

Global models have been developed over the last years to provide such information on nuclei with very large N/Z ratios ($\gtrsim$1.8-2.5 in the vicinity of ^{78}Ni). One of the earliest theoretical approaches to β-decay properties employed was the gross theory of β decay. Often used is the finite-range droplet model (FRDM) with the addition of quasi-particle random-phase approximation (QRPA) for the Gamow-Teller (GT) part with an empirical spreading for the quasi-particle strength and the so-called gross theory to describe the first forbidden (ff) transitions [4] (from here on called FRDM+QRPA). More recently a microscopic model was developed to self-consistently calculate ground-state properties, GT and ff transitions of neutron-rich nuclei, based on the energy density-functional + continuum QRPA (CQRPA) approximation [5]. Within the latter framework, the use of the very recent DF3a energy-density functional [6] optimized for neutron-rich isotopes in the vicinity of N=50 and N=82, produced reliable predictions for neutron-rich zinc and gallium isotopes [7]. The same calculation also showed a stabilization of the gallium isotopes half-life for N$\geq$56. Calculations of the r-process using its predictions showed significant variation in the predicted isobaric abundances strengthening the yield at A>140 [7]. Testing the prediction capabilities with as neutron-rich nuclei as possible is of key importance for their verification, and also to trigger eventual modifications and improvements.

The first decay property of an exotic nucleus that is experimentally accessible is its β half-life: it can be determined from a handful of ions. Hence, measuring the half-life can provide the first test of the predictions from these global models.

This work is the continuation of our efforts reported in [7], extended to the very neutron-rich isotopes $^{84-86}$Ge and $^{84-87}$As. We report here the results of the half-life measurement. The β-decay properties of $^{84-86}$Ge, their daughter nuclei and ^{87}As will be presented in separate publications.

The measurements were performed at the Holifield Radioactive Ion Beam Facility at Oak Ridge National Laboratory (Oak Ridge, TN, USA) [8]. Almost pure radioactive beams of $^{84-86}$Ge and ^{87}As were obtained by combin-

ing two-stage electromagnetic separation and ion-source chemistry. A proton beam from the ORIC cyclotron impinged on an $^{238}UC_x$ target placed inside the IRIS-2 ion source. Fission fragments were then ionized to charge state +1. In order to suppress isobaric contaminants H_2S gas was added to the ion source: the ^{A}Ge fragments diffusing out of the target material formed $^{A}GeS^{+}$ molecules [9]. By setting the mass pre-separator to mass A+32, the mass A contaminants could be highly suppressed. The ions went then through a charge-exchange cell [8], which broke the $^{A}GeS^{+}$ molecule, leaving $^{A}Ge^{-}$ ions. The latter were sent through the second stage of electromagnetic separation consisting of a separator optimized for mass A ions, suppressing the mass A+32 contaminants (e.g., silver isotopes abundantly produced in fission). The same technique was applied for ^{87}As.

The purified beams were then sent to the Low Energy Radioactive Ion Beam Spectroscopy Station (LeRIBSS). The 200 keV ions were implanted onto the Moving Tape Collector (MTC), which periodically moved the activity away from the measuring spot [8]. The implantation point was surrounded by two plastic scintillation counters for detecting β particles and four clover high-purity germanium detectors in close geometry for γ-ray spectroscopy. The photopeak efficiency for the clover array amounted to 6% at 1.3 MeV and 32% at 100 keV. The signals from all the detectors were read out by a digital-electronics-based data acquisition system [10, 11].

