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Nuclear Instruments and Methods in Physics Research A 541 (2005) 583–589
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
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YAG:Nd crystals as possible detector to search for 2β and α decay of neodymium

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Received 8 November 2004; accepted 1 December 2004
Available online 22 January 2005

Abstract

Energy resolution, α/β ratio, pulse-shape discrimination for γ-rays and α particles and radioactive contamination were studied with yttrium–aluminum garnet doped with neodymium (YAG:Nd). Applicability of YAG:Nd scintillators to search for 2β decay and α activity of natural neodymium isotopes is discussed.

PACS: 29.40.Mc; 23.40.-s; 23.60.+e; 95.35.+d

Keywords: Scintillation detector; Double β decay; α decay; Dark matter; Yttrium–aluminum garnet doped with neodymium

1. Introduction

The great interest to the double beta (2β) decay search [1–5] is related, in particular, with the recent evidence for neutrino oscillations, which strongly suggests that neutrinos have nonzero mass. While oscillation experiments are sensitive to the neutrino mass difference, only measurement of neutrinoless (0ν) 2β decay rate could define the nature of the neutrinos (Majorana or Dirac) and the absolute scale of the effective neutrino mass.

^{150}Nd is one of the most promising candidate for 2β decay study because of its high transition energy ($Q_{2\beta} = 3368$ keV). As a result, the calculated value of the phase space integral $G^{0\nu}_{mm}$ of the 0ν2β decay of ^{150}Nd is the largest among 35 possible $2\beta^-$ decay candidates [6,7]. The theoretical predictions for the product of half-life with the effective neutrino mass $T^{0\nu}_{1/2}\langle m_\nu\rangle^2$ are in the range of 3.4×10^{22}–3.4×10^{24} yr eV^2 [8]. Moreover, on the experimental point of view, the larger is the $Q_{2\beta}$ energy, the simpler is to overcome background problems, especially since the background from natural radioactivity drops sharply above 2615 keV, the energy of γ's from ^{208}Tl decay (^{232}Th family). In addition, the contribution of cosmogenic activation, which is very important for

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doi:10.1016/j.nima.2004.12.014

the next generation of 2β decay experiments [9], decreases at higher energies.

There exists no appropriate detector containing neodymium, and the so-called "active source" experimental method [2] cannot be used. It should be noted that such an experimental approach would ensure high detection efficiency, great advantage when using very expensive enriched ^{150}Nd isotope.

The purpose of our work was to investigate the scintillation properties and carry out initial test of radioactive contamination of neodymium doped yttrium–aluminum garnet (YAG:Nd) as a possible detector for 2β decay experiment with ^{150}Nd. The active source method allows also to investigate other rare processes in Nd nuclei, such as α decay of naturally occurring neodymium isotopes.

2. Measurements and results

2.1. Energy resolution and α/β ratio

Main properties of YAG:Nd (chemical formula $Y_3Al_5O_{12}$: Nd) are presented in Table 1. These crystals are well known and their production is well developed due to their wide application in laser technique. The scintillation properties of cerium doped yttrium–aluminum garnet (YAG:Ce) have been studied in Ref. [10], properties of YAG:Yb have been reported in Ref. [11], whereas YAG:Nd, to our knowledge, was never studied as a scintillator.

All the measurements were carried out for the ⊘17 × 6 mm YAG:Nd crystal with ≈ 2 mol% of Nd. The mass of the crystal is 7.16 g. The photoelectron yield was estimated with the Philips XP2412 bialkali photomultiplier (PMT) as 8% of NaI(Tl). The energy resolution FWHM = 13.6% was measured for 662 keV γ line of ^{137}Cs with the YAG:Nd crystal wrapped by PTFE reflector tape and optically coupled to the PMT XP2412. A substantial improvement of the light output (≈ 22%) and energy resolution was achieved by placing the crystal in liquid (silicone oil with index of refraction ≈ 1.5). The crystal was fixed in the center of the teflon container ⊘70 × 90 mm and viewed by two PMTs XP2412. Fig. 1 demonstrates the energy spectra of ^{137}Cs and ^{207}Bi obtained in such a way with the YAG:Nd crystal. The energy resolution of 9.3% was obtained with the YAG:Nd crystal scintillator for 662 keV γ line of ^{137}Cs.

Table 1
Properties of YAG:Nd crystal scintillators

Density (g/cm^3)	4.56
Melting point	1970 °C
Crystal structure	Cubic Garnet
Hardness (Mohs)	8.5
Refractive index	1.82
Average decay time[a]	4 μs
Photoelectron yield relatively to NaI(Tl)[a]	8%

[a]For γ-rays, at room temperature.

