Radiation Measurements 87 (2016) 24–28

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Timing characteristics of the scintillation response of $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce single crystal scintillators

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HIGHLIGHTS

- Scintillation time response of GAGG:Ce crystals was studied.
- $Gd_3Al_2Ga_3O_{12}$:Ce exhibited superior timing characteristics.
- Energy resolution of 5.6% @662 keV was obtained for $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce.
- Coincidence time resolution of 345 ps was obtained for $Gd_3Al_2Ga_3O_{12}$:Ce.

ARTICLE INFO

Article history:
Received 27 November 2015
Received in revised form
1 February 2016
Accepted 6 February 2016
Available online 10 February 2016

Keywords:
Coincidence time resolution
Energy resolution
$Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce
$Gd_3Al_2Ga_3O_{12}$:Ce
Scintillation decay

ABSTRACT

The timing characteristics of scintillation response of Czochralski-grown $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce single crystals were compared. The photoelectron yield, scintillation decay times, and coincidence time resolution were measured. At 662 keV γ-rays, the photoelectron yield of 6200 phe MeV^{-1} obtained for $Gd_3Al_2Ga_3O_{12}$:Ce is higher than that of 4970 phe MeV^{-1} obtained for $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce, while an inferior energy resolution of the former (7.2% vs. 5.6%) is observed. Scintillation decays are approximated by sum of exponentials with the dominant fast component decay time and its relative intensity of 89 ns (73%) for $Gd_3Al_2Ga_3O_{12}$:Ce and 136 ns (69%) for $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce. The coincidence time resolution obtained for $Gd_3Al_2Ga_3O_{12}$:Ce is superior than that of $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce. The normalized time resolution was also discussed in terms of a number of photoelectrons and decay characteristics of the light pulse.

1. Introduction

In the last two decades, new scintillator materials characterized by high light output, fast response time and high detection efficiency were intensively investigated due to an increasing demand in modern medical imaging, high-energy physics research and advanced instruments. Ce^{3+}-doped $Lu_3Al_5O_{12}$ (LuAG:Ce) single crystal is a prospective scintillator with a relatively high density of 6.7 g/cm^3 and a short decay time of about 60–80 ns (Nikl et al., 2000). However, its scintillation performance is degraded by shallow electron traps which delay an energy delivery to Ce^{3+} emission centers and scintillation decay contains high content of slow component (Nikl et al., 2007; Chewpraditkul et al., 2009). By Ga^{3+} admixture in aluminum garnet host, the shallow electron traps are buried in a bottom of conduction band (Fasoli et al., 2011) which result in a reduction of the trapping effects (Nikl et al., 2006), and somewhat increase of light yield (LY) was obtained for Ga content up to 20 at % in $Lu_3(Al,Ga)_5O_{12}$:Ce (Ogino et al., 2009). Undesired thermal ionization of $5d_1$ excited state of Ce^{3+} center can be reduced by a Gd^{3+} admixture. Balanced admixture of Gd and Ga into the $Y_3Al_5O_{12}$ host leads to dramatic LY increase in $(Gd,Y)_3(Al,Ga)_5O_{12}$:Ce ceramics (Cherepy et al., 2010). The recent studies thus focus on the Ce-doped $(Gd,RE)_3(Ga,Al)_5O_{12}$, RE = Lu,Y multicomponent garnets (Cherepy et al., 2010; Kamada et al., 2011a, 2011b), the influence of Gd and Ga admixture on the band gap value and band edge positioning (Dorenbos, 2013) and Ce^{3+}

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http://dx.doi.org/10.1016/j.radmeas.2016.02.018
1350-4487/

emission quenching (Ogiegło et al., 2013). Scintillation performance of single crystal and ceramics of Ce-doped multicomponent garnets has been also directly compared (Wu et al., 2015). The development of all groups of garnet scintillators within last decade has been recently reviewed (Nikl et al., 2013). The best performing $Gd_3(Ga,Al)_5O_{12}$:Ce samples reported so far showed LY above 58,000 photons per MeV (ph MeV^{-1}). The scintillation decay of $Gd_3Al_2Ga_3O_{12}$:Ce can be accelerated by Ca codoping with an expense of LY value (Tyagi et al., 2013; Wu et al., 2014; Meng et al., 2015), somewhat better result was reported for B cooping (Tyagi et al., 2015) and especially for Mg codoping (Kamada et al., 2015). It has been found that the ratio of Al and Ga in this compound influences the scintillation performance (Kamada et al., 2014), and the highest LY value was obtained with Al_2Ga_3 combination whereas the best energy resolution was obtained with $Al_{2.6}$ $Ga_{2.4}$ combination (Kamada et al., 2014; Sibczynski et al., 2015).

In this paper we compare the timing characteristics of the scintillation response of Czochralski-grown $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce crystals at room temperature. It comprises the photoelectron yield, scintillation decay and coincidence timing measurements. The coincidence time resolution was also discussed in terms of a number of photoelectrons and decay characteristics of the light pulse.

2. Experimental

Polished samples of $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce single crystals with the same size of 5 × 5 × 5 mm^3 were supplied by Materials Research Laboratory, Furukawa Co. Ltd. in Japan. According to the manufacturer, the $Gd_3Al_2Ga_3O_{12}$:Ce crystal was grown by the Czochralski method with cerium concentration of 1 at % from Ir crucible under Ar +1.5% O_2 atmosphere (Kamada et al., 2012), while the $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce crystal was grown by the same method but under Ar+30% CO_2 atmosphere (Kamada et al., 2014).

