Determination of the emission probability of the 21.54 keV γ ray in the decay of ^{151}Sm

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The emission probability of the 21.54 keV γ ray in the decay of ^{151}Sm was experimentally determined by means of the measurements of the 21.54 keV γ-ray emission rate and the activity of ^{151}Sm with high-resolution Ge, Si(Li) γ-ray spectrometry and liquid scintillation counting (LSC), respectively. The resulting emission probability of the 21.54 keV γ ray is 0.0324% with a standard uncertainty of 0.0013% based on a quadratic summation of the uncertainty components from the measurements of the γ-ray emission rate and the activity of ^{151}Sm. The result agrees with the latest literature value (partially theoretically based), 0.0318 $\pm$ 0.0022%, with reduced uncertainty.

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I. INTRODUCTION

^{151}Sm is a long-lived fission product with a fission yield of ~0.4% and can be used as a tracer in the fields of environment and life sciences. ^{151}Sm also is a important nuclide in astrophysics for characterizing nucleosynthesis in the slow neutron capture process, and an excellent thermometer for the *s* process [1]. The present investigation on the emission probability of 21.54 keV γ rays in the decay of ^{151}Sm was undertaken to resolve some of the discrepancies in the literature and to reduce the uncertainties in the existing decay schemes of ^{151}Sm. Furthermore, an accurate value of 21.54 keV γ-ray intensity is needed to accurately assay ^{151}Sm by γ-ray spectrometry for various applications, such as nuclear physics, medicine, and nuclear chemistry.

A decay scheme constructed in Ref. [2] is displayed in Fig. 1 with the new half-life measured by our laboratory [3]. A large majority of β^- transition events go directly to the ground state of ^{151}Eu, only a weak β branch (0.91%) feeds the first excited state at 21.54 keV followed only by a 21.54 keV γ transition to the ground state of ^{151}Eu via the emission of either γ rays or internal conversion electrons. The emission probability of the γ rays depopulating the 21.54 keV state has not been accurately measured up to now principally because of the very weak β branch and the very large internal conversion coefficient (ICC). In the Nuclear Data Sheets, the latest BR value of the 21.54 keV γ ray was calculated using the equation $I_\beta/(1+\alpha)$ to be 0.0318 $\pm$ 0.0022% [4], where the β branch I_β is 0.91 $\pm$ 0.06% [5,6], and the theoretical ICC α is 27.6 [7] with 1.4% uncertainty coming from Raman-band calculation. The large uncertainties in this value hindered their use in the above-mentioned fields. So, the emission probability of 21.54 keV γ rays in the decay of ^{151}Sm should be experimentally determined in an effort to examine the calculated value and reduce its uncertainty.

In present work, the nuclide ^{151}Sm was produced by the neutron irradiation of a Sm_2O_3 target enriched in ^{150}Sm to 87.34% in the heavy water research reactor at China Institute of Atomic Energy for ~15 days. The neutron flux was 4.5 $\times$ 10^{13} n/cm^{2}s at the sample irradiation site. The ratio of ^{151}Sm/^{150}Sm in the activated sample was measured by a thermal ionization mass spectrometer (TIMS) to be ~3.36 $\times$ 10^{-3} [3]. The emission probability of the γ ray was obtained by the equation $BR = \gamma PS(21.54)/A$, where the gamma emission rate, $\gamma PS(21.54)$, of the ^{151}Sm sample was measured with both a high-resolution Ge and a Si(Li) spectrometer, and the radioactivity, A, with liquid scintillation counting (LSC).

II. EXPERIMENTAL

A. Measurement of emission rate of 21.54 keV γ ray in ^{151}Sm decay

After a cooling time for more than 5 yr, an irradiated Sm_2O_3 sample (enriched in ^{150}Sm) was dissolved with 68% HNO_3. The solution was heated on a hot plate and evaporated to near dryness, then 20 mL 0.01M HNO_3 was added to produce an essentially salt-free ^{151}Sm solution to eliminate the influence of the self-absorption effect on the γ emissivity. The concentration of Sm in sample solution, measured with ICP-MS, is (2.044 $\pm$ 0.031) $\times$ 10^{-4} g Sm/mL. In order to improve the accuracy, two point sources (the diameter of the active zone is ~0.5 mm with an accurately known activity ratio were made of the salt-free ^{151}Sm solution.

