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Developing ^{152}Eu into a standard for detector efficiency calibration

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Abstract

A γ–γ coincidence experiment was performed to check the ^{152}Eu 13-year decay scheme and the placement of the observed γ-ray transitions. The multi-detector array for residual activity measurement of the Linear Accelerator Laboratory was used. The source activity was 1 MBq, and about 10^9 coincidence events were observed. About 30 γ's were placed in the decay scheme and four ^{152}Sm levels were added to the previously known set of levels fed in ^{152}Eu 13-year decay.

Keywords: ^{152}Eu 13-year decay; Gamma transitions; Level scheme

1. Introduction

There are many aspects in the development of a multi-gamma-ray source standard for an accurate efficiency calibration. As noted in a companion paper (Castro et al., 2004), the statistical correlations between the gamma-ray intensities must be known and used in the calibration procedure. At the same time, the precision in the intensities depends on the accuracy of the decay scheme. A γ–γ coincidence experiment is described in the current study with the aim of checking the level scheme of the daughters of 13-year ^{152}Eu decay and the placement of the observed γ-ray transitions.

About 130 γ-ray transitions are known to follow the decay, with intensities above 10^{-5} photons per parent decay. A recent evaluation of the ^{152}Eu decay scheme for the Decay Data Evaluation Project (Vanin et al., 2001a, b) found 16 unplaced transitions, totaling $\sim$0.0035 photons per decay. Were all these unplaced gamma transitions to feed the ground state, this extreme degree of population would be equivalent to the magnitude of the relative error in the efficiency calibration, which would be excessive in a standard.

A number of ^{152}Eu γ–γ coincidence measurements have been undertaken using two semiconductor detectors (Riedinger et al., 1970; Barrette et al., 1971a; Baker et al., 1972; Ismail et al., 1981; Stewart et al., 1990). The measurement reported here was performed with a multi-detector array, developed for residual radioactivity. Data were accumulated for a long period, and the resulting sensitivity is much higher than the measurements reported in the listed references.

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2. Experimental method

The multi-detector array for residual activity measurements at the Linear Accelerator Laboratory consists of four HPGe detectors with efficiencies varying in the range 15–40% relative to $3'' \times 3''$ NaI(Tl). These detectors are mounted in the horizontal plane with the axis of neighboring detectors at right angles, and shielded to reduce the detection of photons scattered from the active volume of other detectors in the array (using lead bricks at their sides and pierced lead sheets between their faces), with the source placed at the center of this arrangement.

The timing signals were taken from constant-fraction discriminators without slow rise-time rejection. A controller designed to control pile-up rejection and the busy status of the amplifiers reduced the number of single events acquired, and drove the ADCs and the TDC. Acquisition was performed through CAMAC, with a controller driven by a PC trough the ISA bus (Bärg Filho et al., 1998). Data were acquired in list mode, and the analysis was performed off-line.

A 1 MBq source was monitored over a three-month run, resulting in $\sim 10^9$ coincidence events. The ratio between the ^{154}Eu and ^{152}Eu activities was 0.218(6)%. Time windows about 80 ns wide were selected for total and chance coincidences, the detector gains were matched, and the corresponding 4k × 4k channels^2 γ–γ matrices were constituted. The resolution at the high-energy end of the spectrum was 2.5 keV, and at the low-energy end was 1.5 keV. A peak in the resulting total coincidence matrix that corresponds to a γ–γ cascade once in 10^5 decays is about 1000 counts, and constitutes a peak that in most cases will stand out from the background.

The peak-areas were determined using the least-squares method and fitting two-dimensional Gaussian peaks, plus ridges for coincidences with Compton-scattered photons and a suitable plane for the continuum background events (Guimarães Filho, 1999). The full-width at half-maximum was calibrated using the observed values for strong, isolated peaks, and kept fixed at the interpolated values for the other peaks. Approximately 1500 two-dimensional peaks were observed, about 65% corresponding to chance coincidences.

