

Home Search Collections Journals About Contact us My IOPscience

Discovery of the $^{151}\text{Eu}\,\alpha$ decay

This content has been downloaded from IOPscience. Please scroll down to see the full text. 2014 J. Phys. G: Nucl. Part. Phys. 41 075101 (http://iopscience.iop.org/0954-3899/41/7/075101) View the table of contents for this issue, or go to the journal homepage for more

Download details:

IP Address: 141.108.25.109 This content was downloaded on 29/01/2015 at 14:49

Please note that terms and conditions apply.

J. Phys. G: Nucl. Part. Phys. 41 (2014) 075101 (8pp)

```
doi:10.1088/0954-3899/41/7/075101
```

Discovery of the ¹⁵¹Eu α decay

N Casali^{1,2}, S S Nagorny^{1,3}, F Orio⁴, L Pattavina¹, J W Beeman⁵, F Bellini^{4,6}, L Cardani^{4,6}, I Dafinei⁴, S Di Domizio⁷, M L Di Vacri¹, L Gironi^{8,9}, M B Kosmyna¹⁰, B P Nazarenko¹⁰, S Nisi¹, G Pessina⁹, G Piperno^{4,6}, S Pirro⁹, C Rusconi⁹, A N Shekhovtsov¹⁰, C Tomei⁴ and M Vignati⁴

¹ INFN - Laboratori Nazionali del Gran Sasso, I-67010 Assergi (AQ), Italy

² Dipartimento di Scienze Fisiche e Chimiche, Università degli Studi dell'Aquila, I-67100 Coppito (AQ), Italy

³ Institute for Nuclear Research, National Academy of Sciences of Ukraine, 03680 Kyiv, Ukraine

⁴ INFN - Sezione di Roma, I-00185 Roma, Italy

⁵ Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

⁶ Dipartimento di Fisica - Università di Roma La Sapienza, I-00185 Roma, Italy

⁷ Dipartimento di Fisica, Università di Genova and INFN - Sezione di Genova,

I-16146 Genova, Italy

⁸ Dipartimento di Fisica, Università di Milano-Bicocca, Milano I-20126, Italy

⁹ INFN - Sezione di Milano Bicocca, Milano I-20126, Italy

¹⁰ Institute for Single Crystals, National Academy of Sciences of Ukraine, 61001 Kharkov, Ukraine

E-mail: luca.pattavina@lngs.infn.it

Received 9 January 2014, revised 6 March 2014 Accepted for publication 10 March 2014 Published 11 April 2014

Abstract

We report on the first compelling observation of α decay of ¹⁵¹Eu to the ground state of ¹⁴⁷Pm. The measurement was performed using a 6.15 g Li₆Eu(BO₃)₃ crystal operated as a scintillating bolometer. The *Q*-value and half-life measured are: $Q = 1948.9 \pm 6.9$ (stat.) ± 5.1 (syst.) keV, and $T_{1/2} = (4.62 \pm 0.95$ (stat.) ± 0.68 (syst.)) $\times 10^{18}$ y. The half-life prediction of nuclear theory using the Coulomb and proximity potential model are in good agreement with this experimental result.

Keywords: alpha decay, scintillation detectors, bolometers

(Some figures may appear in colour only in the online journal)

1. Introduction

All europium isotopes with mass number ranging from 130 to 153 a.m.u. are potentially unstable nuclides. They can disintegrate through a number of channels, including the α decay [1, 2]. However, competing large branching ratio for electron capture and β^- decay process make α decay amenable for investigation only for artificially produced ^{147,148,150}Eu and naturally occurring ¹⁵¹Eu and ¹⁵³Eu.

Modern calculations of the partial half-lives of 147,148,150,151,153 Eu mainly follow two approaches, namely the semi-empirical one [3], developed in [4] and the Coulomb and proximity potential model (CPPM) [5], proposed in [6]. The results obtained by the two approaches for the half-life of 151 Eu range between 8.5×10^{18} y [3] using semi-empirical calculations and 8.0×10^{17} y [5] using CPPM for deformed nuclei (CPPMDN). Given this order of magnitude incompatibility in the theoretical predictions, experimental investigation of such α decays is needed in order to establish which nuclear model better describes these nuclei.

