Emission probability of the 66.7 keV γ transition in the decay of ^{171}Tm

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The γ-emission probability of the 66.73 keV line in the decay of ^{171}Tm has been experimentally determined using γ spectrometry and inductively coupled mass spectrometry. Using a set of two reference sources, namely ^{60}Co as primary standard and ^{44}Ti/^{44}Sc in secular equilibrium as secondary standard, we were able to deduce the detection efficiency at 66.73 keV of the used γ-spectrometry setup. The emission probability of the 66.73 keV γ transition in the decay of ^{171}Tm has been determined afterwards with a set of three radioisotopically pure ^{171}Tm samples to be 0.159(5)%.

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I. INTRODUCTION

The radioactive isotope ^{171}Tm, $t_{1/2} = 1.92(1)$ a, is a β emitter with a low Q value of 96.4(10) keV [1]. This isotope represents one out of 21 astrophysical interesting branching point nuclei along the s-process path [2] to synthesize elements beyond iron. The competition between the β decay of ^{171}Tm and an additional neutron capture influences the abundance of stable ^{171}Yb and ^{172}Yb and hence the abundance of isotopes of heavier elements. Recently, the isotopic ratio of these isotopes was measured in presolar SiC grains showing significant deviations from stellar models describing the evolution of asymptotic giant branch stars [3]. This discrepancy might be caused by insufficient knowledge of the nuclear properties of ^{171}Tm. For instance, very recently experiments were performed to investigate the neutron capture cross section of ^{171}Tm [4] but precise characterization of the amount of ^{171}Tm target material used in these experiments strongly depends on the knowledge of the γ-emission probability of this isotope [5].

^{171}Tm can be produced by neutron activation of ^{170}Er and exhibits a weak γ transition at 66.73 keV, which can be used to deduce the activity of ^{171}Tm samples by γ spectroscopy using HPGe detectors. The first estimate of the emission probability I_γ of this γ line, with a value of $I_{66.7} = 0.14\%$, given with no uncertainty, dates back to 1964 [6]. Within this work of Hansen, ^{171}Tm has been produced by neutron irradiation of ^{170}Er and was separated by ion exchange chromatography. The sample was then measured with a NaI scintillation detector, a Xe-filled proportional counter and an x-ray escape spectrometer. The internal-conversion line spectrum and the β spectrum were additionally measured with a six-gap spectrometer. The γ-emission probability was then calculated based on assumed probabilities of K and L shell electron transitions during the ^{171}Tm decay.

Very recently, the 66.73 keV γ-line intensity has been measured to be $(0.144 \pm 0.010)\%$ using neutron activation of enriched ^{170}Er [7]. Within this work, the number of ^{171}Tm atoms was determined by following the decay of ^{171}Er, subsequently deducing the 66.73 keV photon emission probability using γ spectrometry.

The knowledge of the exact emission probability and its uncertainty plays a crucial role in experiments using this isotope, where the quantification of ^{171}Tm is based on γ spectrometry only [5,8–9]. Complementary to parallel efforts described in Ref. [7], we report here on the redetermination of $I_{66.7}$ providing an uncertainty analysis following the recommendations of the "guide to the expression of uncertainty in measurement" (GUM) [10]. The determination of the emission probability of the 66.73 keV γ line of ^{171}Tm was based on measuring the number of photons emitted from a quantified mass of ^{171}Tm. This task was accomplished by using a combination of γ spectrometry and mass spectrometry with radioisotopically pure solutions of ^{171}Tm.

All uncertainties are calculated according to GUM [10] and are quoted with a coverage factor $k = 1$, i.e., confidence level of about 68%, if not stated otherwise. The combined standard uncertainties are given in brackets in units of last significant digit.