We estimate the energy resolution of the YAG:Nd scintillation detector at the energy $Q_{2\beta}$ of ^{150}Nd using the results of measurements with ^{137}Cs and ^{207}Bi γ sources. The data were fitted by function FWHM (keV) $= a + \sqrt{b \times E_\gamma}$ with values $a = 2$ keV and $b = 5.2$ keV, where energy of γ quanta E_γ is in keV. The energy resolution

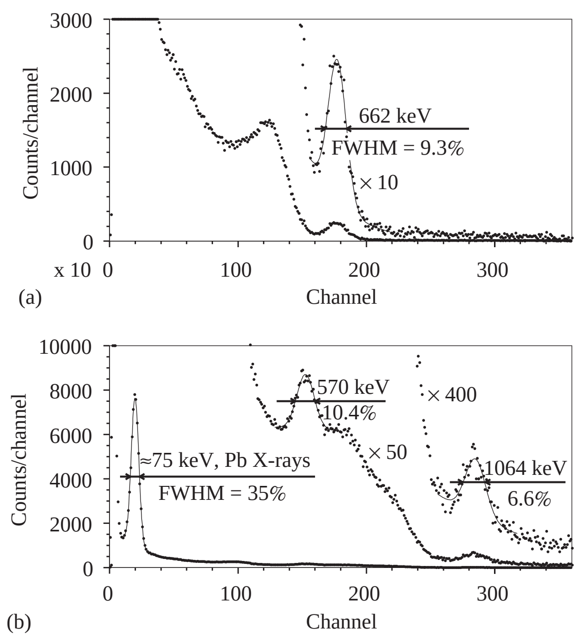

Fig. 1. Energy spectra of ^{137}Cs (a) and ^{207}Bi (b) γ-rays measured with the YAG:Nd scintillation crystal. The crystal was located in liquid and viewed by two distant PMTs (see text).

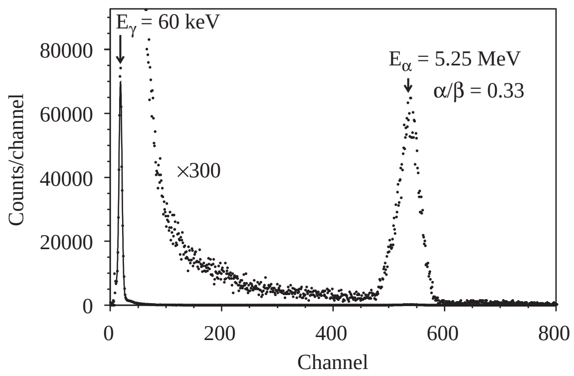

Fig. 2. The energy spectrum of α particles from collimated ^{241}Am source.

FWHM ≈ 4% could be achieved at the $Q_{2\beta}$ energy of ^{150}Nd.

The α/β ratio was measured with the help of the collimated α particles of a ^{241}Am source. As it was checked by surface–barrier detector, the energy of α-particles was reduced to about 5.25 MeV by ≈1 mm of air, due to passing through the collimator. Fig. 2 shows the energy spectrum of the α particles measured by the YAG:Nd scintillator. The α/β ratio is 0.33. The YAG:Nd crystal was irradiated in three perpendicular directions in order to check a possible dependence of the α signal on the direction of irradiation. While such a dependence was found for $CdWO_4$ [12] and $ZnWO_4$ [13] scintillators, it was not observed for YAG:Nd crystal.

2.2. *Pulse-shape discrimination for γ quanta and α particles*

The shapes of scintillation light pulses in YAG:Nd crystals were studied for 5.25 MeV α particles and ≈ 1.5–1.8 MeV γ quanta with the help of a 12 bit 20 MHz transient digitizer as described in Refs. [12,14]. The pulse shape can be fitted by sum of exponential functions:

$$f(t) = \sum A_i/(\tau_i - \tau_0)(e^{-t/\tau_i} - e^{-t/\tau_0}), \quad t > 0$$

where A_i are intensities (in %), and τ_i are decay constants for different light emission components, τ_0 is integration constant of electronics (≈ 0.2 μs). The values of A_i and τ_i obtained by fitting the average of ≈ 3 thousand individual α and γ pulses in the time interval 0–60 μs (see Fig. 3) are presented in Table 2. Differences in light pulse shapes allow to discriminate γ(β) events from α particles. We applied for this purpose the optimal filter method proposed in Ref. [15] and developed in Ref. [14]. To obtain the numerical characteristic of YAG:Nd signal, the so-called shape indicator (SI), the following formula was applied for each pulse

$$SI = \sum f(t_k)P(t_k) \Big/ \sum f(t_k),$$

where the sum is over time channels k, starting from the origin of the pulse up to 50 μs, $f(t_k)$ is the digitized amplitude (at the time t_k) of a given signal. The weight function $P(t)$ is defined as: $P(t) = \{f_\alpha(t) - f_\gamma(t)\}/\{f_\alpha(t) + f_\gamma(t)\}$, where $f_\alpha(t)$