The β and γ radiation emitted in the decay of the nuclei of interest was measured during the implantation of the activity on the tape (beam-on or grow-in) and while the beam was deflected away (beam-off or decay). The implanted activity was then transported away for 0.36 s and a new cycle began. The time distribution of the β-gated decay radiation measured within such a cycle, background subtracted, was used to determine the half-lives by fitting the appropriate solutions of the Bateman equations for the growth and decay patterns of γ transitions belonging to the β-γ and β-n-γ decay channels of interest. Two different solutions of the Bateman equations [12] were used in order to determine the half-lives of $^{84-86}$Ge and of $^{84-87}$As:

$$f(t) = \begin{cases} n\left(1 - e^{-\lambda \cdot t}\right) & \text{for } t < T_0 \\ \\ n\left(1 - e^{-\lambda \cdot t}\right) e^{-\lambda \cdot (t - T_0)} & \text{for } t \geq T_0, \end{cases} \quad (1)$$

where n and λ are fitting parameters corresponding to the activity and the decay constant, respectively, and T_0 is the time at which the beam is deflected away;

$$f(t) = \begin{cases} (n_1 + n_2)\left(1 - e^{-\lambda_2 \cdot t}\right) \\ \quad + n_1 \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(e^{-\lambda_2 \cdot t} - e^{-\lambda_1 \cdot t}\right) \\ \qquad\qquad\qquad\qquad \textit{for } t < T_0 \\ \\ n_1 \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-\lambda_1 \cdot T_0}\right) e^{-\lambda_1 \cdot (t - T_0)} \\ \quad + \left(n_1 + n_2 - n_1 \frac{\lambda_2}{\lambda_2 - \lambda_1}\right)\left(1 - e^{-\lambda_2 \cdot T_0}\right) e^{-\lambda_2 \cdot (t - T_0)} \\ \qquad\qquad\qquad\qquad \textit{for } t \geq T_0, \end{cases} \quad (2)$$

where n_1 and n_2 are fitting parameters corresponding to the activity for germanium and arsenic isotopes, respectively, λ_2 is the fitting parameter corresponding to the arsenic decay-constant, λ_1 is the decay constant of the germanium precursor, fixed to the value determined in this work, and T_0 is the time at which the beam is deflected away.

Equation 1 was used for $^{84-86}$Ge and ^{87}As, since they can be considered as the precursors in the decay chain. Equation 2 was used for $^{84-86}$As, where the $^{84-86}$As precursor is present both in the beam as a mass contaminant and is produced in the decay of the respective $^{84-86}$Ge mother nucleus.

The results of the time analysis of all the nuclei investigated are summarized in Table I: the results of the fits of the growth and decay pattern, background subtracted, for each γ-transition considered are given, as well as the adopted value for the half-life as obtained from the weighted average of the best fits of each transition assigned to that isotope's decay. The cycle used in each case is also given. In Figure 1 growth and decay time distributions, gated on γ transitions, are plotted for the cases of ^{86}Ge and ^{86,87}As and the best fit is superimposed.

Most of the transitions used to infer half-life values to

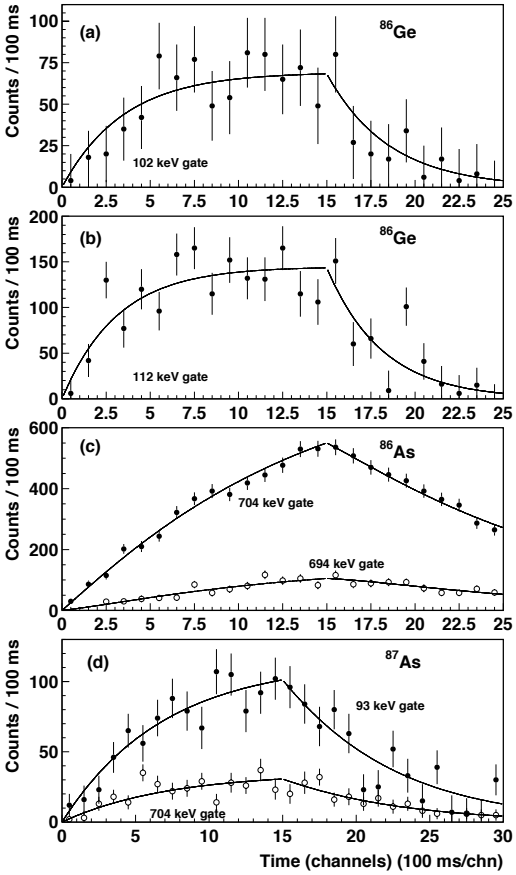

FIG. 1. Growth and decay pattern gated on γ-ray transitions, with the respective fit result, for: (a) and (b) ^{86}Ge; (c) ^{86}As and (d) ^{87}As. The transitions are given next to the respective fit result. See text and Table I for details.

