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Search for α decay of ^{151}Eu to the first excited level of ^{147}Pm using underground γ-ray spectrometry

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Abstract. The alpha decay of ^{151}Eu to the first excited level of ^{147}Pm ($J^\pi = 5/2^+$, $E_{exc} = 91.1$ keV) was searched for at the HADES underground laboratory (≈ 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^3) 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 α 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 α active with an α decay energy $Q_\alpha = 1964.9(1.1)$ keV and $Q_\alpha = 272.5(2.0)$ keV, respectively [2,3]. The first indication on α decay of ^{151}Eu to the ground state of ^{147}Pm with the half-life $T_{1/2} = 5^{+11}_{-3} \times 10^{18}$ 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 α transition (obtained by using different approaches [7–10]) are in the range of 7×10^{18}–1×10^{20} y, which is at the level of present sensitivity accessible in low background experiments. Indeed, recently the α decay of ^{190}Pt to the first excited level of ^{186}Os with the half-life 2.6×10^{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 ^{190}Pt (0.012%). The decay ^{151}Eu → ^{147}Pm* 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 $Li_6Eu(BO_3)_3$ 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 α decay of ^{153}Eu to the ground state of ^{149}Pm by using a γ-ray detector because of instability of the daughter ^{149}Pm nucleus relative to β decay

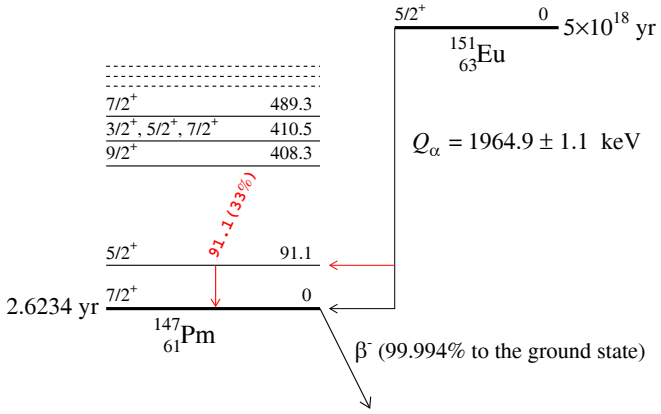

Fig. 1. (Color online) Expected scheme of α decay of ^{151}Eu (levels above 489.3 keV are omitted). The energies of the levels and of the de-excitation γ quantum are given in keV [5,6].

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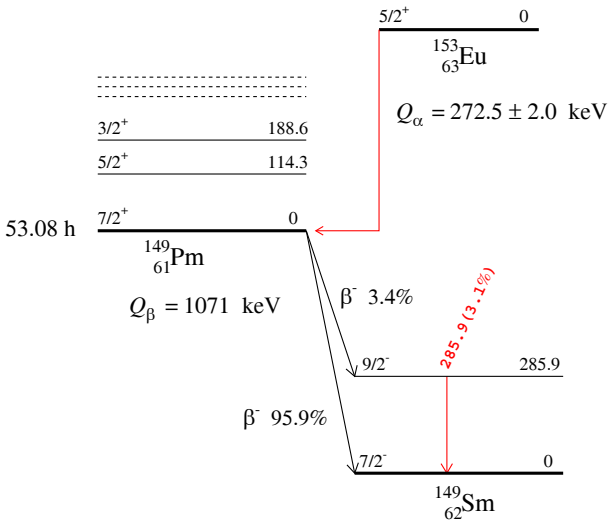

Fig. 2. (Color online) Expected scheme of α decay of ^{153}Eu. The energies of the levels and of the deexcitation γ quantum are given in keV [5,13]. Only the main branches of ^{149}Pm β decay are shown.

with half-life $T_{1/2} = 53.08$ h and $Q_\beta = 1071$ keV (the decay scheme of ^{153}Eu is shown in fig. 2). Up to date only the limit $T_{1/2} \geq 1.1 \times 10^{16}$ y on α decay of ^{153}Eu was set in [12].

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

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 sand and clay overburden of about 500 m of water equivalent assures a muon flux reduction factor of about 5000.

