Search for $\alpha$ decay of $^{151}$Eu to the first excited level of $^{147}$Pm using underground $\gamma$-ray spectrometry

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Abstract. The alpha decay of $^{151}$Eu to the first excited level of $^{147}$Pm ($J^\pi = 5/2^+$, $E_{\text{exc}} = 91.1$ keV) was searched for at the HADES underground laboratory ($\approx 500$ m w.e.). A sample of high-purity europium oxide with mass of $303\,$g and a natural isotopic composition has been measured over $2232.8$ h with a high energy resolution ultra-low background n-type semi-planar HPGe detector ($40\,$cm$^2$) with sub-micron deadlayer. The new improved half-life limit has been set as $T_{1/2} \geq 3.7 \times 10^{18}$ y at $68\%$ CL. Possibilities to improve the sensitivity of the experiment, which is already near the theoretical predictions, are discussed. New half-life limit for $\alpha$ decay of $^{153}$Eu is also determined as $T_{1/2} \geq 5.5 \times 10^{17}$ y.

1 Introduction

Natural europium consists of only two isotopes, $^{151}$Eu and $^{153}$Eu, with the natural abundances of $47.81(0.06)\%$ and $52.19(0.06)\%$, respectively [1]. Both isotopes are potentially active with an $\alpha$ decay energy $Q_\alpha = 1964.9(1.1)$ keV and $Q_\alpha = 272.5(2.0)$ keV, respectively [2,3]. The first indication on $\alpha$ decay of $^{151}$Eu to the ground state of $^{147}$Pm with the half-life $T_{1/2} = 5.3 \times 10^{18}$ y was obtained in [4] with the help of a CaF$_2$(Eu) low background scintillation detector.

Alpha decay of $^{151}$Eu is also energetically allowed to the excited levels of $^{147}$Pm, with a highest probability of transition to the first $91.1$ keV $5/2^+$ level (the decay scheme of $^{151}$Eu is shown in fig. 1). Theoretical estimations for this $\alpha$ transition (obtained by using different approaches [7–10]) are in the range of $7 \times 10^{18} - 1 \times 10^{20}$ y, which is at the level of present sensitivity accessible in low background experiments. Indeed, recently the $\alpha$ decay of $^{180}$Pt to the first excited level of $^{186}$Os with the half-life $2.6 \times 10^{14}$ y was observed by using an ultra-low background HPGe detector and a sample of platinum with the natural composition of isotopes [11], despite very low isotopic abundance of $^{180}$Pt ($0.012\%$). The decay $^{151}$Eu $\rightarrow$ $^{147}$Pm$^\ast$ seems to be a detectable process with the present experimental technique, taking into account more than three orders of magnitude higher isotopic abundance of $^{151}$Eu.

To our knowledge, there were two experimental attempts to detect the process. The limit $T_{1/2} \geq 2.4 \times 10^{16}$ y was set in the measurements with a HPGe detector with a small ($2.72\,$g) sample of Li$_6$Eu(BO$_3$)$_3$ crystal (containing only $1.1\,$g of Eu; the purpose of the experiment was to investigate radioactive contamination of the material) [12]. One order of magnitude higher limit $T_{1/2} \geq 6.0 \times 10^{17}$ y was set in the experiment with CaF$_2$(Eu) crystal scintillator [4].

One can also search for $\alpha$ decay of $^{153}$Eu to the ground state of $^{149}$Pm by using a $\gamma$-ray detector because of instability of the daughter $^{149}$Pm nucleus relative to $\beta$ decay.

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2 Materials and methods

The measurements were performed at the HADES underground facility, which is located at the premises of the Belgian nuclear centre (SCK-CEN) and operated by EURIDICE [14]. The depth of the laboratory is 225 m. The Belgian nuclear centre (SCK-CEN) and operated by EU-RIDICE [14]. The depth of the laboratory is 225 m. The

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| 153Eu | 153Eu |
|-------|-------|
| 5/2+ 0 | 7/2+ 0 |
| 188.6 | 114.3 |

Fig. 2. (Color online) Expected scheme of α decay of 153Eu. The energies of the levels and of the deexcitation γ quantum are given in keV [5,13]. Only the main branches of 149Pm β decay are shown.

with half-life T1/2 = 53.08 h and Qβ = 1071 keV (the decay scheme of 153Eu is shown in fig. 2). Up to date only the limit T1/2 ≥ 1.1 × 1016 y on α decay of 153Eu was set in [12].

