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An E5 decay from the $J^\pi = 11/2^-$ isomer in ^{137}Ba

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A new γ decay branch has been found from the well-known 661.659(3) keV $J^\pi = 11/2^-$ $T_{1/2} = 2.552(1)$ min isomer in ^{137}Ba which is populated in the β-decay of ^{137}Cs. The new 377.9(3)-keV γ ray connects the isomer to the low-lying 283.5 keV, $J^\pi = 1/2^-$ state. It is of near-pure E5 character. The decay has a γ branching ratio ($Br_\gamma = \Gamma_\gamma/\Gamma_{tot}$) of $1.12(9) \times 10^{-7}$. The new decay has a B(E5) of 0.71(6) W.u. ($B(E5)\downarrow = 6.5(6) \times 10^5 e^2 fm^{10}$), a value consistent with other "single particle" E5 decays in the region. The new decay branch is of topical interest, as it competes with the much-sought "2-photon" second-order electromagnetic decay from this state.

The second-order electromagnetic process involving the decay of quantum states by the simultaneous emission of two photons has attracted interest since the beginning of quantum electrodynamics [1–3]. By now the atomic case is quite thoroughly studied and understood, particularly for hydrogen and helium-like atoms [4, 5]. In the analogous nuclear decays, two photon emission has been studied for cases where the first order process is forbidden, such as the decay of excited $J^\pi = 0^+$ states in ^{16}O and ^{40}Ca [6]. However, two-photon decay directly competing with one-photon decay has not been definitively seen, despite several searches [7, 8]. One of the best experimental situations for studying such competition is the decay of the well-known 662-keV isomer in ^{137}Ba [9]. This is an especially interesting case as it involves a large change in angular momentum, with competing two-photon multipole modes involved. This aspect is significantly different to the atomic and $J = 0$ nuclear cases, where the two photons are dominantly both dipole. The key observable for determining the competition between multipoles is in how the energy is shared between the two photons [8].

A technical shortcoming of using a ^{137}Cs source for two-photon physics is the presence of a low-lying $J^\pi = 1/2^+$ 283.5-keV level and the possibility of a true 2-step cascade γ decay path. The 283.5 γ-ray was observed in several works [10–13] as the state is also populated via β-decay, but the 378-keV transition was not seen. Fig. 1 shows the key decay characteristics. The $11/2^-$ to $1/2^+$ transition is a small branch as it involves the decay photon carrying angular momentum of $L = 5\hbar$. This branch is expected to be $<10^{-6}$ of the normal 662-keV γ decay, so observation requires a special experiment. This transition is dominantly an E5 electric decay which allows a rare opportunity to add to the small collection of known E5 matrix elements and re-examine their systematic behavior. It is also important to quantify this "cascade" decay mode, as it is always competing with the higher-order two-photon process, so any two-photon experiment will need to take this two-step decay into account in analysis. In the course of building an experiment to investigate 2-photon decay, we have successfully identified the $11/2^-$ to $1/2^+$ transition and precisely determined its intensity. The branch turns out to be quite small, (less than 5% of the preliminary experimental measurement reported for the 2-photon decay [8]), so, in the end, most likely will reflect a small correction to any 2-photon analysis. However, it represents a clear benchmark for 2-photon experiments, with well-defined photo peaks, and so its observation establishes that a suitable level of statistics and sensitivity has been achieved, which is required for attempting the more difficult continuum physics.

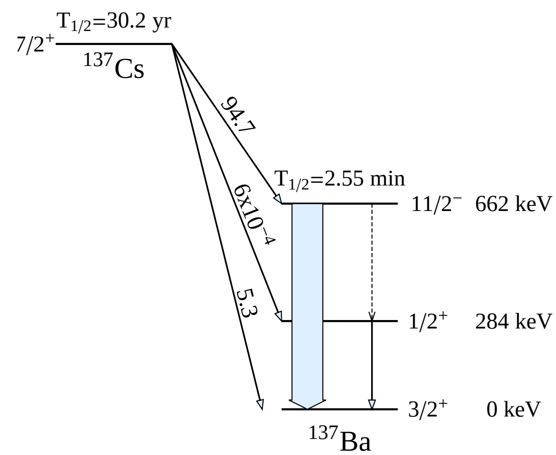

FIG. 1. (Color online)Key features of the decay of ^{137}Cs to states in ^{137}Ba. γ-ray transitions are labeled by their $\Sigma I(\gamma + ce)$ values.