An interesting side-application of observing the α decay of ¹⁵¹Eu and measuring its halflife is the impact on the evaluation of promethium concentration in the entire Earth crust [7]. In fact, one expected promethium production mechanism, besides spontaneous fission of ²³⁸U, is α decay of natural ¹⁵¹Eu to the ground state of ¹⁴⁷Pm.

2. State of the art

A new era of sensitivity to rare α decays started in 2003 with the application of the scintillating bolometer technique to observe the α decay of ²⁰⁹Bi with a half-life T_{1/2} =1.9 × 10¹⁹ y [8]. Since then this technique has been successfully exploited to detect α decay of ¹⁸⁰W [9], α decay of ²⁰⁹Bi to the first excited level [10] and to search for α decays of lead isotopes [11]—demonstrating the versatility and power of this technology.

A key advantage of bolometers is the wide choice of detector materials, the detector can often be composed of a significant amount of the nucleus of interest. This results in excellent detection efficiency—the detector is made of the decay source—which is vital to investigate rare decays. Moreover, high energy resolution (of the order of parts per thousand [12]) is routinely achieved exploiting this technology. When the bolometer is also an efficient scintillator then the simultaneous read-out of the heat and light channels enables the identification of the interacting particle nature, i.e. whether it is an α , β/γ , or neutron. This leads to tremendous background suppression, especially for rare α decays with energy transition lower than 2.6 MeV which would otherwise be overwhelmed by the near omnipresent background from γ emission of ²⁰⁸Tl.

Natural europium consists of two isotopes: ¹⁵¹Eu (47.81%) and ¹⁵³Eu (52.19%) [13]. Both are potentially α -active with decay energies of $Q_{\alpha} = 1965.0 \pm 1.1$ keV and $Q_{\alpha} = 272.4 \pm 2.0$ keV [1, 2], respectively. The low Q_{α} for ¹⁵³Eu gives no hope for an experimental observation due to the extremely long expected half-life, more than 10¹⁴⁰ y [14]. On the other hand theoretical predictions of the ¹⁵¹Eu half-life lie in range of 10¹⁷–10¹⁹ y [14], and such sensitivity levels can be achieved by scintillating bolometers.

The α decay of europium isotopes has been recently investigated using a 370 g CaF₂:Eu low background scintillator [14], which contained Eu only as a dopant with mass fraction of just 0.5%. The authors of that paper mention the first indication of the α decay of ¹⁵¹Eu nuclide to the ground state of ¹⁴⁷Pm with a half-life of T_{1/2} = 5⁺¹¹₋₃ × 10¹⁸ y. Nevertheless, due to the large statistical uncertainty, the authors report also a limit on the half-life of the process: T_{1/2} > 1.7 × 10¹⁸ y at 68% C.L.

In this work we report the first compelling experimental observation of the α decay of ¹⁵¹Eu to the ground state of ¹⁴⁷Pm, using a 6.15 g Li₆Eu(BO₃)₃ crystal (41.1% of Eu in mass) operated as scintillating bolometer.

3. Experimental technique

The crystal was grown using the Czochralski technique in a platinum crucible under air atmosphere in accordance with the thermal conditions described in [15]. The stoichiometric mixture of initial reagents (Li_2CO_3 , Eu_2O_3 and B_2O_3) was used to synthesize the $Li_6Eu(BO_3)_3$ charge by solid phase synthesis as described in [16].

The crystal used in this work has an irregular shape due to the compromise of having the largest possible mass combined with an acceptable crystal quality. For this reason part of the crystal shows unpolished rounded edges. Moreover, the crystal shows some small cleavage planes as well as some tiny inclusions in part of the structure. Our typical experience with bolometers is that this can negatively affect the energy resolution.