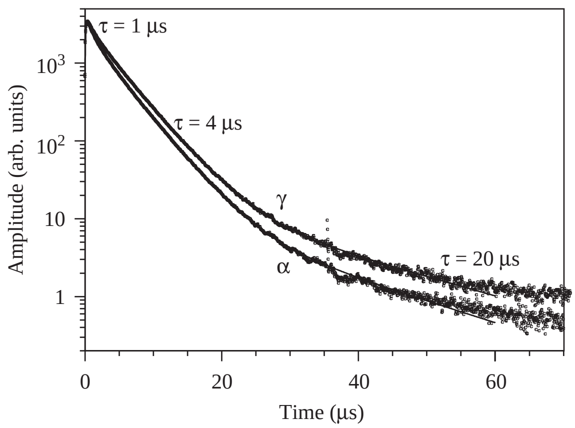

Fig. 3. Decay of scintillation in YAG:Nd for γ-rays and α particles (≈3000 forms of γ and α events were added) and their fit by sum of three exponential components with decay constants τ_i ≈ 1, ≈ 4, and ≈ 20 μs.

Table 2
Decay time of YAG:Nd scintillator for γ quanta and α particles. The decay constants and intensities (in percentage of the total intensity) are denoted as τ_i and A_i, respectively.

Type of irradiation	Decay constants (μs)		
	τ_1 (A_1)	τ_2 (A_2)	τ_3 (A_3)
γ-rays	1.1 (10%)	4.1 (87%)	20 (3%)
α particles	1.0 (15%)	3.9 (83%)	18 (2%)

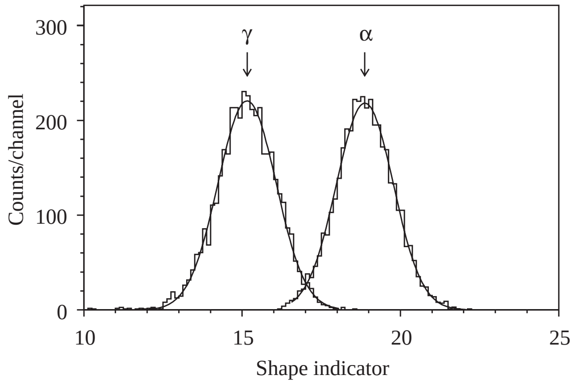

Fig. 4. The shape indicator (see text) distributions measured by YAG:Nd scintillation crystal with α particles ($E_{\alpha} = 5.25$ MeV) and γ quanta (≈ 1.5–1.8 MeV).

and $f_{\gamma}(t)$ are the reference pulse shapes for α particles and γ quanta. Reasonable discrimination between α particles and γ-rays was achieved using this approach as one can see in Fig. 4 where the shape indicator distributions measured by the YAG:Nd scintillation crystal with α particles ($E_{\alpha} \approx 5.3$ MeV) and γ quanta (≈ 1.5–1.8 MeV) are shown.

2.2.1. *Radioactive contamination of the YAG:Nd crystal*

To determine activity of α active nuclides from U/Th contamination in the YAG:Nd crystal, the pulse shape analysis was applied to 14.8 h of low background data carried out in the laboratory based on the earth surface in Kiev. The YAG:Nd crystal was viewed by the low-radioactive PMT (EMI D724KFLB) through plastic scintillator light-guide 10 cm in diameter and 4 cm long. The active light-guide reduces effect of γ-radiation from the PMT and provides suppression of cosmic rays induced background due to the pulse-shape discrimination of scintillation signals. The passive shielding consisted of steel (2 cm) and lead (10 cm). The energy resolution of the detector was 11% for 662 keV γ-rays.

To estimate contamination of the crystal by α active nuclides of U/Th families, the α events were selected from the data accumulated with the YAG:Nd crystal with the help of the pulse-shape discrimination technique. No peculiar feature of the α spectrum could be surely attributed to α emitters from the U/Th chains setting an upper limit on the total α activity of ^{235}U, ^{238}U, and ^{232}Th daughters in the YAG:Nd crystal to be $\leqslant 20$ mBq/kg.