A high-purity europium oxide (Eu_2O_3) sample (in the form of white powder) with mass 303 g, contained in a polyethylene bag, was placed directly on the 0.7 mm thick aluminium endcap of the HPGe detector Ge-2 in HADES. The shape of the sample was almost cylindrical (diameter 6 cm, height 11 cm) defined by the inner copper shield. Detector Ge-2 is an ultra-low background n-type semiplanar HPGe detector (volume 40 cm^3) with a sub-micron top deadlayer. The shielding surrounding the detector consists of 15 cm of lead with an inner 10 cm layer of low radioactive lead (the activity of ^{210}Pb < 0.5 Bq/kg) originating from old French monuments. There is also an inner lining of 6 cm of freshly produced electrolytic copper. The ^{222}Rn activity in air of the laboratory is rather low (average of about 6 Bq/m^3) and is reduced further inside the shield by flushing it with the boil-off nitrogen from the detector's Dewar. Although detector Ge-2 is the smallest detector in HADES, it was chosen because of its excellent energy resolution and because the main interest of this study is the energy region around 91 keV. The detector energy resolution (the full width at the half of maximum, FWHM, keV) can be approximated in the energy region of 60–662 keV by function FWHM $= 0.4699 + 0.02928\sqrt{E_\gamma}$, where E_γ is energy of γ quanta in keV. In particular, FWHM $= 0.749$ keV at 91 keV.

The stability of the measurement system is very high due to the underground location with a cast iron lining and due to the use of a UPS (uninterruptable power supply). Generally, one spectrum per day is collected and acquisition is stopped when liquid nitrogen is filled. The energy stability of the system was possible to check daily due to the two main peaks at 122 keV and 344 keV of ^{152}Eu and with a bin width of 82 eV they showed no detectable drift. For long measurements, the first spectrum after changing a sample is usually excluded in order to assure that radon is removed by the boil-off nitrogen.

The data with the Eu_2O_3 sample were collected over 93.03 days, while the background spectrum of the detector was measured over 68.95 days. The energy spectrum accumulated with the Eu_2O_3 sample in the energy range of 10–670 keV is shown in fig. 3 together with the background data (normalized to the measurement time of the sample).

With the aim of quantifying radionuclides emitting gamma-rays up to 2.7 MeV, an additional measurement for 15.4 days was carried out using detector Ge-5. This is a planar HPGe detector from Canberra of a so-called BEGe type (Broad Energy Germanium). It is a p-type crystal with a sub-micron deadlayer. Its diameter and thickness are 80 mm and 30 mm, respectively, resulting in a relative efficiency of 50% (volume 150 cm^3). The endcap of the detector is fabricated from high-purity aluminium. The Eu_2O_3 sample of a cylindrical shape (diameter 8.1 cm, height 6.2 cm) was placed directly on the endcap of the HPGe detector Ge-5.

3 Results

3.1 Radioactive contamination of Eu_2O_3 sample

The comparison of the Eu_2O_3 and of the background spectra shows that the europium sample is contaminated by daughters of ^{232}Th and ^{238}U, and radioactive europium isotopes ^{152}Eu, ^{154}Eu and ^{155}Eu. 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 ^{176}Lu in the sample. Radioactive ^{152}Eu and ^{154}Eu nuclei were produced by neutron captures in ^{151}Eu and ^{153}Eu, respectively. ^{155}Eu can be produced by the double neutron capture reaction ^{153}Eu(n, γ) $\rightarrow$ ^{154}Eu(n, γ) $\rightarrow$ ^{155}Eu 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.