TABLE I. Half-lives of $^{84-86}$Ge and $^{84-87}$As. The values of the half-life obtained for each transition considered are given, together with the energy of the γ-line and its decay branch. For each isotope studied the adopted value of the half-life is the weighted average of the corresponding γ-transitions and is also given in the table, as well as the grow-in/decay cycles used.

mass	cycle (growth/decay)	isotope	γ-transition keV	decay channel	$T_{1/2}$ from this work	$T_{1/2}$ from literature
84	3.0 s/3.0 s	^{84}Ge	100	β-γ	936±22 ms	
			243	β-γ	962±27 ms	
			347	β-γ	858±72 ms	
				weighted average:	942±17 ms	954±14 ms [13]
		^{84}As	667	β-γ	3.08±0.78 s	
			1455	β-γ	3.27±0.87 s	
				weighted average:	3.16±0.58 s	4.2±0.5 s [13]
85	1.5 s/1.5 s	^{85}Ge	100 and 102 [a]	β-n-γ and β-γ	484±9 ms	
			116	β-γ	526±22 ms	
			267 [b]	β-γ	549±24 ms	
			395 [b]	β-γ	444±37 ms	
				weighted average:	494±8 ms	535±47 ms [14]
	10.0 s/10.0 s	^{85}As	1115	β-γ	2.08±0.14 s	2.032±0.012 s [14]
86	1.5 s/1.0 s	^{86}Ge	102	β-n-γ	217±28 ms	
			112 [b]	β-γ	242±56 ms	
			116 and 119 [a b]	β-n-γ and β-γ	235±39	
				weighted average:	226±21 ms	>150 ns [15]
		^{86}As	694	β-γ	743±216 ms	
			704	β-γ	873±67 ms	
				weighted average:	861±64 ms	945±8 ms [16]
87	1.5 s/1.5 s	^{87}As	92 [b]	β-γ	478±44 ms	
			704	β-n-γ	495±60 ms	
				weighted average:	484±35 ms	560±80 ms [17]
						$1450(550)^{+3900}_{-1100}$ ms[c] [18]

[a] the two transitions cannot be fully resolved and the growth and decay time fit was done by gating simultaneously on both transitions.
[b] see text for details on the assignment of the γ transition.
[c] value based on maximum likelihood fit of 12 events; the first and second numbers following the half-life value represent the systematic and statistical uncertainties, respectively.

the isotopes of interest are known from literature. As far as ^{86}Ge decay is concerned, no information was available on this nucleus prior to this experiment, except its existence with a half-life limit longer than 150 ns [15] inferred from the observation of these nuclei at the final focus of a fragment separator. The ^{86}Ge half-life value was determined here for the first time. For this purpose four γ-transitions were considered: the known β-n-γ transition at 102.0 keV [19], the new γ-transition at 111.7 keV and the doublet at 116 and 119 keV composed of the known β-n-γ 116.3 keV line [19] and the new 118.9 keV γ-transition, see Figure 2. The 112 and 119 keV lines were assigned to the β-γ decay of ^{86}Ge on the basis of the following considerations: only isobarically-separated A=86 activity was deposited on the tape at the measuring point; the two transitions are in coincidence with each other and no similar-energy γ-transitions have been reported to date in A=86 or A=85 (β-n decay chain) isobars and, last but not least, their time distribution is not compatible with the longer lifetimes of the less exotic A=86 and A=85 isotopes. We adopt the weighted average of 226±21 ms as the ^{86}Ge half-life, see Table I.