The sources were measured by an ORTEC Si(Li) γ(x) ray spectrometer system composed of a Si(Li) detector of SLP-10180-P, a DSPEC-PLUS 92X unit, and a computer [8]. The system has an energy resolution of 0.18 keV for a 5.9 keV γ ray. Efficiency functions were established for the energy range of interest (properly covering 21.54 keV) by using 21.99, 22.16, and 23.17 keV x rays of ^{110m}Ag; 20.78 and 21.34 keV x rays and 26.34 keV γ ray of ^{241}Am; and the 32.19 keV x ray of ^{137}Cs for measurement positions of 0, 1.0, and 2.0 cm from the detector surface. The emission rates of the above x(γ) rays were obtained from the accurately known activities of the standard

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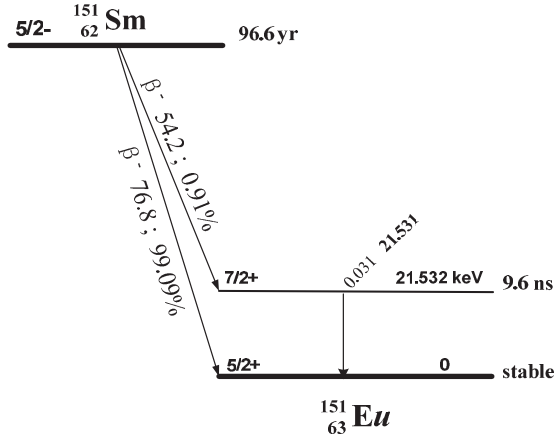

FIG. 1. The decay scheme of ^{151}Sm.

^{110m}Ag, ^{241}Am, and ^{137}Cs sources and their respective branching ratios from the literature [2]. As shown in Fig. 1, there are no cascade or crossover transitions related to 21.54 keV γ ray, thus no coincidence summing effect on its emission rate measurement. The resulting peak efficiency curves at three counting positions are expressed by Eqs. (1)–(3):

$$\ln(\varepsilon_P) = -9.964590 + 4.83383\ \ln(E) - 0.93256\ \ln(E)^2 \times 0\ \text{cm}\ 20\ \text{keV} \leqslant E \leqslant 32\ \text{keV}, \quad (1)$$

$$\ln(\varepsilon_p) = -14.4430 - 7.4269\ \ln(\text{E}) - 1.36746\ \ln(E)^2 \times 1.0\ \text{cm}\ 20\ \text{keV} \leqslant E \leqslant 32\ \text{keV}, \quad (2)$$

$$\ln(\varepsilon_p) = -12.0512 + 5.43801\ \ln(\text{E}) - 1.02625\ \ln(E)^2 \times 2.0\ \text{cm}\ 20\ \text{keV} \leqslant E \leqslant 32\ \text{keV}. \quad (3)$$

The intrinsic full peak efficiency of 21.54 keV, $\varepsilon_p(21.54)$, was calculated using formulas (1), (2), and (3) to be 1.997%, 1.078%, and 0.655% for 0, 1.0, and 2.0 cm counting positions, respectively, with a relative standard uncertainty of 3.0% derived from the uncertainties of the peak efficiency curves, counting statistics of 21.54 keV peak, and the emission rates of the x rays in the standard sources used for efficiency calibration. A background γ spectrum showed no interference with the measurement of the 21.54 keV peak. A Si(Li) γ(x) spectrum is shown in Fig. 2.

In addition, an N-type HPGe-detector system (resolution: 1.8 keV, relative efficiency: 47%) was also used to measure the same samples. The two samples were measured at the position 7 cm from the surface of the HPGe detector for more than 20 h. A γ spectrum is shown in Fig. 3. The detector was previously calibrated for efficiency at the above counting geometry. All results of 21.54 keV γ ray emission rate are listed in Table I. The final value of $\gamma PS = 0.2029 \pm 0.0069$ Bq/mg was obtained by Eq. (4):

$$\overline{\gamma PS} = \frac{\sum_i^N \gamma PS_i/u_i^2}{\sum_i^N 1/u_i^2}, \quad (4)$$

where γPS_i and μ_i stand for the γ emissivities of 21.54 keV γ ray and uncertainties measured in different positions of different detectors. The average 21.54 keV γ emission rate per mg Sm is $\gamma PS = 994.5$, with a relative standard deviation of 3.0%.

