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The standard  $\gamma$ -ray energy calibration source  $^{152}\mathrm{Eu}$  is well known based on the 13.5 year decay of its ground state. However, in addition to this decay  $^{152}\mathrm{Eu}$  also has two relatively long lived isomeric states: a 9 hour J<sup> $\pi$ </sup>=0<sup>-</sup> state at  $E^*$  = 46 keV and a 96 minute J<sup> $\pi$ </sup>=8<sup>-</sup> state at  $E^*$  = 148 keV. Here we report a new measurement of the half-lives of both of these isomeric states. Excited states in  $^{152}\mathrm{Eu}$  were populated following the  $^{154}\mathrm{Sm}(\mathrm{p},3\mathrm{n})$  reaction using a 25 MeV proton beam from the K-150 cyclotron at the Cyclotron Institute of Texas A&M University. Post irradiation,  $\gamma$  rays from the de-excitation of the long lived isomeric states were measured using the six BGO shielded high purity germanium (HPGe) clover detectors that are part of the STARLiTeR array. The half-life of the  $J^{\pi}$  = 8<sup>-</sup> isomer  $^{152m_2}\mathrm{Eu}$  was obtained by measuring the decrease in intensity of the 90 keV  $\gamma$  ray from the cascade to the ground state. The half-life of this state was measured to be 95.8(4) minutes which is in agreement with and significantly more precise than the previously measured value of 96(1) minutes. In a manner similar to the ground state the second long lived isomer  $^{151m_1}\mathrm{Eu}$ , the J<sup> $\pi$ </sup>=0<sup>-</sup> state at 46 keV,  $\beta$ -decays to excited states in  $^{152}\mathrm{Gd}$  and  $^{152}\mathrm{Sm}$ . The half-life of this state was measured to be 9.39(7) hours using five  $\gamma$ -ray transitions.

# I. INTRODUCTION

Nuclear isomers, metastable states in atomic nuclei with lifetimes significantly longer than the typical prompt decay, occur throughout the chart of the nuclei [1]. The predominant decay mode is via  $\gamma$  decay although particle emission mediated either by the strong force ( $\alpha$ -decay), weak force ( $\beta$ -decay), or fission is also possible and observed. Isomeric lifetimes range from a few ns to  $10^{15}$  years for  $^{180m}$ Ta, longer than the accepted lifetime of the universe. The lifetimes of these excited states probe the detailed wave functions of the underlying nuclear structure. Many isomers can be characterized as shell model states where the valence protons and neutrons couple to a higher spin state than other states at similar or lower excitation energies. The isomer decay then requires the emission of a  $\gamma$  ray typically of low energy that removes considerable angular momentum, resulting in long half-lives. Doubly odd <sup>152</sup>Eu lying in the heart of a region of the chart of the nuclei where the equilibrium shape is rapidly changing from spherical/vibrational to deformed rotational character has a relatively large number of long lived states with half-lives greater than 100 ns [2] including two known isomers with half-lives of the order of hours: a  $J^{\pi}=0^{-}$  state at 46 keV with a half-life of 9 hours and a  $J^{\pi}=8^-$  state at 148 keV with a half-life of 96 minutes. The 46 keV isomer is significantly populated in the astrophysical s-process via the

 $^{151}\mathrm{Eu}(n,\gamma)$  reaction [3]. This could be important if thermal equilibrium is not achieved with the ground state, and with respect to this process the isomeric half-lives may be of some interest.

The isomer  $^{152m_2}$ Eu, the 8<sup>-</sup> level at 148 keV, decays via a 40 keV stretched E3 transition to a lower lying 5<sup>+</sup> level and thence via a cascade of  $\gamma$ -ray transitions to the 3<sup>-</sup> ground state, as shown in Figure 1. The relatively short half-lives of the 5<sup>+</sup> and 4<sup>+</sup> states involved in this cascade, <20 ns and 384 (10) ns respectively [2, 4], allow the 90 kev transition to be used to measure the lifetime of  $^{152_{m2}}$ Eu. The first measurement of this cascade dates from a 1963 article by Kirkby et al. [5] who identified a 92(2) keV  $\gamma$  ray with a half-life of 96(5) minutes. Numerous further measurements, for example [4, 6, 7], have identified this transition as belonging to the cascade of  $\gamma$ rays that originates from the decay of the  $8^-$  isomer, specifically from a transition between the 4<sup>+</sup> state at 90 keV and the 3<sup>-</sup> ground state, as shown in Figure 1. The most recent measurement of the half-life of  $^{152m_2}$ Eu was performed in 1974 by Pruys et al. [8] in which it was found to be 96(1) minutes.