The working principle of a scintillating bolometer consists of measuring the temperature rise produced by the interaction of particles with the crystal absorber, which is operated at a temperature of a few mK. When an ionizing particle deposits energy in the crystal, a large portion of it is converted to heat, while some is converted to scintillation light which is detected by another bolometer facing the crystal. The heat and light signals are both recorded as positive temperature variations by temperature sensors on the main absorber and on the LD. For the same energy deposition, α and β/γ particles will generate different heat-to-light ratios.

The Li₆Eu(BO₃)₃ crystal was operated in a dilution ³He/⁴He refrigerator in the Gran Sasso underground laboratory of INFN. The crystal was held in position by means of a high-purity copper structure. Germanium neutron transmutation doped thermistors (Ge-NTD) were used as temperature sensors. The Ge-NTDs were coupled to the Li₆Eu(BO₃)₃ crystal and to the light detector (LD) by means of resin epoxy glue. In order to maximize the light collection efficiency, the crystal was surrounded by a reflecting foil (3M VM2002). The operational features of the LD, namely a high-purity Ge wafer (diameter 44.5 mm and thickness 300 μ m), are described in [17].

4. Data analysis

The detector was operated for 462.2 h. The $Li_6Eu(BO_3)_3$ crystal and the LD have completely independent read-out. Detailed information on the cryogenic facility electronics, the data acquisition and processing can be found in [18–20].

The amplitude of the heat and light signals is energy-calibrated by means of calibration sources of known energies. The scintillation light is calibrated using a permanent ⁵⁵Fe x-ray source facing the LD, we assume a linear dependence of the signal amplitude on the light energy. The heat channel is calibrated using the Q_{α} -value of the α decays from internal contamination in the crystal. The calibration curve used is a second order polynomial with zero intercept.

A useful quantity to consider is the light yield (LY), which we define as the ratio of the energy measured in the LD in keV to the energy measured in the heat channel of the main absorber in units of MeV. Figure 1 shows LY as a function of the heat-energy measured in the main crystal for each event in the 462.2 hour exposure. The β/γ and the α interactions in the detector occupy well separated regions of the LY versus heat-energy plan in agreement with expectation from Birks law, where α interactions produce lower light signals than those



Figure 1. Light yield versus heat-energy scatter plot corresponding to 462.2 hours of background measurement with the 6.15 g $\text{Li}_6\text{Eu}(\text{BO}_3)_3$ crystal. Three classes of events are identified, each one is characterized by a different light yield. Colours are used to highlight the α and the dark counts band.

of $\beta s/\gamma s$ [21]. In the lower energy region of the scatter plot there is a continuum of events that do not produce light. These so-called dark counts can be induced by several mechanisms. For example, defects such as those present in our crystal can give rise to points in which the scintillation process is strongly reduced. Furthermore, an imperfect crystal in which the structure is stressed by an inhomogeneities can give rise to sudden lattice relaxations, resulting in thermal pulses without the emission of photons. Finally, considering the ratio of the volumes of the Ge-NTD thermistor and the scintillating crystal—roughly 1:600—a fraction of dark counts will arise from interactions that release energy only inside the thermometer thereby producing a thermal signal without photon emission. In order to reject these events that produce an almost flat background in the region of interest, a cut on the absolute light signal is performed.

4.1. Alpha energy spectrum

As is clear from figure 1, it is possible to reject β/γ events by performing a selection on the heat-to-light ratio. The choice LY < 3.3 keV MeV⁻¹ has rejection efficiency of (99.99 ± 0.01)%. To exclude dark counts, we select events with detected light higher than 0.4 keV. This cut has an efficiency of (96.8 ± 0.2)%. The energy spectrum of the remaining events is shown in figure 2. The most prominent peak in the energy spectrum is due to an internal contamination of Sm, namely ¹⁴⁷Sm with an activity of 454 mBq kg⁻¹. The events in the high energy region, above 4 MeV, have well understood sources: ²³²Th decay daughters present at a level of 3.5 mBq kg⁻¹; and ²³⁸U daughters particularly ²²⁶Ra and ²¹⁰Po, present at the level of 2.9 mBq kg⁻¹ and 6.2 mBq kg⁻¹, respectively. The most intense peaks namely ¹⁴⁷Sm (2311 keV), ²³²Th (4083 keV) and ²²⁶Ra (4871 keV) were used for the calibration of the α energy scale. The error on the energy-calibration function at 2311 keV is less than 1 keV.