In the case of the β decay of ^{87}As no β-γ information was available prior to this work. Only one β-n-γ transition at 704 keV was assigned to its decay [16]. Another γ-transition at 92 keV was observed here and assigned to the β-γ decay branch of ^{87}As. Its assignment to the β-γ decay of ^{87}As is based on the fact that only isobarically separated A=87 activity was deposited on the tape at the measuring point; its assignment to ^{87}Ge decay is highly unlikely because of the much lower production cross-section; no similar γ-transitions have been reported

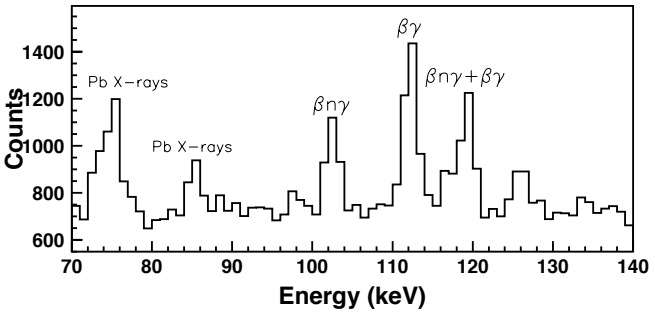

FIG. 2. Portion of the $\beta\gamma$ spectrum of ^{86}Ge showing the γ transitions used to determine its half-life. The peaks of interest are labelled according to the decay channel. See text for details.

in A=87 or A=86 (β-n decay chain) isobars and its time distribution is not compatible with the longer lifetimes of the less exotic A=87 and A=86 isobars. We adopt the weighted average of 484±35 ms as the ^{87}As half-life. This value, which is based on characteristic γ-line decay pattern, is compatible within error bars with the two results known from literature based on β and/or neutron measurements [18, 20], but is of better precision. As far as ^{85}Ge is concerned, little information was known on its decay before this work. In order to determine its half-life, we considered transitions known from previous work [19] at 100 keV (β-n-γ), 102 and 116 keV (β-γ) and the new transitions at 267 and 395 keV identified in this work and assigned to the β-decay of ^{85}Ge. These two transitions were assigned on the basis of similar considerations to those done above for ^{87}As and ^{86}Ge with the additional support of β-γ-γ coincidences.

In all other cases (^{84}Ge, $^{84-86}$As), known β-γ or β-n-γ transitions were used. In addition for ^{86}As the new γ transition at 694 keV was used. Its assignment to the β-decay of ^{86}As is based on β-γ-γ coincidences. In the case of ^{84,86}As the large uncertainty on the half-life is due to the fact that the MTC cycle was optimized for the shorter lifetime of their precursors.

In Figure 3 the half-lives of germanium and arsenic isotopes are plotted as a function of the respective mass. The new experimental values obtained in this work agree with available literature values [13, 14, 16, 17], and agree with the theoretical predictions from the FRDM+QRPA [4] and the DF3a+CQRPA [5–7] models, while the gross theory of β decay [21] systematically underestimates the half-life by large factors. The DF3a+CQRPA-calculated half-lives show very good agreement with the experimental values for the germanium isotopes and provide a robust prediction for the new half-life of ^{86}Ge, while the FRDM+QRPA predictions are systematically larger than the measured values. As already observed for the gallium isotopes [7], DF3a+CQRPA predicted half-lives stabilize for large neutron excesses (A≥88, N≥56 for Z=31 gallium and A≥86, N≥54 for Z=32 germanium isotopes) and become systematically longer than the respective FRDM+QRPA predictions.