3. Discussion

3.1. 2β *decay of neodymium*

Among neodymium isotopes there are three possible 2β candidates: ^{146}Nd, ^{148}Nd, and ^{150}Nd, listed in Table 3 where the $Q_{2\beta}$ energies and the isotopic abundances are given. As mentioned above, the most interesting of them is ^{150}Nd because of its high transition energy. Two neutrino 2β decay of ^{150}Nd with the half-life $T_{1/2} = 1.9^{+0.7}_{-0.4} \times 10^{19}$ yr was observed in experiment [18] by using a time projection chamber and samples of enriched ^{150}Nd and natural neodymium. The double beta decay of ^{150}Nd was studied with similar technique in Ref. [19] (however, only enriched ^{150}Nd sample was used), and the two-neutrino half-life was measured to be $T_{1/2} = (6.8 \pm 0.8) \times 10^{18}$ yr. Recently, the observation of ^{150}Nd 2ν2β decay to the second excited level of ^{150}Sm (0^+_1 at 741 keV) with $T_{1/2} = 1.4^{+0.5}_{-0.4} \times 10^{20}$ yr was reported in Ref. [20], while only the limit $T_{1/2} > 1.5 \times 10^{20}$ yr was determined previously at 90% C.L. [21]. The most stringent limit on the neutrinoless mode was set in the experiment [19] as $T_{1/2} \geqslant 1.2 \times 10^{21}$ yr.

YAG:Nd scintillators provide a possibility to search for 0ν2β decay of ^{150}Nd by "active source" experimental method. The potential for pulse-shape discrimination is an important advantage of YAG:Nd scintillators as a low counting rate

Table 3
2β unstable neodymium isotopes

Transition	Isotopic abundance (%) [16]	$Q_{2\beta}$ (keV) [17]
^{146}Nd → ^{146}Sm	17.2(0.3)	70.2(2.9)
^{148}Nd → ^{148}Sm	5.7(0.1)	1928.8(1.9)
^{150}Nd → ^{150}Sm	5.6(0.2)	3367.5(2.2)

detector. This technique allowed to reduce significantly the background due to U/Th contamination in scintillators as it was demonstrated in experiments using $CdWO_4$ [12,22] and $CaWO_4$ [9,23] crystal scintillators. Obvious disadvantage of YAG:Nd-based detector is the rather small content of Nd. In the present study the mass concentration is only of $\approx 0.03\%$ of ^{150}Nd. It should be noted, however, that YAG:Nd crystals can be produced with neodymium concentration up to 8 mol%, which corresponds to ≈ 0.1 mass% of ^{150}Nd. In this connection we would like to refer the project CANDLES [24] which intends to use a few tons of nonenriched CaF_2 crystals to search for $0\nu2\beta$ decay of ^{48}Ca. The concentration of target ^{48}Ca nuclei in CaF_2 detector is at the same level: ≈ 0.1 mass% of ^{48}Ca.

The value of energy resolution FWHM = 4% could allow *to discover* [25] the $0\nu2\beta$ decay of ^{150}Nd with the half-life $T_{1/2}^{0\nu2\beta} \sim 10^{25}$ yr, which corresponds to the effective Majorana neutrino mass $m_\nu \sim 0.06$–0.6 eV (taking into account all existing calculations of nuclear matrix elements for $0\nu2\beta$ decay of ^{150}Nd). As for the *sensitivity* [25], an experiment involving ≈ 20 tons of nonenriched YAG:Nd crystals with ≈ 8 mol% of Nd could reach the half-life sensitivity $\approx 3 \times 10^{26}$ yr (supposing zero background during ten years of measurements), which corresponds to a Majorana neutrino mass of 0.01–0.1 eV.

3.2. α decay of neodymium

The α decay is energetically allowed for five natural isotopes of neodymium; energies of α decay (Q_α) and natural abundances are listed in Table 4, as well as calculation of half-lives based on models [26–28], and the half-life of ^{145}Nd calculated in Ref. [29]. Alpha decay was observed only for ^{144}Nd. Average half-life $T_{1/2} = 2.29 \pm 0.16 \times 10^{15}$ yr [30] was derived on the basis of four measurements: $T_{1/2} = 2.2 \times 10^{15}$ yr [31], $T_{1/2} = 1.9 \times 10^{15}$ yr [32], $T_{1/2} = 2.4 \pm 0.3 \times 10^{15}$ yr [33] and $T_{1/2} = 2.65 \pm 0.37 \times 10^{15}$ yr [34]. It is obvious that, due to low Q_α values and long expected $T_{1/2}$'s, there is no perspective to observe the α decay of ^{143}Nd, ^{146}Nd and ^{148}Nd. As for ^{145}Nd, only two $T_{1/2}$ limits are known: $T_{1/2} > 6 \times 10^{16}$ yr [32] and $T_{1/2} > 1 \times 10^{17}$ yr [35].