Looking at the spectrum in figure 2, to the left side of the ¹⁴⁷Sm line we observe a clear peak in the 1900–2000 keV window. There are three known α active nuclides with energy transitions that would reconstruct close to the position of the observed peak. They are ¹⁴⁴Nd $(Q = 1905 \pm 1 \text{ keV}, T_{1/2} = 2.3 \times 10^{15} \text{ y})$, ¹⁴⁸Sm $(Q = 1986 \pm 1 \text{ keV}, T_{1/2} = 7.0 \times 10^{15} \text{ y})$ and ¹⁵²Gd $(Q = 2203.0 \pm 1.4 \text{ keV}, T_{1/2} = 1.1 \times 10^{14} \text{ y})$. The concentration of these elements in the



Figure 2. Energy spectrum for alpha events selected in the 462.2 h exposure of the 6.15 g $\text{Li}_6\text{Eu}(\text{BO}_3)_3$ crystal. The events are attributed to internal contamination of the crystal, the most intense one is ¹⁴⁷Sm α decay at 2311 keV, while the peaks above 4 MeV are due to α decays of daughter nuclides from ²³⁸U/²³²Th chains.

crystal was determined using a high resolution inductively coupled plasma-mass spectrometric (HR-ICP-MS) analysis. The obtained results are 4.9×10^{-6} g g⁻¹, 4.7×10^{-6} g g⁻¹ and 91.3×10^{-6} g g⁻¹ for elemental Nd, Sm and Gd. Thus the contribution of the afore mentioned isotopes to the peak are independently constrained to be only 0.5, 0.1 and 1.5 events.

The robustness of the HR-ICP-MS results is further confirmed by the fact that the measured concentration of the elemental Sm, scaled for the natural isotopic abundance of ¹⁴⁷Sm, agrees with the activity measured with our spectrum analysis within an the error of 15%.

Given that known α decays are independently constrained to contribute negligibly to this peak, we interpret it as being due to the α decay of ¹⁵¹Eu.

4.2. Response function and discovery significance

We use a crystal ball (CB) function to fit the response of the Li₆Eu(BO₃)₃ bolometer to α particles. We find that the response function (RF) that best describes the ¹⁴⁷Sm peak is the sum of two crystal balls [22] with the same mean (*Q*-value), one with a left tail and one with a right tail: RF(*Q*, *E*, δ) = $N \cdot [CB_{left}(Q, E) + \delta \cdot CB_{right}(Q, E)]$. Here *N* is an arbitrary normalisation and δ is the ratio between the two CB functions. The shape of the RF evaluated on the ¹⁴⁷Sm peak is shown in figure 3.

With this RF of the bolometer for α particles we perform an unbinned, extended likelihood fit to the α energy spectrum in order to evaluate the number of ¹⁵¹Eu events. The fit function (FF) used is:

$$FF(E) = N_{Sm} \cdot RF(Q_{Sm}, E, \delta) + N_{Eu} \cdot RF(Q_{Eu}, E, \delta).$$
(1)

 $N_{\rm Sm}$ and $N_{\rm Eu}$ are the number of events in the ¹⁴⁷Sm and ¹⁵¹Eu peaks and are free parameters of the fit. The ¹⁴⁷Sm *Q*-value is constrained to its established value [1, 2], while the *Q*-value of ¹⁵¹Eu is left as a free parameter.

In figure 4, the best-fit α energy spectrum from 1.2 to 4 MeV is shown. The best-fit number of ¹⁵¹Eu events (N_{Eu}) corrected for the event selection efficiency, evaluated on the ¹⁴⁷Sm peak to be (96.8 ± 0.2)%, is equal to 37.6 ± 7.5 counts. The best-fit *Q*-value for the ¹⁵¹Eu α transition is 1948.9±6.9(stat.) keV, and the FWHM energy resolution is 65 ± 7 keV.