Conceptually, an experiment to find this cascade branch is simple: place a source before a single γ ray detector and record data until the 378-keV peak becomes statistically significant. In practice, Compton scattering makes this approach untenable. For more sensitivity, one may deploy two γ ray counters and collect all events with either one or two detectors firing, and then measure the ratio of 662-keV photo peak to the 378/284-keV coinci-

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dences.

In practice, use of a modern large array of high purity germanium counters, like the national γ ray facility Gammasphere [14] offers many advantages. High purity germanium have excellent energy resolution, $\sim$1 keV at 300 keV, enhancing the signal/ background ratio. The use of Compton suppression improves the peak-to-total ratio to >55% for 1 MeV γ rays and thus the sensitivity for low intensity branches. The segmentation of the array into 100 elements lowers the count rate in each channel while maintaining high efficiency. Finally, the near 4π coverage allows sub-sets of counters to be selected in geometries that have the lowest Compton scattering background. The use of an absolutely calibrated source allows the strength of the main branch to be calculated, not measured, as the β-branching of ^{137}Cs is extremely well known [9]. The experimental challenge then remains to observe the cascade E5 path. This procedure is discussed in more detail below.

The experiment was conducted using Gammasphere at Argonne National Laboratory with 93 detectors functional for data collection in December 2012. Each detector had a 5 cm "Hevimet" collimator to lower the rates in the BGO suppressors, and block some of the Compton scattering between germanium counters. Technically, each detector triggered in "Constant Fraction" mode, with a pre-trigger resolving time of 400 ns, and the energy threshold in each germanium counter was set at $\sim$80 keV. Data were recorded on disk for all events with multiplicity of at least two "clean" events, at pre-trigger, main trigger, and late trigger times. A $\sim$20 μCi source was selected to provide a high individual count rate ($\sim$2500 counts/s (c.p.s) suppressed) while avoiding excessive pile-up and chance coincidence events. A calibrated ^{137}Cs source was prepared by Eckert & Ziegler Isotope Products [15]. The activity at the start of the experiment was 19.27(58) μCi. Under these conditions, the Gammasphere readout rate was, on average, 7127 c.p.s, a rate completely dominated by 662-keV photons Compton scattering into two or more counters. With no source present the room background rate was $\sim$40 c.p.s. Each readout of the analog electronics caused dead time of the data acquisition system for $\sim$20 μs. This dead time could be directly extracted from the data by comparing "time stamps" between subsequent events written on disk. Dead time information is embedded in two places and was found to vary slightly from event to event. The minimum dead time can be seen as a gap in the time difference spectrum for the first 15 μs; every valid trigger has at least this. A sharp rise follows reaching a peak, and then dropping off exponentially. This exponential decay reflects the time difference between the system recovering and the occurrence of the next detected trigger. The exponential downslope can be fit in order to extract the mean time between triggers. This corresponds to a raw trigger rate of 8300(5) c.p.s. The rate of data actually stored and written to disk is 7350(10) c.p.s. This ratio reflects the loss of events when the system is dead during readout: 13.9(1)%. This is consistent with a value of 13.5(1)% that one obtains from multiplying the readout rate 7350(10) c.p.s. by a mean readout time of 18 μs, (which we have taken as the median of the low time cutoff). The variation of dead times is related to how many detectors are involved in each event. Ideally, we are only interested in multiplicity two events, but at the acquisition level, events of all multiplicities greater than two are collected, including occasional cosmic rays which fire twenty or more counters. Data were collected during a two week period for a total acquisition time of 237.5 hrs. Though mostly continuous, the run contained short gaps when the data collection system was off line. The trigger rate was fairly constant, as one would expect, but shows small variations due to data buffering. The logged data rate allowed a precise determination both of the actual run time and the varying dead time during the data acquisition cycle. The total number of decays of the source during the "live" experiment was calculated to be $6.1(2) \times 10^{11}$. Data were reduced using the GSSort program [16] embedded in the CERN ROOT package [17]. $\sim 5 \times 10^9$ events were recorded for analysis.

Following an initial sort, some detector modules were rejected as they did not have neighbors in the array for "honeycomb" suppression, so their peak-to-total performance was not optimal. The remaining 68 modules were retained for the main analysis. The first-level event selection was to indentify events with exactly two or more good energies and create a time difference spectrum for these pairs. The majority of the prompt $\gamma\gamma$ coincident events fell within a $\pm$15 ns time window. The "chance" or "random" coincident background could be sampled over a much wider time range, as it is constant everywhere. To improve the "chance" spectrum statistics, a 150 ns window was used to select background counts on either side of the peak, and the resulting spectra downscaled during time random subtraction. The time-random subtracted γ ray coincidence matrix (E_1 vs. E_2) is shown in Fig. 2. The dominant feature is the diagonal stripe corresponding to Compton cross-scattering between detectors.