II. EXPERIMENTAL

Radioisotopically pure ^{171}Tm was taken from leftovers of an earlier experiment performed in 2014 [4], where 140 GBq of ^{171}Tm were produced and separated from 250 mg of 98.1% enriched ^{170}Er [5]. The last separation of Tm from Yb for the sample used within this work was performed on 01.11.2014. The total content of remaining Yb was measured using multicollector high-resolution inductively coupled plasma mass spectrometry (ICP-MS) to be only 0.017% [5]. The specific activity of the ^{171}Tm solution was approximately 200 MBq/mL as of August 2017 [5]. An aliquot of 20 µL of this solution was diluted with 1 mL of MilliQ water (resistivity 18.2 MΩ cm at 25 °C) provided by an in-house water purification system.

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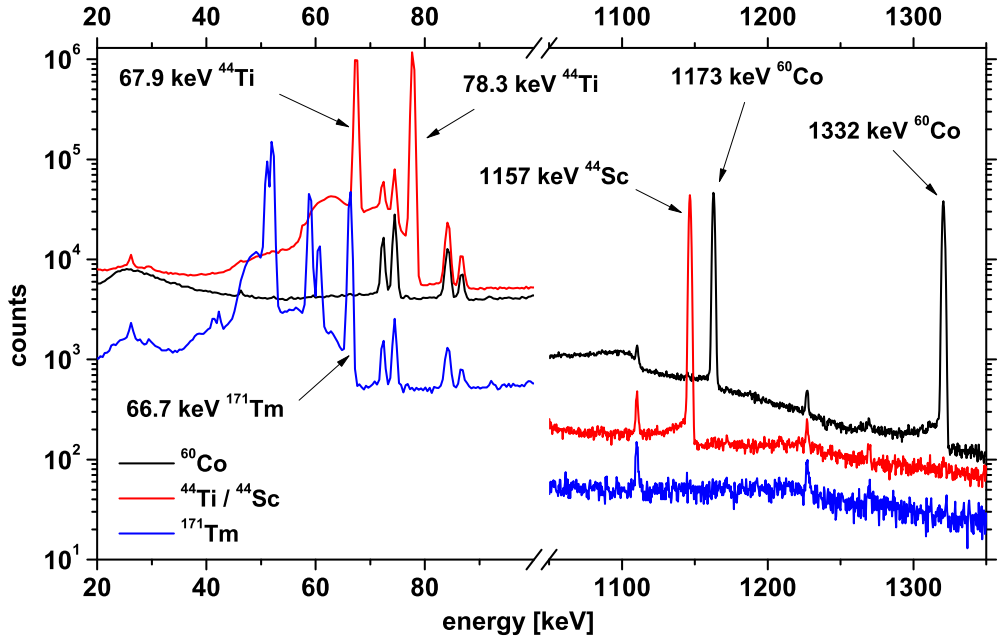

FIG. 1. Superimposed γ spectra of the ^{60}Co, ^{44}Ti/^{44}Sc, and ^{171}Tm samples.

Three different ^{171}Tm pointlike samples were prepared from this solution by evaporating one droplet ($\sim$20 μL) on circular Teflon supports. A Mettler-Toledo AT261 DeltaRange (last significant digit 0.01 mg, certified uncertainty 0.1475 mg by a total load of 51.25000 g by DKD-Kalibrationslaboratorium at 17.12.2014) balance was used to gravimetrically trace all dilution and sample preparation steps.

The total mass contribution of the 171 isobar, i.e., the abundance of ^{171}Tm and ^{171}Yb in the used solution, was determined by ICP-MS using an Element 2, Thermo Fischer Scientific, Bremen, Germany, operated in low-resolution mode and wet plasma conditions. The quantification was performed by the standard addition method [11] using a 10.0(2) mg/L ($k = 2$) Yb reference solution provided by Element Scientific, Omaha, USA.

For γ spectrometry, a ^{60}Co pointlike source served as primary reference standard with an activity of 1831(13) Bq certified with $k = 2$ on 01.07.2011 by Physikalisch-Technische-Bundesanstalt (PTB), Braunschweig, Germany. A secondary ^{44}Ti/^{44}Sc pointlike calibration source was prepared from a purified stock solution available from earlier experiments [12]. An aliquot of this stock solution was diluted with MilliQ water to yield a solution containing approximately 50 kBq/mL of ^{44}Ti/^{44}Sc. From this dilution, a secondary ^{44}Ti reference standard was prepared by evaporating 20 µL of the solution on a Teflon support. This source and the ^{171}Tm samples were sealed using a 50 µm thick Kapton tape.