Figure 3. Normalized response function of the detector used for α particles interacting in the Li₆Eu(BO₃)₃ crystal. The two dashed lines represent the left and right crystal ball functions, while the solid line is their sum.



Figure 4. Zoom of the α energy spectrum around the region of interest. Two peaks are visible: ¹⁴⁷Sm at 2311 keV and one at 1949 keV, interpreted as ¹⁵¹Eu. In the inset the full y-scale plot is shown. The response function of the detector is assumed to be the sum of two crystal ball functions and this is used for fitting the two observed peaks in the α energy spectrum. The dashed lines represent the detector response function, while the solid line is the sum of the response function of the two peaks.

In order to quantify the statistical significance of the excess over the background-only expectation from ¹⁴⁷Sm we adopt the same test used in [23]. Following that reference, we define

$$q_0 = -2 \cdot \ln \frac{L(\text{data} \mid \text{bkg}(\hat{\theta}_0))}{L(\text{data} \mid \hat{\mu} \cdot \text{signal}(\hat{\theta}) + \text{bkg}(\hat{\theta}))}, \qquad (2)$$

where $\hat{\theta}_0$ is the set of fit parameter values that maximize the likelihood in the background-only hypothesis, $\hat{\theta}$ is the set of fit parameter values that maximize the likelihood in the background plus signal hypothesis, and $\hat{\mu}$ is the number of signal events that maximizes the likelihood. Cases where a fit finds a positive signal, i.e. $\mu > 0$, correspond to positive values of q_0 , while in the absence of a signal, $\mu = 0$, q_0 is zero. For our fit, the excess of events that we observed gives $q_0 = 54$, corresponding to a 7.4 σ statistical significance. Therefore the probability that the measured excess of events is produced by a background fluctuation is of the order of 10^{-14} .

5. ¹⁵¹Eu half-life

The number of ¹⁵¹Eu atoms in the 6.15 g Li₆Eu(BO₃)₃ crystal is $(4.76 \pm 0.07) \times 10^{21}$, evaluated using the isotope composition of Eu precisely measured with HR-ICP-MS $(47.6 \pm 0.7)\%$.

To estimate the systematic uncertainty from the choice of fit interval and FF, we varied both the fitting interval and RF used. For example, we find shrinking the fit range from (1200–4000 keV) to (1800–2800 keV) reduces the best-fit number of ¹⁵¹Eu events by 5.3 counts, while changing the RF to a CB plus a Gaussian increases the number of ¹⁵¹Eu events by 0.9 counts. We include these effects in the systematic uncertainty. Finally, we do not consider the systematic uncertainty arising from the small calibration error (<0.1% at 2311 keV) because it is negligible compared to the detector energy resolution.

Considering the best-fit number of ¹⁵¹Eu events corrected for the event selection efficiency (37.6 \pm 7.5 counts), the containment efficiency of ¹⁵¹Eu α decay evaluated using Monte Carlo simulation (ϵ = 99.98%), and the live time of the measurement (462.2 h), we obtain the following value for the half-life for α decay of ¹⁵¹Eu to the ground state of ¹⁴⁷Pm:

$$T_{1/2} = (4.62 \pm 0.95(\text{stat.}) \pm 0.68(\text{syst.})) \times 10^{18} \text{ y.}$$
 (3)

This value is in agreement with half-life interpreted from the indication of ¹⁵¹Eu α decay reported in [14]. Our measurement concurs, within 1 σ with the theoretical prediction using the CPPM reported in [5], and is not compatible with the semi-empirical calculation reported in [3, 24, 25].

6. Conclusion

In this work we reported the discovery of ¹⁵¹Eu α decay to ground state of ¹⁴⁷Pm with a statistical significance of 7.4 σ . The measured half-life is $T_{1/2} = (4.62 \pm 0.95(\text{stat.}) \pm 0.68(\text{syst.})) \times 10^{18}$ y.