Efficiency and energy were calibrated using the most intense transitions of ^{152}Eu in order to study the weak unplaced transitions. The relative efficiency precision is about 5%, while the energy precision is 30 eV. Angular correlation effects were corrected using the most likely spins and transitions multipolarities (Vanin et al., 2001a, b), making sure that a uniform mixing-parameter phase convention was used throughout (Taylor et al., 1971; Krane and Steffen, 1970; Anicin et al., 1972). Finite solid-angle corrections were estimated according to Barrette et al. (1971b). However, there was no need for correction in most cases because we analyzed the sum spectra of all pairs of detectors, where the number of spectra of detectors forming 90° was twice the number of detectors forming 180°. The biggest multiplicative correction factor for the peak-areas of 1.36 applied to the $0^+ \rightarrow 2^+ \rightarrow 0^+$ cascade, while the smallest had a value of 0.97. There were only two other cascades requiring factors that differed significantly from unity, with the values of 1.10 and 1.03.

3. Results

Table 1 summarizes the results of the experiment that correspond either to new transitions or to important changes in previously accepted values. Gamma-ray transition intensities were calculated from the coincidence peaks using the known value of the intensity for the coincidence partners, and adopting the values from the recent evaluation (Vanin et al., 2001a, b) whenever available; otherwise, the value obtained in this work was used. Since each transition was observed in several coincidences, the intensity corresponds to an average of the intensities calculated after the corrections due to sum, pile-up, chance coincidences, efficiency and angular correlation. For each listed gamma ray (besides energy, intensity and placement in the decay scheme), we add a comment either to define what was previously known to this study or specify the unfolded multiplet peak.

Compared to previous work, we found evidence for four additional levels of ^{152}Sm being populated in the decay of 13-year ^{152}Eu. Two peaks, previously assigned as gamma-ray transitions, were identified as secondary detection effects. We present briefly the arguments used to place the levels, the main changes in the gamma-ray assignments, and the list of transitions that remain unplaced.

3.1. Level at 1083 keV

Already known from the ^{152}Pm decay, the observation of the 210-keV transition from the 1293-keV level in coincidence with the 272- and 961-keV transitions feeding the 810-keV and 122-keV levels, respectively, establishes the population of this level in the decay of 13-year ^{152}Eu. All these transitions are weak and the peaks are located close to strong peaks or ridges. Fig. 1 shows the region of the coincidence spectrum where the 210–272 keV coincidence peak can be seen, providing an example of a γ–γ coincidence peak observed near the detection threshold.

Table 1
Most important results from the γ–γ coincidence measurement

E_γ (keV)	Photons/10^5 decays	Parent and level energies			Comments (I_γ in photons/10^5 decays)
		Z	E_i (keV)	E_f (keV)	
150.09(10)	0.83(6)	Sm	1371.691	1221.5	New
174.80(22)	1.38(18)	Gd	930.562	755.396	Kawaldeep et al. (1993), $I_\gamma = 21(1)$, unplaced
195.22(15)	2.04(18)	Gd	1318.5	1123.184	Stewart (1990), $I_\gamma = 6(1)$, unplaced
209.41(13)	1.2(4)	Gd	1643.395	1434.025	Previous $I_\gamma = 5.5(5)$
209.976(26)	4.3(7)	Sm	1292.771	1082.8	Unfolds 209.41(13), $I_\gamma = 5.5(5)$
237.106(26)	6.24(22)	Sm	1529.807	1292.771	Unplaced
272.39(10)	0.88(11)	Sm	1082.8	810.447	New
286.53(11)	1.28(12)	Sm	1579.365	1292.771	Unfolds 285.98(3), $I_\gamma = 10.0(6)$
287.10(12)	0.81(7)	Gd	1605.255	1318.5	Unfolds 285.98(3), $I_\gamma = 10.0(6)$
315.10(3)	39.4(13)	Gd	930.562	615.563	Previous $I_\gamma = 49.6(17)$
316.2(2)	10.2(4)	Sm	1022.967	706.91	Previous $I_\gamma = 3.1(10)$
320.08(5)	2.18(10)	Sm	1612.94	1292.771	Unplaced
324.914(26)	4.97(24)	Gd	1643.395	1318.5	Unfolds 324.83(3), $I_\gamma = 73.8(15)$
328.764(26)	3.47(20)	Gd	1643.395	1314.71	Unfolds 329.425(21), $I_\gamma = 129(6)$
345.538(10)	10.1(6)	Sm	1579.365	1233.866	Unfolds 344.2785(12) 26580(1 2 0)
348.751(15)	1.78(10)	Sm	1371.691	1022.967	New
354.16(15)	0.97(9)	Gd	1109.076	755.396	New
358.45(7)	1.50(9)	Sm	1730.241	1371.691	Unfolds 357.26(5), $I_\gamma = 4.0(5)$
391.17(8)	1.49(8)	Sm	1612.94	1221.5	Unplaced
401.25(11)	0.59(6)	Sm	1085.850	684.714	New
476.43(10)	1.54(10)	Sm	1769.3	1292.771	New
514.77(6)	0.38(3)	Sm	1221.5	706.91	New
535.33(14)	1.85(23)	Sm	1769.3	1233.866	Unplaced
589.74(9)	1.32(9)	Sm	1612.94	1022.967	New
683.70(9)	4.9(3)	Sm	1769.3	1085.850	Unplaced
707.15(7)	1.33(8)	Sm	1730.241	1022.967	New
734.12(12)	0.83(7)	Sm	1757.151	1022.967	Unfolds 735.43(4), $I_\gamma = 5.8(10)$
735.43(4)	4.32(19)	Sm	1776.56	1041.134	Unplaced
756.17(3)	4.22(19)	Sm	1779.11	1022.967	Unplaced
813.20(6)	3.91(20)	Sm	1776.56	963.36	New
850.10(13)	0.77(7)	Gd	1605.255	755.396	New
855.19(7)	1.94(14)	Sm	1221.5	366.479	New
919.73(4)	6.12(28)	Sm	1730.241	810.447	Unfolds 919.337(4), $I_\gamma = 429(5)$
961.08(4)	10.9(9)	Sm	1082.8	121.782	New
968.64(4)	7.8(3)	Sm	1779.11	810.447	Kawaldeep et al. (1993) $I_\gamma = 3.7(24)$, unplaced
970.22(9)	1.2(3)	Gd	1314.71	344.280	New
1084.41(5)	10.3(7)	Sm	1769.1	684.714	Meyer (1990), $I_\gamma = 224(8)$, unplaced
1318.38(17)	1.58(14)	Gd	1318.5	0	New