The present paper describes a new search for α decay of 151Eu to the first excited level of 147Pm and α decay of 153Eu to the ground state of 149Pm by using a high-purity europium oxide sample installed on an ultra-low background HPGe γ detector.

2.1 Radioactive contamination of Eu2O3 sample

The comparison of the Eu2O3 and of the background spectra shows that the europium sample is contaminated by daughters of 232Th and 238U, and radioactive europium isotopes 152Eu, 154Eu and 155Eu. There are also peaks in the spectrum with the energies 201.7 ± 0.2 keV and 306.5 ± 0.2 keV, which indicate presence of 176Lu in the sample. Radioactive 152Eu and 154Eu nuclei were produced by neutron captures in 151Eu and 153Eu, respectively. 155Eu can be produced by the double neutron capture reaction 153Eu(n,γ) → 154Eu(n,γ) → 155Eu and also due to fission of uranium and thorium. Indeed, europium is extracted from minerals with typically high concentration of thorium and uranium: bastnaesite, loparite, xenotime, and monazite. There are no unidentified peaks in the spectrum.
Massic activities of the contaminant radionuclides \( A \) were calculated with the formula

\[
A = \frac{S_{\text{sample}}/t_{\text{sample}} - S_{\text{bg}}/t_{\text{bg}}}{(\vartheta \cdot \varepsilon \cdot m)},
\]

where \( S_{\text{sample}} \) (\( S_{\text{bg}} \)) is the area of a peak in the sample (background); \( t_{\text{sample}} \) (\( t_{\text{bg}} \)) is the time of the sample (background) measurement; \( \vartheta \) is the \( \gamma \)-ray emission probability [5]; \( \varepsilon \) is the efficiency of the full energy peak detection; \( m \) is the mass of the sample. The full energy peak efficiencies were calculated using the Monte Carlo code EGS4 [15]. A detailed model of the detector, sample and shield were used for the calculations. Coincidence summing corrections for cascading gamma-rays were also included in the simulation. The systematic uncertainty of the calculated efficiency is at the level of 5% as confirmed by several proficiency testing exercises. The systematic uncertainty of the detection efficiency calculation arising from the nonperfect cylindrical geometry of the sample (contained in a plastic bag) was estimated to be \( \approx 10\% \). Estimations of radioactive contamination of the sample from the 15.4 day measurement on Ge-5 were performed in the same way. The summary of radioactive contamination of the \( \text{Eu}_2\text{O}_3 \) sample is presented in table 1. The results of the independent measurements are in reasonable agreement, taking into account a systematic uncertainty due to the nonperfect cylinder shapes of the sample in both the measurements.

### 3.2 Limit on \( \alpha \) decay of \( ^{151}\text{Eu} \) to the first excited level of \( ^{147}\text{Pm} \)

Gamma quanta with an energy of 91.1 keV should be emitted after alpha decay of \( ^{151}\text{Eu} \) to the first excited level of \( ^{147}\text{Pm} \). There is no peak at energy 91 keV in the energy spectrum accumulated with the \( \text{Eu}_2\text{O}_3 \) sample (see fig. 4). Therefore we can only set a lower half-life limit \( (\lim T_{1/2}) \) on the effect according to the formula

\[
\lim T_{1/2} = \frac{N \cdot \varepsilon \cdot \vartheta \cdot t}{\ln 2} / \lim S,
\]

where \( N \) is the number of \( ^{151}\text{Eu} \) nuclei \( (4.96 \times 10^{23}) \), \( \varepsilon \) is the detection efficiency, \( \vartheta \) is the \( \gamma \) yield \( (\vartheta = 0.33 \) for the level 91.1 keV due to a high electron conversion coefficient of 2.03 [6]), \( t \) is the measuring time, and \( \lim S \) is the number of events of the effect searched for which can be excluded at a given confidence level (CL). The detection efficiency for 91.1 keV \( \gamma \) quanta emitted in the \( \text{Eu}_2\text{O}_3 \) sample was calculated with the EGS4 package as \( \varepsilon = 0.00434 \) (see sect. 3.1).

