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Scintillation properties of $Gd_3Al_2Ga_3O_{12}:Ce^{3+}$ single crystal scintillators

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ABSTRACT

The scintillation properties of $Gd_3Al_2Ga_3O_{12}:Ce^{3+}$ (GAGG:Ce) single crystals grown by the Czochralski method with 1 at% cerium in the melt were investigated and results were compared with so far published results in the literature. The light yield (LY) and energy resolution were measured using a XP5200B photomultiplier. Despite about twice higher LY for GAGG:Ce, the energy resolution is only slightly better than that of LuAG:Ce due to its worse intrinsic resolution and non-proportionality of LY. The LY dependences on the sample thickness and amplifier shaping time were measured. The estimated photofraction in pulse height spectra of 320 and 662 keV γ-rays and the total mass attenuation coefficient at 662 keV γ-rays were also determined and compared with the theoretical ones calculated using the WinXCom program.

1. Introduction

Single crystal scintillators are widely used for the detection of ionizing radiation in nuclear and high energy physics, modern medical imaging, space exploration, and industry. The fast and efficient 5d→4f luminescence of Ce^{3+} makes it a well-suited emission center in scintillator applications. As a result, new types of Ce-doped inorganic scintillators were intensively studied in the last two decades and some of them were successfully commercialized; for reviews see Refs. [1–4].

Oxide materials based on garnet structure are promising candidates for scintillator applications because of the well-mastered technology developed for laser hosts and easy doping by rare-earth elements. The Ce-doped $Lu_3Al_5O_{12}$ (LuAG:Ce) single crystal was shown to be a prospective scintillator material with a relatively high density of 6.7 g/cm^3 and a fast scintillation response of about 60–80 ns [5]. Due to technological improvements the reported light yield (LY) has gradually increased from about 14,000 up to 25,000 photons/MeV [6,7]. Scintillation performance of LuAG:Ce is strongly degraded by the presence of shallow electron traps which delay an energy delivery to the Ce^{3+} emission centers and give rise to intense slow components in the scintillation decay [8,9]. These traps were ascribed to the antisite Lu_{Al} defects in the LuAG host [10] which are typical defects in the garnet crystals grown from high temperature melt [11,12]. It has been reported that Ga admixture in LuAG host diminishes the energy trapping effects [13] and somewhat increased LY was obtained for Ga concentration up to 20 at% in $Lu_3(Al,Ga)_5O_{12}$:Ce [14]. Recently, $Gd_3Al_2Ga_3O_{12}$:Ce single crystal grown by the micro-pulling down method demonstrated high LY of 42,000 photons/MeV (ph/MeV) and energy resolution of 8.3% at 662 keV as measured with avalanche photodiode (APD) at room temperature [15]. An improvement of the scintillator performance was achieved in $Gd_3Al_2Ga_3O_{12}$:Ce single crystal grown by the Czochralski method which shows high LY of 46,000–50,600 ph/MeV and good energy resolution of 4.9–5.5% at 662 keV [16,17]. However, lower LY of about 33,000 ph/MeV and energy resolution of 6.1% at 662 keV were reported at the samples of the same composition by another group [18]. The composition of multicomponent garnet scintillators influences strongly the energy transfer processes and interaction of Ce^{3+} and Pr^{3+} emission centers with the host. Fundamental aspects of these processes became intensively studied and of abroad interest [19–22] and the research results in the field of garnet scintillators in the last decade have been recently summarized in a review paper [23].

In this paper we have investigated the scintillation properties of the latest generation of $Gd_3Al_2Ga_3O_{12}$:Ce (GAGG:Ce) single crystals grown by the Czochralski method. The measurements of

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pulse height spectra were performed in order to evaluate LY, its non-proportionality and energy resolution. The LY dependences on the sample thickness and amplifier shaping time were measured. The estimated photofraction in pulse height spectra of 320 and 662 keV γ-rays and the total mass attenuation coefficient at 662 keV γ-rays were also determined and compared with the theoretical ones calculated using the WinXCom program.

2. Experimental

GAGG:Ce single crystals were produced in Materials Research Laboratory, Furukawa Co. Ltd. in Japan [16]. The crystals were grown by the Czochralski method with cerium concentration of 1 at% ($Ce_{0.03}Gd_{2.97}Ga_3Al_2O_{12}$) from Ir crucible under Ar with adding 1.5% of O_2 atmosphere. A stoichiometric mixture of high purity 4N CeO_2, Gd_2O_3, β-Ga_2O_3, and α-Al_2O_3 powders was used as starting materials. Polished plates of 1, 2 and 5 mm thickness cut from the parent sample were used for the measurements. The crystal density was 6.69 g/cm^3 determined by the Archimedes method.

Light yield measurements were performed under the excitation of 662 keV γ-rays from a ^{137}Cs source using a photomultiplier (PMT) based-setup described in Ref. [24]: the signal from a Photonis XP5200B PMT anode was sent to a CANBERRA 2005 preamplifier and then to a Tennelec TC243 spectroscopy amplifier. The PC-based multichannel analyzer (Tukan 8k MCA) was used to record the pulse height spectra. The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) of energy deposited in the crystal, was determined by means of a single photoelectron method [24,25]. In this method the number of photoelectrons is measured by comparing the position of a full energy peak of γ-rays detected in the crystals with that of the single photoelectron peak from the photocathode. The measurements of LY non-proportionality and energy resolution were carried out for a series of X/γ-rays emitted by different radioactive sources in the energy range from 32.1 to 1274.5 keV.