As the focus for this experiment is observing two photons which combine to 662 keV, it is convenient to rotate the matrix by 45-degrees, producing a $(E_1 + E_2)$ sum vs. $(E_1 - E_2)$ difference matrix, causing the events of interest to fall along a horizontal 662-keV sum line. Selecting events with sum energy equal to 662 ± 2 keV and projecting onto the $E_1 - E_2$ axis yields the spectrum given in Fig. 3. The spectrum is dominated by Compton events of two types: scattering of photons into neighbor detectors (resulting in near equal energy sharing), or scattering of photons by $\sim$ 180 degrees (resulting in very assymetrical energy sharing). Clearly, further sorting restrictions are needed to avoid these scattering events.

For each pair of detectors in an event, the opening angle from the target can be calculated, as the position of each counter (θ, ϕ) are precisely known. The probability distribution of opening angles between two cascade γ

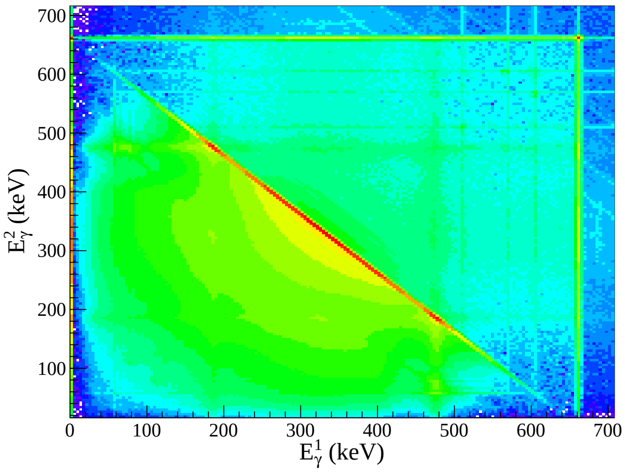

FIG. 2. (Color online)The time-selected, time-random subtracted, compressed $\gamma\gamma$ coincident matrix. All the events of interest lie on the sloping line with a total energy of 662 keV, though the overall topology of this matrix reveals much about interfering background processes.

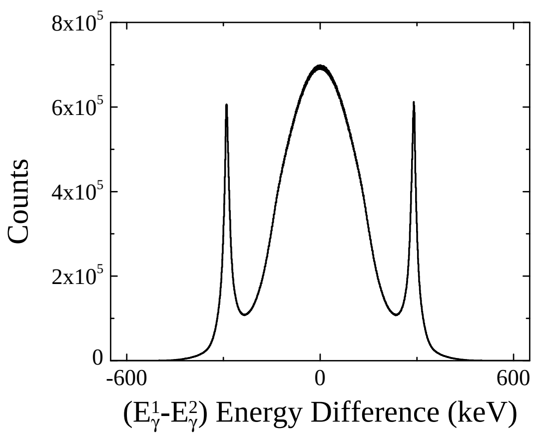

FIG. 3. A projection along the 662-keV ridge, with no angle selection. The total spectrum is dominated by Compton scattering between detectors, either across the array (large energy differences) or into near-neighbors (small energy differences)

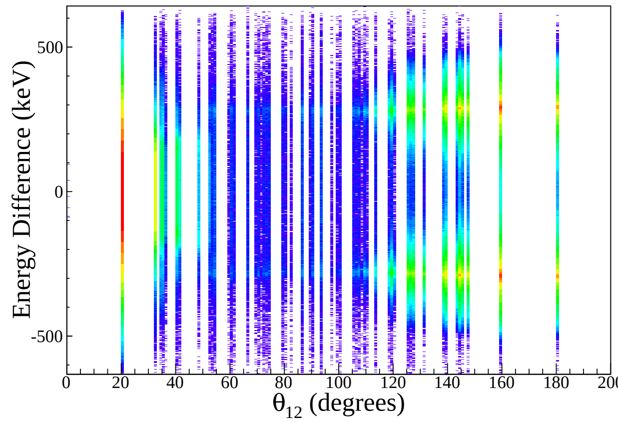

FIG. 4. (Color online)Energy difference vs. opening angle matrix. The suppressed Compton events in the opening angle range from 53° to 130° creates an ideal region for investigating the presence of real 2 photon decays.

