Available online at www.sciencedirect.com
ScienceDirect
Nuclear Instruments and Methods in Physics Research A 572 (2007) 734–738
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Intrinsic radioactivity of a $Li_6Eu(BO_3)_3$ crystal and α decays of Eu

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Received 9 October 2006; received in revised form 14 December 2006; accepted 20 December 2006
Available online 31 December 2006

Abstract

A small $Li_6Eu(BO_3)_3$ crystal (2.72 g) was measured deep underground at the Gran Sasso National Laboratories with a HP Ge detector of 408 cm^3 during 1500 h. While the crystal is not polluted by usual radioactive contaminants (only limits on the activity of U/Th chains, ^{40}K, ^{60}Co, ^{137}Cs were set on the level of 0.01–1 Bq/kg), the presence of radioactive ^{152}Eu and ^{154}Eu is evident; these are created in capture of neutrons by naturally abundant ^{151}Eu and ^{153}Eu. The measured activities correspond to 0.95 and 0.21 Bq/kg for ^{152}Eu and ^{154}Eu, respectively, with a ratio far from the standard expectation. As by-products, limits on half lives for the α decays $^{151}Eu \rightarrow {}^{147}Pm$ ($E_{exc} =$ 91.1 keV) and $^{153}Eu \rightarrow {}^{149}Pm$ are determined as $T_{1/2} > 2.4 \times 10^{16}$ yr and $T_{1/2} > 1.1 \times 10^{16}$ yr, respectively.

PACS: 23.60.+e; 29.30.Kv; 27.70.+q

Keywords: Alpha decay; ^{151}Eu; ^{152}Eu; ^{153}Eu; ^{154}Eu; Neutron capture; $Li_6Eu(BO_3)_3$ crystal

1. Introduction

While alpha decay is an old phenomenon with a history more than 100 years old, it continues to attract interest—which has even increased in the last years—of both theoreticians [1] and experimentalists. Developments of experimental techniques recently lead to two interesting observations of α decays of very long-lived isotopes. The alpha decay of ^{209}Bi was registered in Ref. [2] with a $Bi_4Ge_3O_{12}$ scintillating bolometer; the half life is $T_{1/2} = (1.9 \pm 0.2) \times 10^{19}$ yr. The alpha decay of the ^{180}W isotope with $T_{1/2} = 1.2^{+0.8}_{-0.4}(stat) \pm 0.3(syst) \times 10^{18}$ yr was observed in an experiment with $^{116}CdWO_4$ crystal scintillators [3]. This result was later confirmed with $CaWO_4$ bolometers [4] and $CaWO_4$ crystal scintillators [5]. ^{209}Bi and ^{180}W nuclides are in competition for the most rarely observed α decay: while the half life of ^{209}Bi is one order of magnitude longer than that of ^{180}W, the specific activity of ^{209}Bi (105 disintegrations per year per gram of Bi) is much more convenient for experimental observation than the one of ^{180}W (only 2.3 disintegrations per year per gram of W of natural composition[1]), which is the lowest natural α activity ever observed.

The ^{151}Eu nucleus is also potentially unstable to α decay; the energy release would be $Q_\alpha = 1.964$ MeV [7]. While ^{151}Eu decay was not experimentally detected to-date, it is another candidate for observation of the α decay with the current techniques. Its half life was calculated as $\sim 3 \times 10^{18}$ yr [8], and measurements with a relatively large $CaF_2(Eu)$ crystal scintillator have been carried out in the

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[1]Natural abundance of ^{209}Bi is 100%, while abundance of ^{180}W is only 0.12% [6].

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doi:10.1016/j.nima.2006.12.025

Laboratori Nazionali del Gran Sasso (LNGS); the results will appear soon [8]. It is worth to note that, although Eu is present in CaF_2(Eu) crystals only as a dopant with mass fraction of $\simeq 0.5\%$, the use of CaF_2(Eu) crystals offers many advantages because of the possibility to obtain detectors of good radiopurity and of large mass and because of the exploitation of the active source approach. Nevertheless, future experiments may be considered using materials with greater content of Eu as $Li_6Eu(BO_3)_3$ crystals; their scintillation properties are quite poor, but they might be used as bolometers.

We report here on an investigation of the intrinsic radioactive purity of a small (2.72 g) $Li_6Eu(BO_3)_3$ crystal which is important for low-background measurements. In addition, it should be also mentioned that $Li_6Eu(BO_3)_3$, as well as borates with Eu as dopant, are used as red phosphor in plasma display panels and cathodoluminescent screens (see e.g. Refs. [9,10]). Thus radioactivity of these materials is important also from the safety point of view.

2. Measurements and data treatment

$Li_6Eu(BO_3)_3$ monocrystals with diameter up to 20 mm and length up to 25 mm were grown by the Czochralsky method in air atmosphere with Platinum crucibles in accordance with Ref. [11]. A small crystal was used in the present measurements: its dimensions were $\oslash 14.5 \times 5.6$ mm, and the mass of the crystal was 2.72 g. Radioactive contamination of the crystal, as an external source with respect to the HP Ge detector of 408 cm^3 volume, was measured in the low-background set-up. The measurements were performed deep underground in the Laboratori Nazionali del Gran Sasso (3800 m w.e.). The $Li_6Eu(BO_3)_3$ sample was measured over 1500 h; the background of the HP Ge detector was measured over 689 h. As an example, part of the experimental spectrum in the energy region up to 450 keV together with the background for comparison is presented in Fig. 1.