Our result is extremely relevant to identify the best-performing nuclear models for favoured α decays of odd-mass nuclei. In fact, among the different nuclear models previously mentioned in this work, the one in best agreement with our measurement is the Coulomb and proximity potential model (CPPM), where the penetrability potential was evaluated both from the internal and external parts of the nuclear barrier. The computed theoretical value for ¹⁵¹Eu half-life is: 4.8×10^{18} y, which is less than 1 σ away from our experimental value.

The measured *Q*-value for the decay is $1948.9 \pm 6.9(\text{stat.}) \pm 5.1$ (syst.) keV, in which the statistical error is caused by the small ¹⁵¹Eu statistics, while the systematic by varying both the fitting interval and RF used. Our result is compatible with the one reported in [1, 2] within 2σ interval. In order to better understand and reduce this discrepancy, further measurements with larger source mass and larger statistics are needed.

The discovery of ¹⁵¹Eu α decay to the ground state of ¹⁴⁷Pm also plays a relevant role in the evaluation of the overall mass of promethium in the Earth's crust. Following the calculation reported in [14], we compute the total mass of ¹⁴⁷Pm in the Earth crust to be about 565 g.

Acknowledgments

This project was supported by the Italian Ministry of Research under the PRIN 2010ZXAZK9 2010-2011 grant. This work was also supported by the ISOTTA project, funded within the ASPERA 2nd Common Call for R&D Activities. Part of the work was carried out thanks to LUCIFER Project, funded by the European Research Council (FP7/2007-2013) grant agreement no 247115.

We would like to express special gratitude to Thomas O'Donnell for his careful and accurate revision of the manuscript. Thanks are also due to the LNGS mechanical workshop and in particular to E Tatananni, A Rotilio, A Corsi, and B Romualdi for continuous and constructive help in the overall set-up construction. Finally, we are especially grateful to M Perego and M Guetti for their invaluable help.

References

- [1] Audi G et al 2012 Chin. Phys. C 36 1287
- [2] Wang M et al 2012 Chin. Phys. C 36 1603
- [3] Tavares O A P and Medeiros E L 2007 Phys. Scr. 76 163
- [4] Tavares O A P et al 2006 Nucl. Instrum. Methods Phys. Res. B 243 256
- [5] Santhosh K P et al 2013 Int. J. Mod. Phys. E 22 1350081
- [6] Santhosh K P et al 2008 J. Phys. G: Nucl. Part. Phys. 35 085102
- [7] Attrep M et al 1968 J. Inorg. Nucl. Chem. 3 699703
- [8] de Marcillac P et al 2003 Nature 422 876
- [9] Cozzini C et al 2004 Phys. Rev. C 70 064606
- [10] Beeman J W et al 2012 Phys. Rev. Lett. 108 062501
- [11] Beeman J W et al 2013 Eur. Phys. J. A 49 50
- [12] Beeman J W et al 2012 Eur. Phys. J. C 72 2142
- [13] Berglund M and Wieser M E 2011 Pure Appl. Chem. 83 397
- [14] Belli P et al 2007 Nucl. Phys. A 789 15
- [15] Baumer V N et al 2002 J. Cryst. Growth 242 167
- [16] Grinyov B V 2003 Patent 66072 UA
- [17] Beeman J W et al 2013 J. Instrum. 8 P07021
- [18] Pirro S et al 2000 Nucl. Instrum. Methods A 444 331
- [19] Arnaboldi C et al 2002 Nucl. Instrum. Methods A 520 578
- [20] Piperno G et al 2011 J. Instrum. 6 P10005
- [21] Birks J B 1951 Proc. Phys. Soc. A 64 874
- [22] Gaiser J E 1982 Report p 178 SLAC-R-255
- [23] CMS Collaboration 2012 Phys. Lett. B 710 26
- [24] Poenaru D N and Ivascu M 1983 J. Phys. 44 791
- [25] Poenaru D N et al 1985 Phys. Rev. C 32 2198