rays is always isotropic when the intermediate state has J=1/2 [18]. This is an advantage, both when integrating over directions in the array with no counters, and in optimizing the subset of detectors for the two photon cascade search where Compton scattering is smallest. A matrix of energy difference vs. opening angle between pairs of counters is shown in Fig. 4. It is clear that there is a region of opening angles, near 90-degrees, where Compton scattering is least, as there is most material between detectors to block the cross-scattering, and the crystals are relatively far apart. Although there is a considerable loss of data when using only a subset of pairs, this selection reduces the presence of Compton scattering to a level most conducive to observing weak branches. An energy difference spectrum gated on the prompt sum energy of 662 keV and opening angles between 53° and 130° is shown in Fig 5. The 11/2-1/2-3/2 cascade is now apparent. Two peaks appear at +94.1(4) keV and -94.8(4) keV which correspond to an average energy difference of ±94.5(5) keV between the two γ energies in the cascade. The levels involved in this problem are well known,

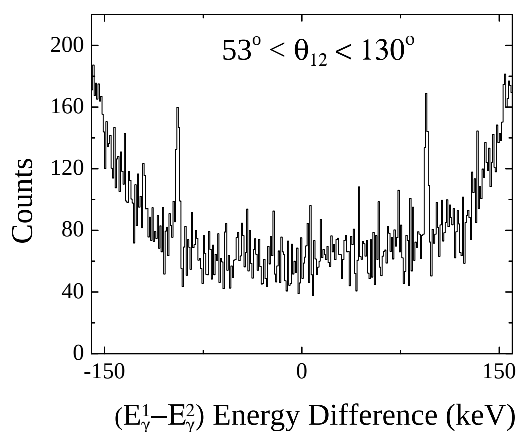

FIG. 5. As for Fig. 3, but with an opening angle selection of 53° to 130° between detectors. This subset of events has the lowest Compton scattering, so is most sensitive for a search for the two-step cascade.

661.659(3) keV [19], 283.50(10) keV [10, 11] and 0.0 keV, so the expected γ-rays are 283.5 and 378.2 keV and the energy difference peaks should appear at ±94.7(2) keV in good agreement with our observation. Consequently, there is little doubt that the 378-keV γ ray originates from the 662-keV level. The total number of counts in these peaks, after background subtraction, is 519(20).

The key remaining issues are to verify that the peaks do not arise from an artifact in the data, and then to carefully appraise the efficiency of the experiment after all the data selection criteria were applied. To ascertain if the sharp peaks arise from a higher energy background γ ray undergoing scattering, we moved the energy selection criterion to higher sum energy than 662 keV and also to lower energy. The overall shape of the difference spectrum (Fig. 5) remained constant, but the sharp peaks vanished when moving away from the 662-keV ridge. Similarly, moving the time window away from zero time difference also completely suppressed the peaks.

The absolute efficiency of Gammasphere was determined from known photo-peak coincidence intensity relations from ^{152}Eu and ^{181}Ta sources. It was determined to be slightly higher than the scaled Gammasphere accepted value for 100 detectors [16], as there was no reaction chamber in place during this experiment. Correcting for this effect gave good consistency. The number of counts in the peaks reflect the number of times the 378/284-keV cascade was detected by Gammasphere. The E5 branching ratio can be defined as the number of γ decays from the 662-keV state to the 284-keV level divided by the number of total decays from the 662-keV state:

$$Br_\gamma^{E5} = I_\gamma^{E5}/\Sigma I(\gamma + ce). \tag{1}$$

The denominator of this equation, the number of decays from the 662-keV state during the experiment, can be determined from the total number of decays of the source and the well-known β-feeding intensity to the 662-keV level [9]. This yields $\Sigma I(\gamma + ce) = 5.8(2) \times 10^{11}$. To obtain the numerator, a number of efficiency factors must be taken into account in order to transform the number of photopeak coincidences into the total number of 378-keV decays from the 662-keV level. The leading efficiency factor is the photopeak efficiency of Gammasphere for detecting both the 284-keV and 378-keV photons. For our arrangement and with the correct number of detectors used in the experiment (68) these are 0.126(4) and 0.121(4), respectively. A second restriction was placed on the detectors used during the opening angle selection, further lowering the absolute efficiency. Out of a total of 2278 detector opening angle pairs created by the 68 detectors used in the sort, only 1474 pairs had opening angles between 53° and 130°. Thus, this opening angle cut results in a further efficiency loss factor of 0.65.