Both reference sources and the ^{171}Tm samples were measured at a distance of 140(1) mm in air on a 1.2 mm thick Al support in front of a Canberra BE2825 planar HPGe detector for at least 21 h to achieve not less than 65000 net counts in the respective full energy peak (FEP) regions of interest. The primary standard (^{60}Co) was measured five times, while the ^{44}Ti/^{44}Sc standard and each ^{171}Tm sample were measured three times. The dead time of the counting setup was always below 0.5%. The GENIE2000 software, CANBERRA, was used to record the spectra and evaluate the area of the FEPs. The energy calibration of the detector was performed by a mixed nuclide standard provided by the Czech metrology institute, Brno, Czech Republic, containing ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y with certified activities. True coincidence summing effects for the measured geometry at 140 mm distance were calculated utilizing the EFFTRAN code [13]. True coincidence summing correction factors (ranging between: 1.00–1.01) were taken into account for all measured nuclides. Background measurements revealed no interfering peaks in the regions of interest. Due to the close proximity of the γ line of ^{171}Tm to the ones of ^{44}Ti, no correction for attenuation effects in air, aluminium, or Teflon was performed. Attenuation effects of the high-energy lines of ^{44}Sc and ^{60}Co were considered as negligible. Values for efficiencies as well as the γ-line emission probability of ^{171}Tm were obtained from the average of replicated measurements.

III. RESULTS

The measured γ spectra of the ^{60}Co reference source, the ^{44}Ti/^{44}Sc and ^{171}Tm samples are given in Fig. 1. Applying decay correction with respect to the center of the counting interval for the measured primary ^{60}Co calibration source, the FEP efficiencies at 1173.23 keV and 1332.49 keV ($I_{1173} = 99.85(3)\%$ and $I_{1332} = 99.9826(6)\%$ [14]) were determined to be $\varepsilon_{1173} = 0.0666(3)\%$ and $\varepsilon_{1332} = 0.0588(2)\%$, respectively, for the measured geometry.

The FEP efficiency ε_x and its relative uncertainty $\Delta\varepsilon_x/\varepsilon_x$ of a given detection system at an energy E_x is extrapolated from two known FEP efficiencies ε_1 and ε_2 measured at energies E_1 and E_2, where $E_x < E_1 < E_2$, assuming a general power law

using the following Eqs. (1) and (2):[1]

$$\varepsilon_x = \varepsilon_1 \cdot \left(\frac{E_x}{E_1}\right)^{\alpha} = \varepsilon_1 \cdot \left(\frac{\varepsilon_2}{\varepsilon_1}\right)^{\beta}, \quad (1)$$

$$\left(\frac{\Delta\varepsilon_x}{\varepsilon_x}\right)^2 = (1-\beta^2)\cdot\left(\frac{\Delta\varepsilon_1}{\varepsilon_1}\right)^2 + \beta^2\cdot\left(\frac{\Delta\varepsilon_2}{\varepsilon_2}\right)^2, \quad (2)$$

where α and β are defined as

$$\alpha = \frac{\ln\left(\frac{\varepsilon_2}{\varepsilon_1}\right)}{\ln\left(\frac{E_2}{E_1}\right)} \qquad \beta = \frac{\ln\left(\frac{E_x}{E_1}\right)}{\ln\left(\frac{E_2}{E_1}\right)}.$$

With the above equations, the FEP efficiency at 1157.02 keV was determined using the FEP efficiencies of both ^{60}Co lines to be $\varepsilon_{1157} = 0.0676(3)\%$. Subsequently, the activity of ^{44}Sc was determined by measuring the absolute count rate of its 1157.02 keV γ line ($I_{1157} = 99.875(3)\%$ [16]) to be 1286(6) Bq on reference date 09/22/2017. Due to secular equilibrium between ^{44}Ti and its decay product ^{44}Sc, the activities of both isotopes are equal.