The first two columns characterize the gamma ray; columns 3–5 give the placement proposed in this work; and the last column shows either previous knowledge or identifies the transition that obscured the transition (energy in keV, and intensity in units of photons/10^5 parent decays). Comments and level energies are from Vanin et al. (2001a, b).

3.2. Level at 1221 keV

There are previous suggestions for this level in ^{152}Sm, decaying through 514- and 855-keV gamma-ray transitions tagged "uncertain" (Firestone et al., 1996). We observed both transitions in coincidence with the 391-keV γ ray, already assigned to ^{152}Eu but unplaced (Vanin et al., 2001a, b). The 855-keV transition was also observed in coincidence with the 150- and 244-keV transitions.

3.3. Level at 1776 keV

Other nuclides undergoing beta disintegration also feed this level in ^{152}Sm, which is known to decay by 735- and 813-keV γ-ray transitions. The previously unplaced 735-keV transition was observed in coincidence with the 674- and 919-keV γ rays, which decay from the 1041-keV level. The 813-keV γ ray has not previously been assigned to the 13-year ^{152}Eu decay, but was observed in coincidence with

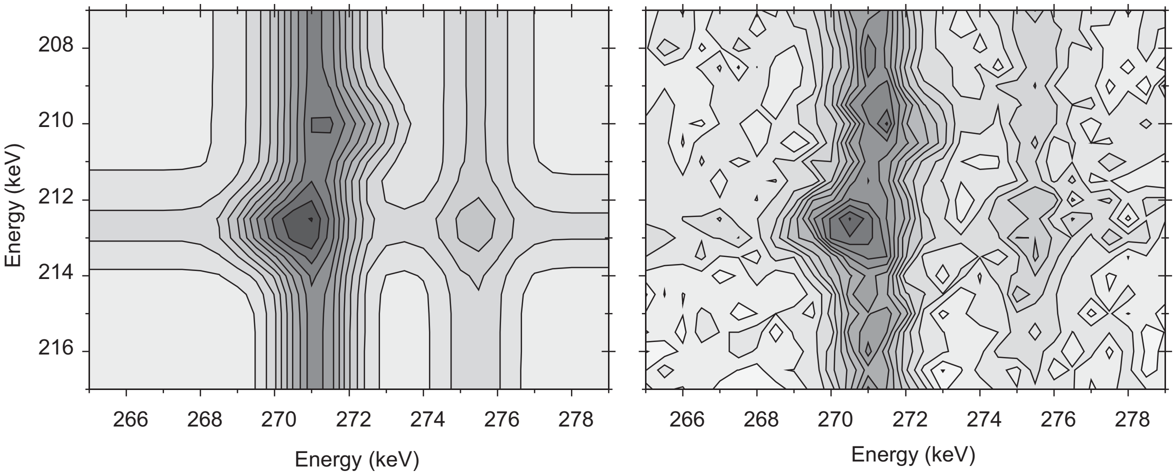

Fig. 1. Contour plots of the coincidence spectrum around the 210–272 keV peak, with the fitted spectrum to the left and raw data to the right. The ridge at 271.1 keV dominates the region. Two other ridges, at 213 and 275 keV, simulate a peak at 213–275 keV. The crossing of the ridges at 271 and 213 keV causes the stronger appearance of the 212–270 keV peak (which effectively has an area similar to 210–272 peak), and displaces the peak positions in relation to the highest points in the plot.

the 963- and 841-keV transitions that decay from the 963-keV level.

3.4. Level at 1779 keV

The unplaced 756-keV γ ray was observed in coincidence with the 901- and 656-keV transitions, and a new 969-keV transition was observed in coincidence with the 688- and 810-keV transitions, establishing firmly the existence of this new level. The decay properties suggest spin $(2,3,4)^{(+)}$.

3.5. 1084 keV peak in the gamma spectrum

The observed intensity of the 1084.1-keV γ ray of $I_\gamma = 0.0103(7)\%$ is much smaller than previous results: $I_\gamma = 0.224(8)\%$ (Meyer, 1990). It is unlikely that a 1084 level decaying exclusively to the ground state can be fed only by the parent, because such relatively strong feeding could only happen if the level spin is 1 or 2 (in which case the level would most probably receive some feeding from more excited levels, and would be seen in this coincidence experiment). We suggest that the previously observed peak corresponds to the shoulder of the pile-up peak of the 121- and 964-keV γ rays.