The other long lived isomer  $^{152m_1}$ Eu, the  $0^-$  level at 45.6 keV,  $\beta$ -decays to a variety of excited states in  $^{152}$ Sm and  $^{152}$ Gd in a manner similar to the ground state. Its half-life has previously been measured five times, most recently in 1990 by Abzouzi et~al.~[9] who obtained a very precise value of 9.3116(13) hours. However this disagrees by 13 standard deviations from the previous, also very precise, measurement of Lagoutine et~al.~[10] who obtained 9.274(3) hours, suggesting that for at least one of these measurements the values of the errors are under-

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estimated.

The transition between  $^{152m_1}$ Eu and the ground state of  $^{152}$ Eu has a hindrance factor of  $\geq 5 \cdot 10^7$  relative to the Weisskopf estimate [2]. This has been explained by considering the Nilsson configurations of the proton and neutron orbitals; the ground state has been assigned main wavefunction components of  $\pi 5/2^+$  [413  $\downarrow$ ]  $\nu 11/2^-$ [505  $\uparrow$ ] whereas the 0<sup>-</sup> isomer has been assigned  $\pi 3/2^+$ [411  $\downarrow$ ]  $\nu 3/2^-$ [532  $\downarrow$ ] [2]. Therefore, for the decay to occur both the neutron and proton must change orbits and the neutron transition is  $\Omega$  forbidden.

In this brief report we present new measurements of the half-lives of both of these isomeric states.

### II. EXPERIMENTAL ARRANGEMENT

The experiment was conducted at the Cyclotron Institute of Texas A&M University using the STARLiTeR array. STARLiTeR consists of STARS (Silicon Telescope Array for Reaction Studies), a highly segmented  $\Delta E-E$ charged particle telescope, a detailed description of which can be found in reference [11], and LiTeR (the Livermore Texas Richmond array), a Compton suppressed HPGe clover detector array for  $\gamma$ -ray detection. In the present work LiTeR consisted of six BGO shielded HPGe clover detectors positioned approximately 13 cm from the target at angles of 47°, 90° and 133° with respect to the incident beam axis. The clover detectors were calibrated using the standard  $\gamma\text{-ray}$  calibration sources  $^{22}\mathrm{Na},\,^{54}\mathrm{Mn},\,^{57}\mathrm{Co},\,^{60}\mathrm{Co},\,^{109}\mathrm{Cd},\,^{133}\mathrm{Ba},\,^{137}\mathrm{Cs}$  and  $^{152}\mathrm{Eu}.$  A final energy resolution of 2.6 keV and 3.5 keV (FWHM) was obtained at 122 keV and 963 keV, respectively. The absolute photopeak efficiency of the array, with addback, was measured to be 4.8% at 103 keV.

A  $^{154}$ Sm target of thickness  $\simeq 1$  mg cm $^{-2}$  and 99% enrichment was bombarded by a 25 MeV proton beam from the K-150 cyclotron with an average beam current of 1.2 nA for approximately 8 hours immediately prior to the activation measurement. Unstable europium isotopes were produced via the  $^{154}$ Sm(p, xn) reactions. Following this irradiation  $\gamma$  rays from the activated target were measured over a continuous period of 38 hours. The energy and time for each  $\gamma$ -ray event was recorded for subsequent offline analysis. The time information was obtained from a 40 MHz clock pulse, digitized using a 64 bit TDC and used to generate  $\gamma$ -ray decay curves.

### III. RESULTS

In Figure 2(a) the low energy region of the  $\gamma$ -ray spectrum obtained during the entire activation measurement is shown. The spectrum obtained during the first hour of the measurement is shown in Figure 2(b). The intense peak at 90 keV is identified as the 90 keV  $\gamma$  ray from the decay of  $^{152m_2}$ Eu shown in Figure 1. This is the only

 $\gamma$  ray from this cascade observed in the current experiment. The 18 keV  $\gamma$  ray lay below the energy threshold of the experiment while the 40 keV transition is highly converted and was therefore also not observed. The 13 keV and 77 keV  $\gamma$  rays are too weak to be observed. Figure 2(c) shows the long lived background remaining after 38 hours. A  $\gamma$ -ray energy-time matrix is shown in Figure 2(d) in which the long lifetimes of the two transitions bordering the 90 keV peak is apparent.