B. Activity measurement of ^{151}Sm

^{151}Sm is essentially a pure β^- decay nuclide with a very weak β branch (0.91%) feeding the first excited state of ^{151}Eu at 21.54 keV. The activity of ^{151}Sm was measured by liquid scintillation counting (LSC) which was already mentioned in our former paper [3]. The main difficulty in LSC is the discrimination between the nuclide of interest, ^{151}Sm, and the radioactive contaminants present in the sample solution which can induce complex spectra overlapping and quenching effects. As can be seen from the γ-ray spectrum of the ^{151}Sm sample shown in Fig. 4, ^{152}Eu and ^{154}Eu are the main

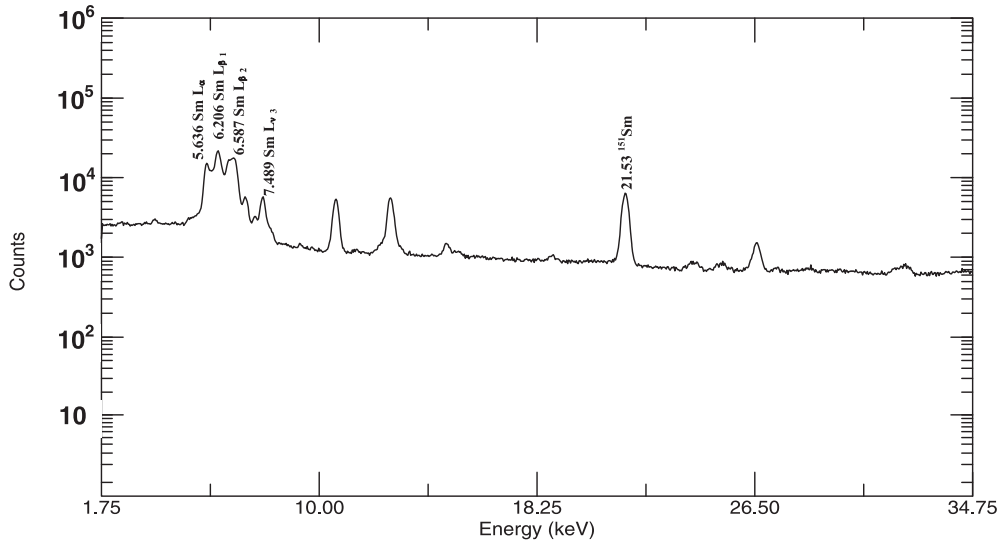

FIG. 2. A Si(Li) γ(x)-ray spectrum of a ^{151}Sm sample.

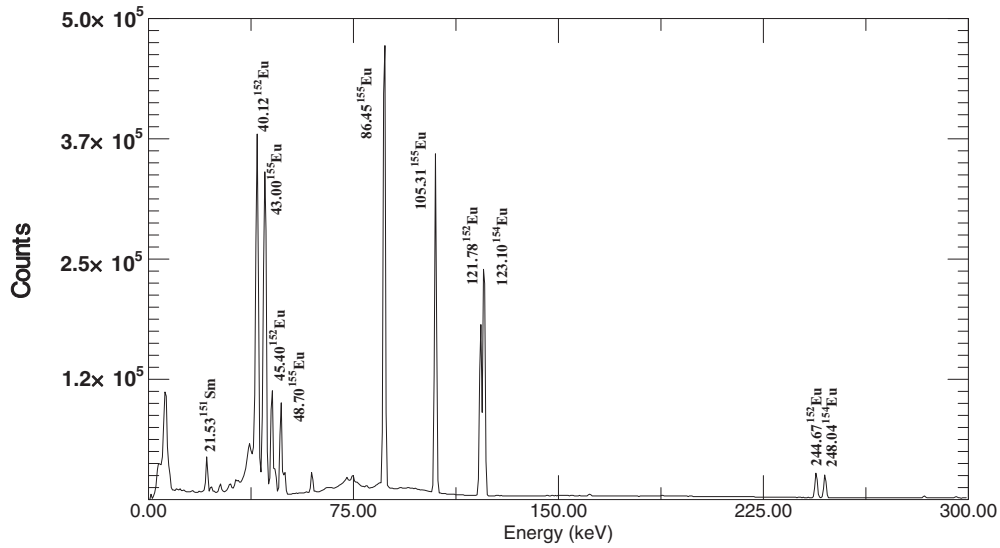

FIG. 3. A HPGe γ-ray spectrum of a ^{151}Sm sample.