The total mass attenuation coefficient at 662 keV γ-rays was determined using the good geometry arrangement of a source (15 mCi ^{137}Cs), absorber ($5 \times 5 \times 1$ mm^3 GAGG:Ce sample) and NaI:Tl detector. A narrow beam of γ-rays is defined by circular apertures (∅4 mm) in the Pb-collimators of the source and detector, placed at a distance of 60 cm. All measurements were carried out at room temperature (RT).

3. Results and discussion

3.1. Light yield and energy resolution

Fig. 1 presents the pulse height spectra of γ-rays from ^{137}Cs (662 keV) source as measured at 4 μs shaping time for GAGG:Ce, LuAG:Ce and BGO crystals with the same size of $5 \times 5 \times 1$ mm^3, whereas the photoelectron yield and energy resolution values are summarized in Table 1. The photoelectron yield (phe/MeV) was recalculated to the LY (ph/MeV) using the average quantum efficiency $\overline{QE}$ of 18.5% for GAGG:Ce and LuAG:Ce and $\overline{QE}$ of 22% for BGO, based on the typical quantum efficiency characteristic of PMT provided by manufacturer and the emission spectrum of the crystals. Despite about twice higher LY for GAGG:Ce, its energy resolution is only slightly better than that of LuAG:Ce. It could be associated with its worse intrinsic resolution and non-proportionality of LY.

We also studied the dependence of LY on the sample thickness (h) for GAGG:Ce. The pulse height spectra of 662 keV γ-rays measured for the studied samples are shown in Fig. 2, whereas the values of LY and energy resolution are summarized in Table 2.

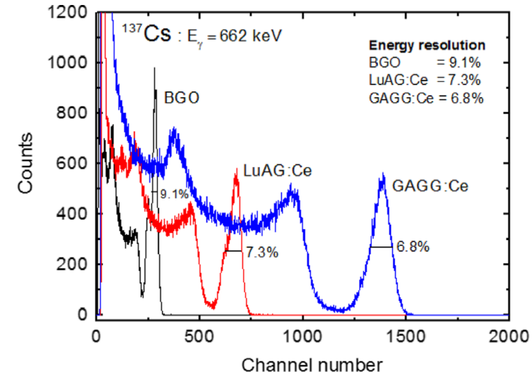

Fig. 1. Pulse height spectra of 662 keV γ-rays (^{137}Cs source) measured with GAGG:Ce, LuAG:Ce and BGO crystals with the same size of $5 \times 5 \times 1$ mm^3.

Table 1
Photoelectron yield, LY and energy resolution for GAGG:Ce, LuAG:Ce and BGO crystals with the same size of $5 \times 5 \times 1$ mm^3 measured at 4 μs shaping time.

Crystal	Photoelectron yield (phe/MeV)	LY (ph/MeV)	$\Delta E/E$ (%)
GAGG:Ce	8860 ± 440	$47{,}900 \pm 4800$	6.8 ± 0.2
LuAG:Ce	4300 ± 220	$23{,}200 \pm 2300$	7.3 ± 0.2
BGO	1860 ± 100	8450 ± 800	9.1 ± 0.3

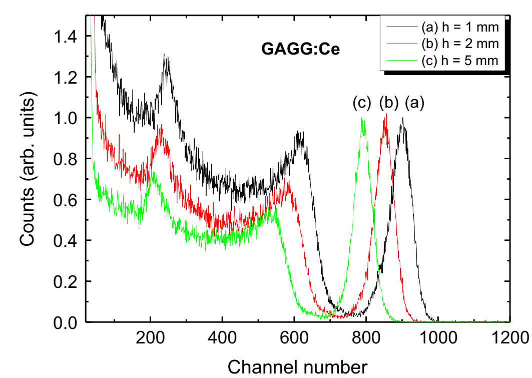

Fig. 2. Pulse height spectra of ^{137}Cs γ-rays measured for the GAGG:Ce samples with thickness (h) of 1, 2 and 5 mm.

The LY value of 47,900 ph/MeV (energy resolution of 6.8%) obtained in this work for a $5 \times 5 \times 1$ mm^3 GAGG:Ce sample is comparable to that of 46,000–50,600 ph/MeV (energy resolution of 4.9–7.3%) recently measured for the same size GAGG:Ce samples [16,17]. We note that the LY value of the studied GAGG:Ce decreases with thickness down to 42,100 ph/MeV (88% LY of 1 mm thick sample) for 5 mm thick sample, which is better than the value (85% LY of 1 mm thick sample) measured for 4.5 mm thick sample in Ref. [17]. It indicates that scintillation light loss due to self-absorption and photon scattering in the studied GAGG:Ce is smaller than that of the samples in Ref. [17]. This feature is of importance for maintaining high LY with increasing sample size. Despite a comparable photoelectron yield, the energy resolution of 6.8% obtained for a $5 \times 5 \times 1$ mm^3 sample is worse than that of 6.1% observed for a $10 \times 10 \times 5$ mm^3 GAGG:Ce sample in Ref. [18], which is due to a higher contribution of the intrinsic resolution (6.0% versus 5.2%) for the studied sample.

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as [26]

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2 \tag{1}$$