The decay rate of the background in the vicinity of the 90 kV peak has a strong energy dependence caused by Compton scattering of multiple high energy gamma rays originating from relatively short lived states. Detailed studies have shown that this can affect the extracted half-life for the 90 keV transition if background regions are chosen even a few keV away from the peak of interest, for example the potential background regions at approximately 110 keV and 125 keV that can be seen in Figure 2(a). Therefore, two 4 keV wide background regions were chosen on both sides of and immediately adjacent to the 90 keV peak, despite the presence of two long-lived contaminant lines in this region of the spectrum. These peaks are visible in Figures 2(a), (c) and (d), but play a relatively unimportant role for short decay times as seen in Figure 2(b). The background spectrum was then appropriately weighted by area and subtracted from the decay curve produced by gating on the 90 keV peak. The resulting decay in intensity of the 90 keV  $\gamma$  rays after background subtraction is shown in Figure 3. A single exponential fit to this data yielded a half-life of 95.8(4) minutes for the decay of  $^{152m_2}$ Eu, where the quoted uncertainty is statistical, and the reduced  $\chi^2$  of the fit is 0.74.

The  $\gamma$  rays at 122 keV, 334 keV, 842 keV, 963 keV, 970 keV and 1315 keV, shown in the level scheme in Figure 1, from the decay of  $^{152m_1}$ Eu were observed. Independent measurements of the half-life of  $^{152m_1}$ Eu using four of these  $\gamma$  rays are shown in Figure 4. The  $\gamma$  rays at 970 keV and 1315 keV had insufficient statistics for a measurement. The contributions from the decay of the ground state of  $^{152}$ Eu, which has a half-life of 13.517(9) years, were negligible due to the relatively long half-life compared to the 9 hour decay of interest. The weighted mean of these measurements is 9.39(7) hours, where the quoted uncertainty is again statistical.

The inset in Figure. 4 compares the value obtained in the present work to five previous measurements. We note that there is a 13 sigma discrepancy between the two recent most precise measurements [9,10], calling into question at least one of the quoted uncertainties. In such cases the Particle Data Group [12] recommends a procedure for scaling the uncertainty. If we apply this procedure in this case the weighted average of all six measurements is 9.306(14) hours. This is to be compared to the current NNDC value of 9.3116(13) hours [13].

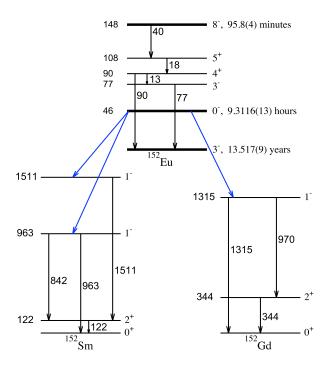


Figure 1. (Color online) Partial decay schemes of  $^{152m1}$ Eu and  $^{152m2}$ Eu.  $\gamma$ -ray energies and excitation energies are in keV.

### IV. SUMMARY AND CONCLUSIONS

In conclusion, by measuring the intensity of the 90 keV  $\gamma$  ray originating from the decay of the  $J^{\pi}=8^-$  isomer  $^{152m_2}$ Eu the half-life of this state was measured to be 95.8(4) minutes, a factor of 2.5 reduction in uncertainty compared to previous measurement of 96(1) minutes. Using the 122, 334, 842, and 963 keV  $\gamma$  rays, four independent measurements of the half-life of  $^{152m_2}$ Eu were made. The weighted mean of these was 9.39(7) hours which is in reasonable agreement with the previous two most precise literature values of 9.3116(13) hours and 9.274(3) hours, values which disagree with each other by 13 sigma. Using the procedure recommended by the Particle Data Group we find the weighted average of our value and the five previous measurements of  $^{152m_2}$ Eu to be 9.306(14) hours.