3.6. 897 keV peak

We identify this peak with the annihilation single-escape peak from the 1408-keV γ ray. This peak is only observed in true coincidence with the 122-keV transition from ^{152}Sm, and is clearly wider than the 901-keV γ-ray peak. The intensity of the observed peak is compatible to what is expected for the 1408-keV single-escape peak.

The energy assigned previously to this peak of 896.58(9) keV differs significantly from the energy expected for the 1408.013(3)-keV single-escape peak, which we explain by the interference of the strong Compton edge of the 1112-keV gamma ray.

3.7. Gamma rays remaining unplaced

The weak γ ray at 406.74(15) keV with $I_\gamma = 0.00083(21)\%$ was clearly seen as pile-up of the 121-keV γ ray with the sum-peak between the 244-keV γ ray and KX-rays from Sm. However, there were some γ rays assigned to the decay of 13-year ^{152}Eu (Vanin et al., 2001a, b) that were not seen in the coincidence experiment, with the following energies and intensities: 379.37(6), 0.83(21); 595.61(1), 3.1(17); 696.87(19), 2.9(10); 1001.1(3), 4.6(10); 1139(1), 1.3(3); 1674.30(6), 6.0(8), in keV and photons/10^5 decays, respectively. Since they were not placed in the decay scheme by the experimentalists that observed them, it is impossible to place an upper limit on their intensity. Even in single γ-ray spectroscopy measurements, difficulties would be experienced in confirming their intensities or placing upper limits significantly lower than the reported intensities, because all of them fall in regions of the spectrum dominated by secondary detection effects. Nevertheless, the sum of the intensities of all these transitions is smaller than 0.0002/decay.

4. Conclusions

About 30 γ-ray emissions were placed in the decay scheme of ^{152}Eu that had either been already assigned but not placed yet, or were observed for the first time. Four ^{152}Sm levels were observed for the first time to be fed in the decay of 13-year ^{152}Eu, three of them already known from the decays of other nuclides and one observed for the first time.

The main difference in the list of γ rays assigned to the decay of 13-year ^{152}Eu was the withdrawal of lines at 1084 and 897 keV, attributed to secondary detection effects. There are many small changes in the intensities of the transitions involved in the unfolded multiplets, all in the range 10^{-4}–10^{-5} photons per parent decay.

The resulting improved decay scheme permits the intensities of the most intense gamma rays to be determined with relative precisions of about 0.1%. The main difficulty resides in the determination of the internal conversion coefficients of the 122-keV transition.

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