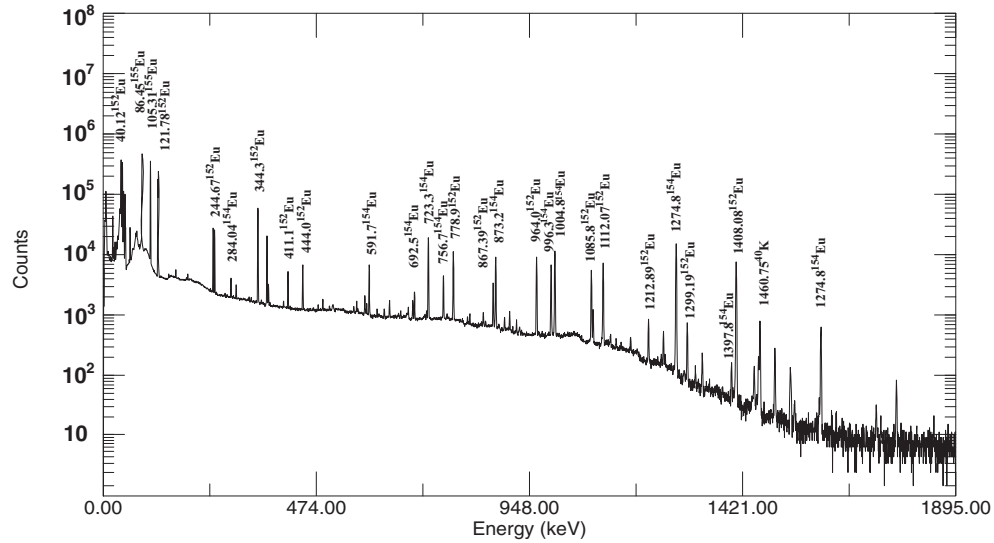

FIG. 4. A HPGe γ-ray spectrum of a ^{151}Sm sample.

TABLE I. Results of 21.54 keV γ emission rate per mg Sm [using the Si(Li) and HPGe].

Instrument, sample No., counting site	Peak count rate (s/mg Sm)	Efficiency	Emission rate (γ/s/mg Sm)	Uncertainty. %
Si(Li), 1, 0 cm	18.89	0.01938	975	3.1
Si(Li), 1, 1 cm	10.72	0.01078	994	3.1
Si(Li), 1, 2 cm	6.738	0.00669	1007	3.1
Si(Li), 2, 0 cm	18.14	0.01938	935	3.2
Si(Li), 2, 1 cm	10.83	0.01078	1004	3.1
Si(Li), 2, 2 cm	6.957	0.00669	1040	3.5
HPGe, 1, 7 cm	17.33	0.01726	1004	3.1
HPGe, 2, 7 cm	17.19	0.01726	995	3.2
Average 21.54 keV γ emission rate per mg Sm $\pm$ RSD			994.5	3.0

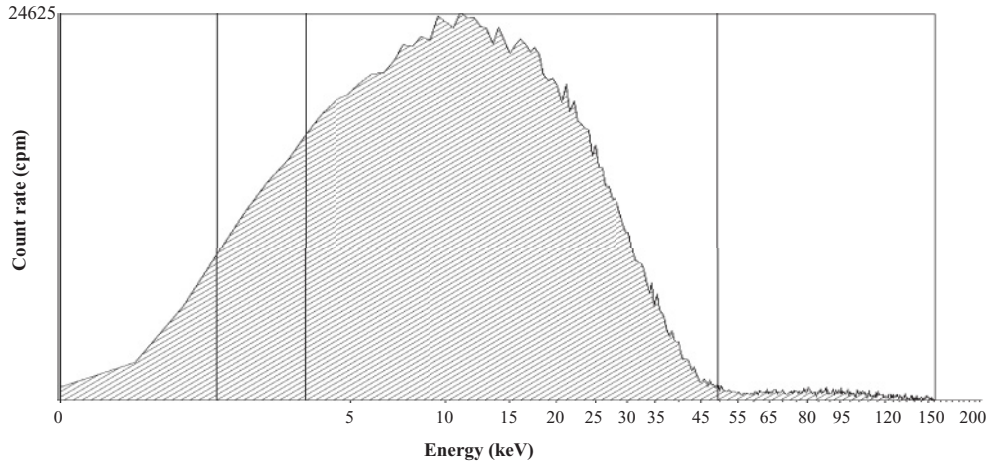

FIG. 5. A LSC entire spectrum of a ^{151}Sm sample (log-log coordinate).