To estimate a value of \( \lim S \) for the 91.1 keV peak, the energy spectrum accumulated with the \( \text{Eu}_2\text{O}_3 \) sample was fitted in the energy region 88–95 keV. The model to fit the data was constructed from a Gaussian function with centre at the energy of 91.1 keV and the energy resolution FWHM = 0.749 keV (the peak searched for), a linear function which describes the background, and two Gaussians to take into account the neighbouring peaks with the
energies ≈ 90 keV (Th X-ray) and ≈ 92.6 keV (92.4 and 92.8 keV peaks of $^{234}$Th from the $^{238}$U chain).

A fit by the chisquare method ($\chi^2$/n.d.f. = 89/78 = 1.14, where n.d.f. is number of degrees of freedom) results in the area of the peak searched for $S = 7 \pm 27$ counts, which gives no evidence for the effect. In accordance with the Feldman-Cousins procedure [16], we took $\lim S = 34$ counts which can be excluded at 68% CL. Thus we obtained the following limit on $\alpha$ decay of $^{151}$Eu to the first 5/2$^+$ 91.1 keV excited level of $^{147}$Pm:

$T_{1/2}^{(151 \text{Eu} \rightarrow 147 \text{Pm} (5/2^+, 91.1 \text{keV})} \geq 3.7 \times 10^{18}$. \[1\]

The data collected with the Eu$_2$O$_3$ sample also allow searching for all possible $\alpha$ decays of $^{153}$Eu to $^{149}$Pm (including the most probable decay to the ground state) due to $\beta$ instability of the daughter isotope (see fig. 2). The most intense $\gamma$ line of $^{149}$Pm has an energy of 285.9 keV and a yield $\vartheta = 0.031$ [13]. The detection efficiency of the $\gamma$ quanta is $\varepsilon = 0.0114$. Part of the spectrum in the energy interval 278–298 keV is shown in fig. 5. There is no peculiarity in the spectrum accumulated with the europium oxide sample, which can be ascribed to the 285.9 keV $\gamma$ peak. However, there are two peculiarities (at 284.3 keV and 284.9 keV, see fig. 5) immediately to the left of the sought 285.9 keV peak with a somewhat higher number of counts compared to the surrounding channels. Calculations show that these “structures” are not peaks as they fall below the detection limit. We attribute them to counting statistical variations. A fit in the energy interval 282–290 keV by a model, consisting of the Gaussian function to describe the peak searched for plus polynomial function of the second degree to describe the background, gives an area of the effect searched for as 34 ± 27 events, which is no evidence for the effect (see fig. 5). Taking $\lim S = 61$ we set the following half-life limit on $\alpha$ decay of $^{153}$Eu to $^{149}$Pm at 68% CL:

$T_{1/2}^{(153 \text{Eu} \rightarrow 149 \text{Pm})} \geq 5.5 \times 10^{17}$. \[2\]
Table 2. Half-life limits on $\alpha$ decay of Eu isotopes (given at 68% CL; the half-life limits in work [12] are given at 90% CL) and comparison with the theoretical predictions. The values in columns 3–5 are presented in years.

| Alpha transition | Level of daughter nucleus | Experimental $T_{1/2}$ | Theoretical estimations |
|------------------|--------------------------|------------------------|------------------------|
| $^{151}\text{Eu}$ → $^{147}\text{Pm}$ | 5/2$^+$, 91.1 keV | $\geq 2.4 \times 10^{16}$ [12] | $9.7 \times 10^{16}$ \textsuperscript{(a)} |
| $^{151}\text{Eu}$ → $^{147}\text{Pm}$ | $\geq 6.0 \times 10^{17}$ [4] | $7.4 \times 10^{18}$ \textsuperscript{(b)} |
| $^{153}\text{Eu}$ → $^{149}\text{Pm}$ | 7/2$^+$, 0 keV (g.s.) | $\geq 1.1 \times 10^{16}$ [12] | $5.2 \times 10^{14}$ \textsuperscript{(a)} |
| $^{272.5}\text{(2.0) keV}$ | $\geq 5.5 \times 10^{17}$ | $2.8 \times 10^{14}$ \textsuperscript{(b)} |

\textsuperscript{(a)} Calculated with semiempirical formulae \[7\].
\textsuperscript{(b)} Calculated with the cluster model of refs. \[8,9\].
\textsuperscript{(c)} Calculated with the approach of ref. \[10\].