A small correction arises from internal conversion of the 284-keV decay, for which there is no coincident photon. The internal conversion coefficient, α, can be precisely calculated [20]. In this case it is insensitive to the 284-keV mixing ratio. The ICC efficiency loss factor was calculated to be 0.95(1). The event losses due to the software time selection and to the event trigger efficiency were also investigated, but were found to be negligible in the current triggering mode. Finally, as discussed above, the system electronic "live time" was determined to be 0.86(2). The inferred number of 378-keV γ rays, I_γ^{E5}, is:

$$I_\gamma^{E5} = N/\left(\varepsilon_{378} * \varepsilon_{284} * fr_{pairs} * \varepsilon_{dt} * \varepsilon_{ec}\right) \tag{2}$$

with N = number of counts in the photo peaks, ε_γ = Gammasphere efficiency for current arrangement, fr_{pairs} = the fraction of opening angle pairs selected, ε_{dt} = dead time efficiency factor, and $\varepsilon_{ec} = (1 + \alpha_{284})^{-1}$ = the correction for internal conversion. Combining these efficiencies and their uncertainties we can evaluate the branching ratio to be $Br_\gamma^{E5} = 1.12(9) \times 10^{-7}$. As a consistency check, a less stringent sort of the data was made using all 93 detectors which were properly functional. This increased the efficiency of the whole experiment by 59%, giving 883(55) peak counts, but at the cost of a somewhat higher background. Following the same extraction procedure, we reach a consistent result, $Br_\gamma^{E5} = 1.00(13) \times 10^{-7}$, a verification of the data reduction. However, this is not a separate determination of the branching ratio, but merely a consistency check.

As the absolute intensity of the 662-keV transition is well known [9], our new measurement of Br_γ^{E5} allows us to calculate the absolute intensity of the 378-keV transition as $1.06(9) \times 10^{-5}$ per 100 decays of ^{137}Cs. In principle, this could affect the determination of the β-feeding intensity to the 284-keV level as this is generally deduced from a balance of intensities into and out of the level. In practice, the 378-keV transition intensity is an order of magnitude smaller than that of the 284-keV transition (with I_γ=5.8(8) x 10^{-4} per 100 decays) and thus, the β-feeding to the 284-keV level is not significantly altered by the present measurement. In Ref. [12], it was assumed that the, then unobserved, 378-keV transition intensity was on the order of 10% that of the 284-keV transition intensity and the β feeding and corresponding log *ft* value adjusted accordingly. In light of the current result, this correction was far too large and we now estimate the log *ft* value to be log*ft*=16.49(12).

In principle, for a $J^\pi = 11/2^-$ to $1/2^+$ decay, both E5 and M6 multipoles compete and interfere, with a multipole mixing ratio δ(M6/E5). However, for all cases when the lower angular momentum multipole is electric and the upper is magnetic, the magnetic transition is extremely suppressed, both as all partial widths fall rapidly with angular momentum, by about two orders of magnitude per multipole, and because magnetic widths are always much smaller than the corresponding electric widths, again by more than two orders of magnitude. Thus, the multipole mixing ratio must be so small that the M6 contribution to the total width will cause a negligible reduction in the E5 transition rate, far below the level of uncertainty of this experiment.

Given our newly measured branching ratio for this decay, and the well-known level half-life $T_{1/2} = 2.552(1)$ min, the partial E5 γ mean-life can be inferred, $1.37(11) \times 10^9$s and thus the partial decay probability. We find the decay to have a B(E5) of 0.71(6) W.u., or reduced transition probability B(E5)$\downarrow = 6.5(6) \times 10^5 e^2 fm^{10}$.

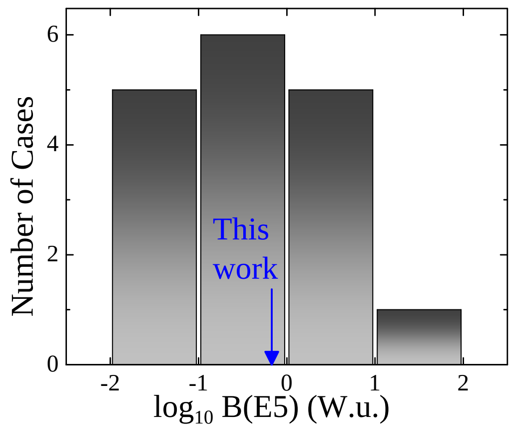

FIG. 6. (Color online)The distribution of currently known B(E5) matrix elements (in log Weisskopf units). The current determination lies right in the middle of the "single particle" transitions.