There are quite a lot of peaks in the spectrum of the $Li_6Eu(BO_3)_3$ crystal. Analysis showed that all of them belong either to (1) usual contaminants such as chains of ^{232}Th, ^{238}U, and also ^{40}K, ^{60}Co, ^{137}Cs or to (2) radioactive decays of ^{152}Eu and ^{154}Eu.

Comparing the rates of the peaks of "usual" contaminants in the $Li_6Eu(BO_3)_3$ spectrum with those in the background, no evidence was found for an additional pollution of the $Li_6Eu(BO_3)_3$ crystal by these nuclides: rates were equal inside statistical uncertainties. Thus only limits on their activities were calculated at 90% C.L. as summarized in Table 1.

Activities were calculated with the formula

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

where S_{sample} (S_{bg}) is the area of a peak in the sample (background) spectrum; t_{sample} (t_{bg}) is the time of the sample (background) measurement; y is the yield of the corresponding γ line [12]; ε is the efficiency of the full peak detection; m is the mass of the sample. Efficiencies were calculated with the GEANT4 package [13].

Measured rates of ^{152}Eu and ^{154}Eu peaks correspond to activities in the crystal of 949 ± 48 mBq/kg for ^{152}Eu and 212 ± 35 mBq/kg for ^{154}Eu, respectively.

3. ^{152}Eu and ^{154}Eu contamination

Natural Europium consists of only two stable isotopes: ^{151}Eu (47.81%) and ^{153}Eu (52.19%) [6]. Radioactive ^{152}Eu and ^{154}Eu nuclei were produced by neutron capture by ^{151}Eu and ^{153}Eu, respectively. These nuclei have quite long half lives: ^{152}Eu—13.537 yr, and ^{154}Eu—8.593 yr [12]. Cross-sections for capture of thermal neutrons are equal to $\sigma_{151} = 5900 \pm 200$ b (^{151}Eu) and $\sigma_{153} = 312 \pm 7$ b (^{153}Eu) [12]. The $Li_6Eu(BO_3)_3$ crystal with a mass of 2.72 g contains $N_{151} = 7.78 \times 10^{20}$ nuclei of ^{151}Eu and $N_{153} = 8.49 \times 10^{20}$ nuclei of ^{153}Eu.

The dynamics of unstable daughter nuclides in a flux of neutrons is given by two terms; the first describes the creation of daughter nuclei, and the second their decay:

$$\mathrm{d}N_{\mathrm{d}}/\mathrm{d}t = \phi \cdot \sigma_{\mathrm{m}} \cdot N_{\mathrm{m}} - \lambda_{\mathrm{d}} \cdot N_{\mathrm{d}}$$

where N_{d} (N_{m}) is the number of daughter (mother) nuclei; ϕ the flux of neutrons; σ_{m} the cross-section of neutron capture by mother nuclei; and $\lambda_{\mathrm{d}} = \ln 2/T_{1/2\mathrm{d}}$ is the decay constant of the daughter isotope.

A solution of this equation is $N_{\mathrm{d}}(t) = R_{\mathrm{m}} \cdot (1 - \mathrm{e}^{-\lambda_{\mathrm{d}} t})/\lambda_{\mathrm{d}}$, where $R_{\mathrm{m}} = \phi \cdot \sigma_{\mathrm{m}} \cdot N_{\mathrm{m}}$. The corresponding activity of the daughter is

$$A_{\mathrm{d}}(t) = \lambda_{\mathrm{d}} \cdot N_{\mathrm{d}}(t) = R_{\mathrm{m}} \cdot (1 - \mathrm{e}^{-\lambda_{\mathrm{d}} t}).$$

Thus, the maximal activity of the daughter is equal to the rate of its creation, as could be expected.

The ratio of ^{152}Eu and ^{154}Eu activities should be equal to

$$A_{152}/A_{154} = (R_{151}/R_{153}) \cdot X,$$

where $R_{151}/R_{153} = (\sigma_{151} N_{151})/(\sigma_{153} N_{153}) = 17.33$, and $X = (1 - \mathrm{e}^{-\lambda_{152} t})/(1 - \mathrm{e}^{-\lambda_{154} t})$. For $t \to \infty$, $X = 1$, and for $t \to 0$, $X = \lambda_{152}/\lambda_{154} = 0.635$. Thus, for given N_i and σ_i, the value of $A_{152}/A_{154} = (R_{151}/R_{153}) \cdot X$ should be in the range of 11.00–17.33, which is inconsistent with the measured ratio $A_{152}/A_{154} = 4.48 \pm 0.77$.

Two solutions were considered to solve this contradiction:

(1) The isotopic composition of natural Eu in the crystal was, for some reason, heavily distorted. To see the observed ratio of activities, we should have 19.1% of ^{151}Eu and 80.9% of ^{153}Eu, instead of 47.81% for ^{151}Eu and 52.19% for ^{153}Eu in natural isotopic composition [6]. However, mass-spectrometric measurements showed that the Eu in the crystal was of normal composition: 47.9(3)% of ^{151}Eu and 52.1(3)% of ^{153}Eu. The measurements of the Eu isotope ratio have been carried out with the help of the secondary ion-mass spectroscopy method with an IMS-4f system (CAMECA, France).