In the next step, the FEP efficiencies of the γ detector at both 67.87 keV ($I_{67.9} = 93.0(20)\%$ [16]) and 78.32 keV ($I_{78.3} = 96.4(17)\%$ [16]) γ lines of ^{44}Ti were determined using the secondary ^{44}Ti standard. The FEP efficiency at these energies were $\varepsilon_{67.9} = 0.778(17)\%$ and $\varepsilon_{78.3} = 0.785(14)\%$, respectively. In analogy to the extrapolation of the efficiency at 1157 keV, the FEP efficiency at 66.73 keV was determined to be $\varepsilon_{66.7} = 0.777(19)\%$ by using Eqs. (1) and (2). Measurements of ^{171}Tm revealed the absolute γ-emission rate of the 66.73 keV line of ^{171}Tm emitted from each prepared sample.

Finally, the concentration of the 171 isobar isotopes, i.e., the content of ^{171}Tm and ^{171}Yb was measured by ICP-MS to be 260.5(28) ng/g in the prepared solution. With the known amounts of evaporated solution used to prepare the ^{171}Tm sources, the last Tm/Yb separation date 01.11.2014 and the ^{171}Tm half-life, we compute the 66.73 keV emission probability to be $\boldsymbol{I_{66.7} = 0.159(5)\%}$.

The contributors to the uncertainty of this value are deduced by a sensitivity analysis and shown in Table I. The main contributors to combined uncertainty originated from the γ-emission probabilities of the ^{44}Ti γ lines and the Yb standard reference solution. Other contributions from systematic uncertainties originating from literature data on half-lives or branching ratios of the utilized radionuclides as well as the contribution from the counting statistics are comparatively small. The uncertainty of the ^{171}Tm activity determination originating from an incomplete separation Tm/Yb was evaluated to be below 0.1%. A detailed description of the data evaluation and uncertainty propagation can be found in the Supplemental Material [17].

TABLE I. Sensitivity coefficients of involved quantities types to the uncertainty of the determined emission probability of the 66.73 keV line of ^{171}Tm.

Uncertainty Source	Sensitivity coefficient (%)
γ-emission probabilities	71.9[a]
Yb reference solution (ICP-MS)	12.5
Type A (counting statistics)	7.6
Half-lives	6.4[b]
^{60}Co reference standard	1.6

[a]Dominated by the uncertainty of the emission probability of the 67.87 keV line of ^{44}Ti.
[b]Dominated by the uncertainty of the ^{171}Tm half-life.

IV. CONCLUSIONS

The photon emission probability of 66.73 keV γ line of ^{171}Tm was measured by means of γ spectrometry combined with ICP-MS using radioisotopically pure ^{171}Tm samples. The uncertainty of the measured value was determined using standard uncertainty propagation following the recommendations of GUM, whereas the main uncertainty contributor was originating from the accepted literature value of the emission probability of the 67.87 keV ^{44}Ti γ line.

In comparison with the previously performed determinations of this quantity with a value of $I_{66.7} = 0.14\%$ by Hansen [6] and $I_{66.7} = 0.144(10)\%$ by [7], our result of $I_{66.7} = 0.159(5)\%$ is significantly higher. The here presented value is by more than 3σ with respect to the given uncertainty away from both previously determined ^{171}Tm emission probabilities. Therefore, the number of target atoms in the recently performed neutron capture experiments [4] will be consequently reduced thus enhancing the measured cross sections by approximately 14%.

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[1] This procedure uses a power-law regression between two reference energies and is applied for extrapolations to energies lying in the vicinity of reference data points. For the present work the extrapolated energies are within a margin of 2% off from the closest reference energy and the approximations as well as their uncertainty are thus considered as acceptable. Refer to Ref. [15] for more details about the physical justification of this approach.

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