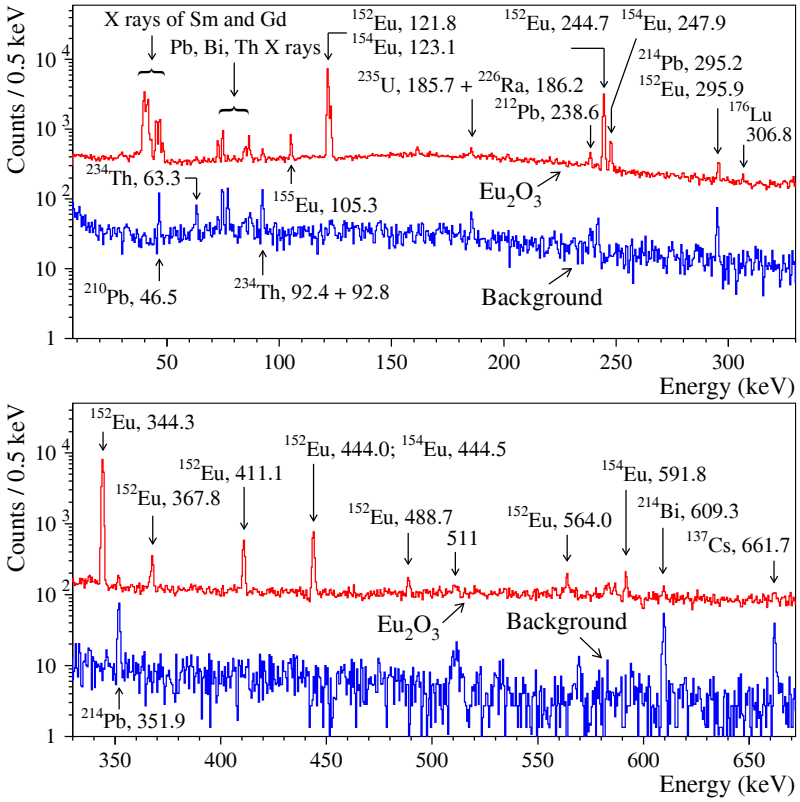

Fig. 3. (Color online) Energy spectrum of the Eu_2O_3 sample measured over 2232.8 h in the 10–330 keV energy interval (upper part), and in the 330–670 keV energy interval (lower part). The background spectrum (measured over 1654.7 h, normalized to 2232.8 h) is also shown. The energies of γ lines are in keV [5].

Massic activities of the contaminant radionuclides (A) were calculated with the formula

$$A = (S_{sample}/t_{sample} - S_{bg}/t_{bg})/(\vartheta \cdot \varepsilon \cdot m),$$

where S_{sample} (S_{bg}) is the area of a peak in the sample (background); t_{sample} (t_{bg}) is the time of the sample (background) measurement; ϑ is the γ-ray emission probability [5], ε 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 relative 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 Eu_2O_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 α decay of ^{151}Eu to the first excited level of ^{147}Pm

Gamma quanta with an energy of 91.1 keV should be emitted after alpha decay of ^{151}Eu to the first excited level of ^{147}Pm. There is no peak at energy 91 keV in the energy spectrum accumulated with the Eu_2O_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

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

where N is the number of ^{151}Eu nuclei (4.96×10^{23}), ε is the detection efficiency, ϑ is the γ 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 γ quanta emitted in the Eu_2O_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 Eu_2O_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 γ peak searched for), a linear function which describes the background, and two Gaussians to take into account the neighbouring peaks with the

Table 1. Radioactive contamination of the Eu_2O_3 sample measured in HADES by the HPGe γ detectors Ge-2 and Ge-5. All uncertainties are presented as the combined standard uncertainty ($k = 1$). The reference dates for the activities are October 2011 for Ge-2 and April 2012 for Ge-5.

Decay chain	Radionuclide	Massic activity (mBq/kg)	
		Ge-2	Ge-5
	^{40}K	–	420 ± 40
	^{60}Co	–	≤ 9
	^{137}Cs	≤ 3	≤ 18
	^{138}La	–	≤ 7
	^{152}Eu	2520 ± 280	3060 ± 330
	^{154}Eu	380 ± 40	440 ± 50
	^{155}Eu	210 ± 40	290 ± 40
	^{176}Lu	4 ± 2	7 ± 2
^{232}Th	^{228}Ra	23 ± 7	≤ 20
	^{228}Th	23 ± 5	23 ± 8
^{235}U	^{235}U	10 ± 3	10 ± 3
	^{231}Pa	≤ 100	≤ 210
	^{227}Ac	≤ 16	≤ 100
^{238}U	^{234m}Pa	290 ± 130	≤ 910
	^{226}Ra	≤ 15	17 ± 4
	^{210}Pb	≤ 1200	–

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 (χ^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 α decay of ^{151}Eu to the first $5/2^+$ 91.1 keV excited level of ^{147}Pm:

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

The data collected with the Eu_2O_3 sample also allow searching for all possible α decays of ^{153}Eu to ^{149}Pm (including the most probable decay to the ground state) due to β instability of the daughter isotope (see fig. 2). The most intense γ line of ^{149}Pm has an energy of 285.9 keV and a yield $\vartheta = 0.031$ [13]. The detection efficiency of the γ 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 γ 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

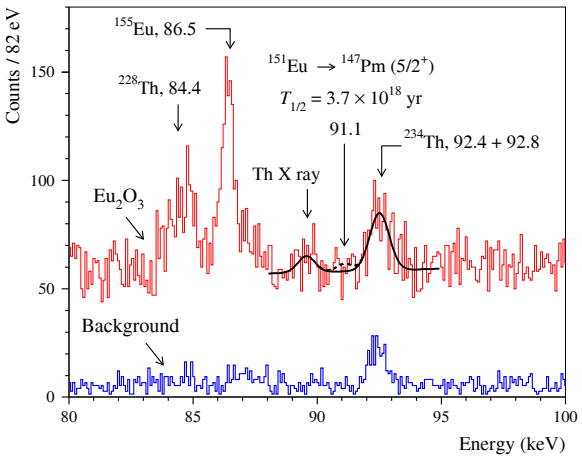

Fig. 4. (Color online) The energy spectrum measured over 2232.8 h with the Eu_2O_3 sample in the 80–100 keV energy interval. Fit of the spectrum in the vicinity of 91.1 keV γ peak is shown by solid line. The expected 91.1 keV γ peak from α transition $^{151}\mathrm{Eu} \rightarrow {}^{147}\mathrm{Pm}^*$ corresponds to the half-life $T_{1/2} = 3.7 \times 10^{18}$ y excluded at 68% CL; it is shown by the dashed line. The background spectrum (measured over 1654.7 h, normalized here to 2232.8 h) is also presented. The energies of γ lines are in keV.

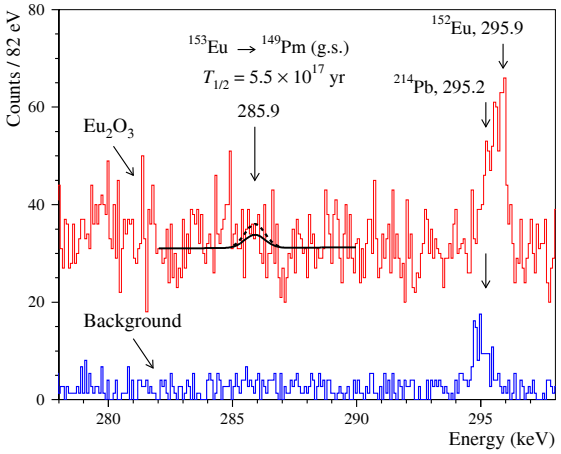

Fig. 5. (Color online) Fragment of the energy spectra accumulated over 2232.8 h with the Eu_2O_3 sample. Fit of the spectrum in the energy interval 282–290 keV is shown by solid line. The expected 285.9 keV γ peak from β decay of ^{149}Pm (which could appear after α decay of ^{153}Eu) is shown by the dashed line. Area of the peak corresponds to the half-life of ^{153}Eu $T_{1/2} = 5.5 \times 10^{17}$ y excluded at 68% CL. The background spectrum (normalized to the time of measurements with the sample) is also presented.

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 α decay of ^{153}Eu to ^{149}Pm at 68% CL:

$$T^{\alpha}_{1/2}(^{153}\mathrm{Eu}) \geq 5.5 \times 10^{17}\,\mathrm{y}.$$

Table 2. Half-life limits on α 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 Q_α [2,3]	Level of daughter nucleus	Experimental $T_{1/2}$		Theoretical estimations
		Previous result	This work	
^{151}Eu $\rightarrow$ ^{147}Pm	$5/2^+$, 91.1 keV	$\geq 2.4 \times 10^{16}$ [12]	$\geq 3.7 \times 10^{18}$	9.7×10^{19} [(a)]
1964.9(1.1) keV		$\geq 6.0 \times 10^{17}$ [4]		7.4×10^{18} [(b)]
				4.5×10^{19} [(c)]
^{153}Eu $\rightarrow$ ^{149}Pm	$7/2^+$, 0 keV (g.s.)	$\geq 1.1 \times 10^{16}$ [12]	$\geq 5.5 \times 10^{17}$	5.2×10^{144} [(a)]
272.5(2.0) keV				2.8×10^{141} [(b)]