The indication of α decay of natural ^{180}W isotope with half-life $T_{1/2} = 1.2^{+0.8}_{-0.4}$ (stat) ± 0.3 (syst) $\times 10^{18}$ yr has been recently observed in the experiment [12] with the help of the low background $^{116}CdWO_4$ crystal scintillators. This result was confirmed with $CaWO_4$ crystal scintillators [23] and bolometers [36]. It should be also referred the excellent result on the detection of α decay of ^{209}Bi with the half-life $T_{1/2} = (1.9 \pm 0.2) \times 10^{19}$ yr [37]. Both experiments [36] and [37] use the cryogenic technique allowing to measure heat and light signals simultaneously. This method provides perfect selection of α events on the background caused by γ-rays (electrons). It should be interesting to test YAG:Nd crystal as cryogenic detectors to search for α decay of ^{145}Nd and to measure more accurately the ^{144}Nd half-life.

It should be also mentioned possibility to search for spin-dependent inelastic scattering of weakly interacting massive particles (WIMP) because of nonzero spin ($7/2^-$) of ^{145}Nd with excitation of its

Table 4
Theoretical calculations of half-lives for α decay of natural Nd isotopes

Isotope	Abund. [16]	Q_α (MeV) [17]	Calculated $T_{1/2}$ (yr) [26]	[27]	[28]	[29]	Exp. $T_{1/2}$ (yr) [30]
^{143}Nd	12.2%	0.520	$>1.1 \times 10^{80}$	5.2×10^{93}	8.1×10^{81}	—	—
^{144}Nd	23.8%	1.905	1.9×10^{15}	4.3×10^{15}	3.8×10^{15}	—	$2.29 \pm 0.16 \times 10^{15}$
^{145}Nd	8.3%	1.578	1.7×10^{22}	3.9×10^{23}	5.0×10^{22}	3.7×10^{22}	$>1 \times 10^{17}$
^{146}Nd	17.2%	1.182	2.0×10^{34}	3.9×10^{34}	1.1×10^{35}	–	–
^{148}Nd	5.7%	0.599	6.1×10^{70}	1.1×10^{71}	1.9×10^{72}	—	—

For ^{143}Nd, we take into account that change in a parity and spin additionally suppresses the ^{143}Nd decay rate.

low energy nuclear levels (the lowest one: $3/2^-$ 67 keV, E2 transition, and the second level: $5/2^-$ 72 keV, M1 transition). Simultaneous detection of heat and light (as in Refs. [38,39]) would allow to discriminate such "mixed" events (nuclear recoil plus γ quanta) from "pure" nuclear recoils or γ (electron) events because of different heat/light ratio.

4. Conclusions

The scintillation properties of YAG:Nd crystals were studied. An energy resolution of 9.3% (662 keV ^{137}Cs γ line) was obtained with the YAG:Nd crystal scintillator placed in liquid and viewed by two PMTs. Shapes of scintillation signals were investigated, and reasonable pulse-shape discrimination for γ-rays and α particles was achieved. The α/β ratio was measured with the YAG:Nd scintillator to be equal to 0.33 for 5.25 MeV α particles. Radioactive contamination of the YAG:Nd crystal by α active nuclides from U/Th chains was estimated to be lower than 20 mBq/kg.

Three potential 2β active neodymium isotopes can be studied using YAG:Nd crystal. Due to good scintillation characteristics and pulse-shape discrimination ability, YAG:Nd seems to be encouraging scintillators for the measurement of ^{150}Nd 2β decay.

The YAG:Nd crystals could also be used to search for α decay of natural neodymium isotopes, in particular, ^{145}Nd. The simultaneous measurement of heat and light with cryogenic technique would provide a strong signature of α decay. However, the use of YAG:Nd crystal as a bolometer still has to be tested experimentally.

Acknowledgements

The authors would like to thank Dr. Yu.D. Glinka from the Institute of Surface Chemistry of the National Academy of Sciences of Ukraine for lending the YAG:Nd crystal used in the present study. We also acknowledge Dr. M. Di Marco for her remarks regarding this manuscript.

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