β^--γ-emitting contaminants. The activity of ^{152}Eu in a 1-mg Sm sample, $A(^{152}\mathrm{Eu})$, was calculated using formula (5) to be 3.74×10^3 Bq, with a relative standard uncertainty of 1.3% mainly derived from the uncertainties of the peak efficiency curve,

$$A(^{152}\mathrm{Eu}) = \mathrm{Cps}(344.3)/[\varepsilon(344.3)\, B(344.3)], \qquad (5)$$

where Cps(344.3), $\varepsilon(344.3)$, and $B(344.3)$ stand for peak counts per second, peak efficiency, and branching ratio of 344.3 keV γ ray in the decay of ^{152}Eu, respectively. The activity of ^{154}Eu was calculated using the same method to be 4.18×10^3 Bq/mg Sm sample, with a relative standard uncertainty of 1.3%. Several other γ rays of these two nuclides (such as 244.7 keV, 1085.8 keV, 1112.1 keV γ ray of ^{152}Eu and 591.7 keV, 723.3 keV, 1004.8 keV γ ray of ^{154}Eu) were also used in calculating the activities of ^{152}Eu and ^{154}Eu, and the results are consistent with the previous results within uncertainty limits, which confirms that the total activity of ^{152}Eu and ^{154}Eu was $<8 \times 10^3$ Bq/mg Sm sample and can be neglected compared to the activity of ^{151}Sm ($\sim 3 \times 10^6$ Bq/mg Sm sample). Furthermore, the end-point energy for ^{151}Sm β^- decay is 76.7 keV, while those for ^{152}Eu and ^{154}Eu β^- decay are $\sim$500 keV; therefore the contribution from $^{152+154}$Eu to the energy region of ^{151}Sm in the LSC spectrum will be even smaller.

A model 3170 TR/SC Tri-Carb LSC spectrometer with low-background double photomultiplier tubes was used to measure the activity of ^{151}Sm. The detection efficiency is a function of the β^- spectrum of the radionuclide and the light yield of the scintillator, which depends on the LSC cocktail itself but also on the chemical composition of the source. The LSC was calibrated by the efficiency tracing method using a ^{14}C standard source ($A = 1896 \pm 19$ DPS, certified by China National Institute of Metrology) with the same chemical and physical parameters (i.e., volume, source-to-cocktail volume ratio, acidity concentration) and quenching agent (nitromethane) as for the ^{151}Sm sample. Blank contribution was corrected for by using the same LS cocktail containing natural Sm. A ^{151}Sm source containing 26.1 mL sample solution (containing 5.335 mg Sm) was measured, and a LSC spectrum is shown in Fig. 5. Taking into account the detection efficiency, the specific activity of ^{151}Sm is therefore $(3.07 \pm 0.08) \times 10^6$ Bq/mg Sm.

III. RESULTS AND DISCUSSION

The emission probability of the 21.54 keV γ ray of ^{151}Sm, BR, was calculated from the absolute γ ray emission rates $\gamma PS(21.54)$ and the activity (A) of ^{151}Sm in samples with the same mass of Sm by Eq. (6):

$$BR = \gamma PS(21.54)/A, \qquad (6)$$

where $\gamma PS(21.54)$ (=994.5 $\pm$ 29.9 Bq/mg) is the specific γ emissivity of 21.54 keV γ ray, A (=$3.07 \pm 0.08 \times 10^6$ Bq/mg) is the specific activity of ^{151}Sm. The emission probability of 21.54 keV γ ray of ^{151}Sm is thus 0.0324 $\pm$ 0.0013%. The uncertainty budget is given in Table II.

TABLE II. Uncertainty budget in measurement of emission probability of 21.54 keV γ ray in the decay of ^{151}Sm.

	Uncertainty component	Relative standard uncertainty (%)
γ ray emission rates	Counting statistics	0.5
	Full energy peak efficiency	3.0
Activity of ^{151}Sm	Impurities	2.3
	Detection efficiency for ^{151}Sm	1.0
	Counting statistics	0.2
	Sample weighing	0.1
	Total	3.9

The result of the emission probability of 21.54 keV γ ray in the decay of ^{151}Sm from this work, $0.0324 \pm 0.0013\%$, agrees with that of the latest literature value, $0.0318 \pm 0.0022\%$ [4], within uncertainty limits. The relative standard uncertainty of our result is reduced by ~50%, relative. More importantly, as mentioned above, since the literature value was derived from a weak β^- branching ratio (with a large uncertainty) and a theoretical ICC, the cited uncertainty may be much smaller than it should be, while the result from the present work was obtained completely by experimental measurements.

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