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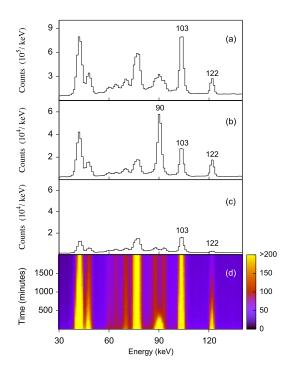


Figure 2. (Color online) Panel (a): The low energy region of the  $\gamma$ -ray spectrum obtained during the entire activation measurement. Panel (b): The  $\gamma$  rays detected during the first hour of the measurement. Panel (c): The  $\gamma$  rays remaining after 38 hours, also measured during a one hour period. Panel (d): A two dimensional plot of  $\gamma$ -ray energy against time showing the decrease in intensity of  $\gamma$  rays from the decay of relatively short lived states as well as the near constant intensity of the long-lived background peaks. The 90 keV and 122 keV  $\gamma$  rays from Figure 1 are labled. The 103 keV  $\gamma$  ray is from the 46 hour decay of  $^{153}{\rm Sm}$ , populated via the (p,d) reaction.

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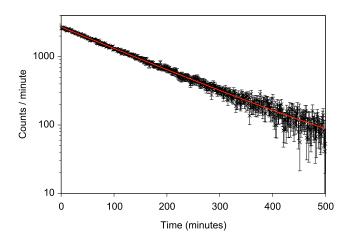


Figure 3. (Color online) The decay in intensity of the 90 keV  $\gamma$  ray which originates from the decay of  $^{152m_2}$ Eu. The red line shows an exponential fit to the data.

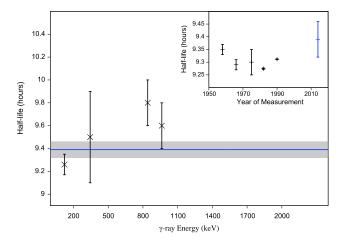


Figure 4. (Color online) Measurements of the half-life of  $^{152m1}\mathrm{Eu}$  obtained by measuring the decrease in intensity of the 122 keV, 334 keV, 842 keV and 963 keV  $\gamma$  rays listed in Figure 1. The blue line and grey band illustrate the weighted mean obtained in the present work and the associated uncertainty. Inset: Previous measurements of the half-life of  $^{152m1}\mathrm{Eu}$  [8–10, 14, 15] (black points) compared to the value obtained in the present work (blue point).

- [1] P. Walker and G. Dracoulis, Nature **399**, 35 (1999).
- [2] T. von Egidy et al., Zeitschrift für Physik A 286, 341 (1978).
- [3] H. Beer, F. Käppeler, K. Yokoi, and K. Takahashi, The Astrophysical Journal 278, 388 (1984).
- [4] K. Takahashi, M. McKeown, and G. Scharff-Goldhaber, Phys. Rev. 137, B763 (1965).
- [5] P. Kirkby and T. Kavanagh, Nuclear Physics 49, 300 (1963).
- [6] J. Reierson, G. Nelson, and E. Hatch, Nuclear Physics A 153, 109 (1970).
- [7] J. Simi, S. Koiki, and M. Radojevi-Stojanovi, Delayed coincidence measurement of 152Eu 89.8488 kev isomeric state feeding, in *Neutron Capture Gamma-Ray Spectroscopy*, edited by R. Chrien and W. Kane, pp. 751–753, Springer US, 1979.
- [8] H. Pruys, E. Hermes, and H. V. Gunten, Journal of Inorganic and Nuclear Chemistry 37, 1587 (1975).
- [9] A. Abzouzi, M. Antony, V. Ndocko Ndongué, and D. Oster, Journal of Radioanalytical and Nuclear Chemistry

- **145**, 361 (1990).
- [10] F. Lagoutine and J. Legrand, The International Journal of Applied Radiation and Isotopes 33, 711 (1982).
- [11] S. Lesher et al., Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 621, 286 (2010).
- [12] K. Nakamura, Journal of Physics G: Nuclear and Particle Physics 37, 075021 (2010).
- [13] Evaluated Nuclear Structure Data File (ENSDF) National Nuclear Data Center (NNDC), Brookhaven National Laboratory, 2014.
- [14] J. Corless, Isotopic Abundance Studies by Neutron Activation Analysis. Annual Report, April 15, 1964-April 14, 1966.
- [15] G. Gueben and J. Govaerts, Inst. Interuniv. Sci. Nucleaires (Bruxelles), Monographie No.2 (1958).