A summary of the obtained results in comparison with the previous studies is given in table 2.

4 Discussion

Half-life values for the $\alpha$ decays of the Eu isotopes were calculated here with the cluster model of refs. \[8,9\] and with semiempirical formulae \[7\] based on liquid-drop model and description of $\alpha$ decay as a very asymmetric fission process. The approaches were tested with a set of experimental half-lives of a few hundred $\alpha$ emitters and demonstrated good agreement between calculated and experimental $T_{1/2}$ values, mainly inside the factor of 2–3. We also successfully used these works to predict $T_{1/2}$ values of the long-living $\alpha$ active $^{180}\text{W}$ \[17\] and $^{151}\text{Eu}$ \[4\] obtaining adequate agreement between the first experimentally measured and calculated results. The results of the theoretical predictions for the $\alpha$ decays of europium isotopes considered in the present study are presented in table 2, together with the result of the recent work \[10\]. One can see that our experimental limit for the $\alpha$ decay $^{151}\text{Eu} \rightarrow ^{147}\text{Pm}^*$ is not so far from the theoretical predictions.

The sensitivity of the experiment can be improved by purification of the Eu$_2$O$_3$ sample from potassium, uranium and thorium. Nevertheless, the main source of the background in the experiment is contamination of the sample by radioactive europium isotopes $^{152}\text{Eu}$, $^{154}\text{Eu}$ and $^{155}\text{Eu}$. One could overcome this problem by using europium extracted from minerals with low concentration of uranium and thorium to avoid production of radioactive Eu nuclides. In this case sensitivity of an experiment can be improved by one order of magnitude (also assuming 2–3 times longer measurements and optimization of efficiency). Further improvement of sensitivity could be achieved by using a Li$_6$Eu(BO$_3$)$_3$ crystal \[12\] as a cryogenic scintillating bolometer with typically very high energy resolution for alpha particles (at the level of a few keV), high detection efficiency, and excellent particle discrimination ability which allow to distinguish single $\alpha$ events, $\alpha$ signals with admixture of $\gamma$ quanta, and pure $\gamma(\beta)$ events. This approach was recently successfully applied in work \[18\] to detect $\alpha$ transitions of $^{209}\text{Bi}$ to the ground state and the first excited level of $^{205}\text{Tl}$ with the half-life $(2.01 \pm 0.08) \times 10^{19}$ y.

5 Summary

The alpha decays of naturally occurring europium isotopes, which are accompanied by emission of $\gamma$ quanta, were searched for with the help of the ultra-low-background HPGe detector at the HADES underground laboratory of the Institute for Reference Materials and Measurements (Geel, Belgium). The new improved half-life limit for alpha decay of $^{151}\text{Eu}$ to the first excited level of $^{147}\text{Pm}$ ($J^\pi = 5/2^+$, $E_{\text{exc}} = 91.1$ keV) is set as: $T_{1/2} \geq 3.7 \times 10^{18}$ y. This value is not so far from the theoretical predictions, which are in the range of $7 \times 10^{18} - 1 \times 10^{20}$ y.

The sensitivity of the experiment can be improved by one order of magnitude by using a radiopure europium sample, increase of the exposure and optimization of efficiency. Taking into account the theoretical estimations, such an improvement of sensitivity could lead to detection of the alpha decay of $^{151}\text{Eu}$ to the first excited level of $^{147}\text{Pm}$ with the half-life on the level of $10^{19}$ y. Further improvement of the sensitivity could be achieved by using Li$_6$Eu(BO$_3$)$_3$ crystals as scintillating bolometers, as proposed in \[12\].

As a by-product of the experiment, the new improved $T_{1/2}$ limit for $\alpha$ decay of $^{151}\text{Eu}$ to the ground state of $^{149}\text{Pm}$ was set as $T_{1/2} \geq 5.5 \times 10^{17}$ y. However, due to the very small energy release in this $\alpha$ decay, the limit is still many orders of magnitude far from the theoretical expectations.

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