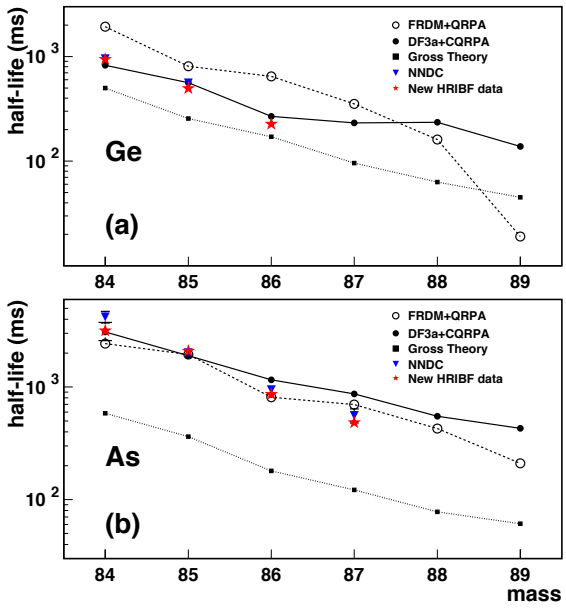

FIG. 3. (Color online) Half-lives for (a) germanium and (b) arsenic isotopes. Theoretical predictions from the FRDM+QRPA calculations [4], the DF3a+CQRPA [5, 6] and gross theory [21] are compared with existing literature data and the new results from this experiment. Experimental data are plotted with error bars, which, when not visible, are smaller than the marker symbol used. See text for details.

As far as arsenic isotopes are concerned, the DF3a+CQRPA predictions depart slightly from experimental values, especially for the higher masses (A=86,87). The FRDM+QRPA provides better agreement with the experiment. The trend of longer half-lives predicted by the DF3a+CQRPA with respect to the FRDM+QRPA model for zinc, gallium and germanium isotopes is respected for N≥54 arsenic isotopes. For ^{85}As, both models show perfect agreement with the experimental result, while that of ^{84}As shows good agreement between our new more precise half-life and the DF3a+CQRPA.

The DF3a+CQRPA proved to give very good estimates for the half-lives of very neutron-rich germanium isotopes. Its predictions seem to depart for the ^{86}As and ^{87}As isotopes, where the FRDM+QRPA provides, in average, a better estimate. Both predictions are similar for the more neutron-rich ^{88}As and depart for ^{89}As. Experiments are needed to verify further the prediction power of these models. A bit worse agreement between the DF3a+CQRPA and experiment for ^{86}As and ^{87}As might be related to the onset of collective effects for nuclei departing from Z=28 and in particular N=50 shell closures.

In summary, we have measured the β-decay properties of several very neutron-rich germanium and arsenic isotopes, providing the first information on ^{86}Ge (half-life 226±21 ms). For this purpose we exploited the chemical selection technique at the IRIS-2 ion source at the HRIBF in combination with two-stage mass separation, which provided almost-pure isotopically separated samples at the measuring station. The half-life values for $^{84-86}$Ge and $^{84-87}$As are generally in a good agreement with the previous values from literature, with a more precise value measured in this work for ^{85}Ge and ^{87}As decay, respectively. We have compared our results to theoretical predictions from three models, the gross theory for β decay, the FRDM+QRPA and the DF3a+CQRPA. While gross theory systematically overestimates the β-decay rate by about an order of magnitude, QRPA-based models provide a better agreement between predictions and measured half-lived. Among these latter models, the DF3a+CQRPA gives the best estimates for the half-lives, in particular for zinc, gallium [7] and germanium isotopes, and it is less accurate for arsenic isotopes. This suggests that further improvements need to be implemented, as well as the development of the next-generation models, in order to have the same quality predictions for nuclei close to the N=50 shell closure and for those further from magic gaps. The half-lives predicted by the DF3a+CQRPA model for the most neutron-rich germanium isotopes (including ^{86}Ge) were used to predict the r-process abundances [7]. The new half-life value of ^{86}Ge provides additional validation of these calculations.

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