The systematics of E5 matrix elements are sparse. Some come from $\Delta J = 4$ transitions, and so require a multipole mixing ratio to infer the matrix element, which results in imprecision. Amongst the $\Delta J = 5$ transitions, less than a dozen E5 matrix elements are well established. They fall into two groups: "single particle" decays from near shell closures, which are in the 0.01-1.0 W.u. regime and "collective" decays which are ~10 W.u. The only exception to this compact distribution is a single K-forbidden decay of 10^{-7} W.u [21]. In the present case, the ^{137}Cs decay involves a structure change near the N=82 shell closure, so it is rather unsurprising that the transition lies right in the middle of the expected single-particle range. Fig. 6 summarizes our current knowledge of E5 matrix elements. This data was compiled from the NNDC website [22], which provides a search engine to investigate transitions based on various criteria; in this case, multipolarity was the selection criteria. This is a considerable improvement over the last published compilation [23] which had only 3 data points. Had this decay fallen in the "collective" regime, the branch would have been an order of magnitude bigger and had a significant impact on 2-photon physics. However, this is clearly not the case.

In conclusion, we have observed the 378-keV γ decay from the well-known 662-keV isomer in ^{137}Ba for the first time. The γ branch is small, only $1.12(9) \times 10^{-7}$. The branch translates into a B(E5) matrix element of 0.71(6) W.u., which is quite typical for "single-particle" decays of this type. The new cascade branch is ~ 30 times weaker than the theoretical estimate of the true "2-photon" decay [8]. However, the 2-photon events lie in a broad bell-shaped distribution centered at zero energy difference. Most of the counts around zero energy difference in Fig. 5 are probably associated with these 2-photon decays. The challenge is to reliably subtract the remaining Compton events in order to extract a 2-photon cross section.

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[1] M. Goppert, Naturweiss **17**, 923 (1929); Ann. Physik (Leipzig) **9**, 273 (1931).
[2] G. Breit and E. Teller, Astrophysics Journal **91**, 215 (1940).
[3] J.R. Oppenheimer and J.S. Schwinger, Phys. Rev. **56**, 1066 (1939).
[4] G.W.F. Drake, Phys. Rev. A **34**, 2871 (1986).
[5] P.H. Mokler and R.W. Dunford, Phys. Scr. **69**, C1 (2004).
[6] J. Kramp *et al.*, Nucl. Phys. **A474**, 412 (1987).
[7] W. Beusch., Helv. Phys. Acta. **33**, 363 (1960).
[8] D.J. Millener, R.J. Sutter, and D.E. Alburger, Bull. Am. Phys. Soc. DNP CF8 (2011).
[9] E. Browne and J.K. Tuli, Nucl. Data Sheets **108**, 2173 (2007).
[10] I. Bikit *et al.*, Phys. Rev. C **54**, 3270 (1996).
[11] B.K. Wagner *et al.*, J. Radioanalytical Chem. **219**, 217 (1997).
[12] V.O. Sergeev and F.F. Valiev, Bulletin of the Russian Academy of Sciences: Physics **71**, 827 (2007).
[13] V.A. Bondarenko and I.L. Kuvaga, Nucl. Phys **A582**, 1 (1995).
[14] I-Yang Lee, Nucl. Phys. A **520**, 641c (1990).
[15] Eckert and Ziegler Isotope Products Inc. 24937 Ave. Tibbetts, Valencia CA 91355.
[16] T. Lauritsen, `http://www.phy.anl.gov/gammasphere/doc/GSSort/GSUtil.html`.
[17] ROOT Data reduction package. `http://root.cern.ch/drupal/`.
[18] A.H. Wapstra, G.J. Nijgh, R. van Lieshout, Nuclear Spectroscopy Tables, North Holland Publishing Company, Amsterdam (1959).
[19] R.G. Helmer, C. van der Leun, P.H.M. Van Assche, Recommended Standards for γ-Ray Energy Calibration (unpublished) (1995).
[20] T. Kibédi, Australian National Laboratory Preprint ANU_P/1839 v. 2.3 (2012); `www.nndc/bnl.gov/nndcscr/ensdf_pgm/analysis/BrIcc/`.
[21] M.B. Smith *et al.*, Phys. Rev. C **68**, 031302(R) (2003).
[22] A. Sonzogni, NNDC, Brookhaven National Laboratory, `http://www.nndc.bnl.gov/chart/`.
[23] P.M. Endt, At. Data Nucl. Data Tables **26**, 47 (1981).