(a) Calculated with semiempirical formulae [7].
(b) Calculated with the cluster model of refs. [8,9].
(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 α 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 α decay as a very asymmetric fission process. The approaches were tested with a set of experimental half-lives of a few hundred α 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 α active ^{180}W [17] and ^{151}Eu [4] obtaining adequate agreement between the first experimentally measured and calculated results. The results of the theoretical predictions for the α 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 α decay ^{151}Eu $\rightarrow$ ^{147}Pm* is not so far from the theoretical predictions.

The sensitivity of the experiment can be improved by purification of the Eu_2O_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}Eu, ^{154}Eu and ^{155}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 $Li_6Eu(BO_3)_3$ 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 α events, α signals with admixture of γ quanta, and pure $\gamma(\beta)$ events. This approach was recently successfully applied in work [18] to detect α transitions of ^{209}Bi to the ground state and the first excited level of ^{205}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 γ 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}Eu to the first excited level of ^{147}Pm ($J^\pi = 5/2^+$, $E_{exc} = 91.1$ 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 7×10^{18}–1×10^{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}Eu to the first excited level of ^{147}Pm with the half-life on the level of 10^{19} y. Further improvement of the sensitivity could be achieved by using $Li_6Eu(BO_3)_3$ crystals as scintillating bolometers, as proposed in [12].

As a by-product of the experiment, the new improved $T_{1/2}$ limit for α decay of ^{153}Eu to the ground state of ^{149}Pm was set as $T_{1/2} \geq 5.5 \times 10^{17}$ y. However, due to the very small energy release in this α decay, the limit is still many orders of magnitude far from the theoretical expectations.

The work performed by the HADES staff of Euridice is gratefully acknowledged. FAD and VIT were supported in part through the Space Research Program of the National Academy of Sciences of Ukraine. We are grateful to V.Yu. Denisov for the theoretical half-life for the α decay ^{151}Eu $\rightarrow$ ^{147}Pm* (91.1 keV) absent in [10] but kindly calculated on our request.

References

1. M. Berglund, M.E. Wieser, Pure Appl. Chem. **83**, 397 (2011).
2. G. Audi, A.H. Wapstra, C. Thibault, Nucl. Phys. A **729**, 337 (2003).
3. G. Audi, M. Wang, private communication (2011).
4. P. Belli *et al.*, Nucl. Phys. A **789**, 15 (2007).
5. R.B. Firestone *et al.*, *Table of Isotopes*, 8th edition (John Wiley, New York, 1996) and CD update 1998.
6. N. Nica, Nucl. Data Sheets **110**, 749 (2009).
7. D.N. Poenaru, M. Ivascu, J. Phys. **44**, 791 (1983).
8. B. Buck, A.C. Merchant, S.M. Perez, J. Phys. G **17**, 1223 (1991).
9. B. Buck, A.C. Merchant, S.M. Perez, J. Phys. G **18**, 143 (1992).
10. V.Yu. Denisov, A.A. Khudenko, At. Data Nucl. Data Tables **95**, 815 (2009).
11. P. Belli *et al.*, Phys. Rev. C **83**, 034603 (2011).
12. P. Belli *et al.*, Nucl. Instrum. Methods A **572**, 734 (2007).
13. B. Singh, Nucl. Data Sheets **102**, 1 (2004).
14. E. Andreotti *et al.*, *Status of underground radioactivity measurements in HADES*, in *Proceedings of the 3rd International Conference NPAE 2010, Kyiv, Ukraine, 2010* Kyiv (2011) p. 601.
15. W.R. Nelson *et al.*, SLAC-Report-265, Stanford (1985).
16. G.J. Feldman, R.D. Cousins, Phys. Rev. D **57**, 3873 (1998).
17. F.A. Danevich *et al.*, Phys. Rev. C **67**, 014310 (2003).
18. J.W. Beeman *et al.*, Phys. Rev. Lett. **108**, 062501 (2012).