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe MeV^{-1}) of γ-ray energy deposited in a crystal, was determined by means of the single-photoelectron method (Bertolaccini et al., 1968; Moszynski et al., 1997). In this method the number of photoelectrons is measured by relating the position of a full-energy peak of γ-rays detected in the crystals with that of a single-photoelectron peak from a photocathode. Under γ-ray excitation, the signal from a Photonis XP20D0 PMT coupled with a crystal was sent to a CANBERRA 2005 preamplifier and then to a Tennelec TC243 spectroscopy amplifier set at 2 μs shaping time constant. The Tukan 8 k MCA was used to record the pulse height spectra.

The scintillation decays were measured by the time-correlated single photon counting technique (Bollinger and Thomas, 1961), using a fast-slow coincidence setup. The crystal coupled to a start PMT (Photonis XP2020Q) was excited with 662 keV γ-rays from a ^{137}Cs source and scintillation photons were detected with a fast stop PMT (Hamamatsu R5320, time jitter of 140 ps) placed at a distance of about 15 cm in front of a start PMT. Anode signals from two PMTs were sent to two ORTEC 935 constant fraction discriminators (CFD) used as the time pick-off units for an ORTEC 566 time-to-amplitude converter (TAC). Dynode signals from two PMTs were sent to two ORTEC 113 preamplifiers and amplified with 0.5 μs shaping time constant in two Tennelec TC 244 amplifiers. Two ORTEC 551 timing single-channel analyzers (SCA) were used to select the events, corresponding to a full-energy peak of 662 keV γ-rays and a single-photoelectron peak detected in a start XP2020Q PMT and a stop R5320 PMT, respectively. Decay time spectra were recorded by the Tukan 8 k MCA. The decay times were determined by performing the multi-exponential fits of the decay curves.

Coincidence timing measurements were performed using 511 keV annihilation quanta from a ^{22}Na source. The studied crystal was coupled to a Photonis XP20D0 PMT. A fast BaF_2 crystal coupled to a Photonis XP20Y0Q/DA PMT was used as the reference detector with its time resolution of 128 ps for 511 keV full-energy peak selected in the side channel (Moszynski et al., 2004). Under excitation with 511 keV annihilation quanta from a ^{22}Na source, placed between these two crystals, a signal from each PMT was processed with an ORTEC 935 CFD. Time spectra were measured with an ORTEC 566 TAC and recorded by the Tukan 8 k MCA. In all measurements, a fast - slow coincidence setup was used for a precise selection of the required (511 keV) energy windows.

3. Results and discussion

3.1. Photoelectron yield and energy resolution

Fig. 1 presents the pulse height spectra of 662 keV γ-rays from a ^{137}Cs source measured at 2 μs shaping time constant for $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce crystals, whereas the photoelectron yield and energy resolution (ΔE/E) are collected in Table 1. The photoelectron yield obtained for $Gd_3Al_2Ga_3O_{12}$:Ce is higher than that of $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce. Despite a lower photoelectron yield, an energy resolution of $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce is better than

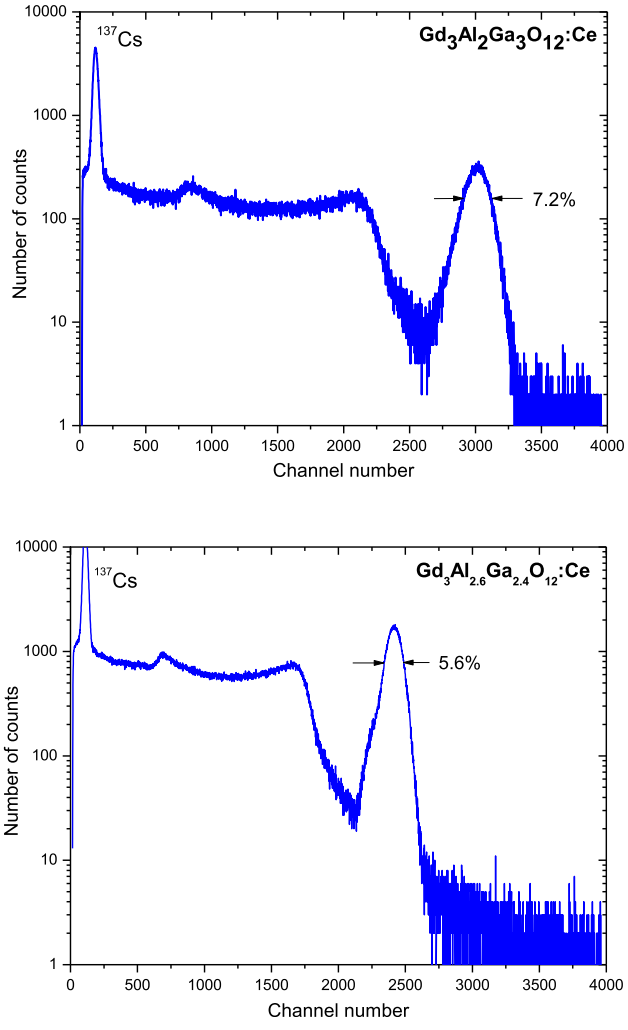

Fig. 1. Pulse height spectra of 662 keV γ - rays from a ^{137}Cs source measured with $Gd_3Al_2Ga_3O_{12}$:Ce and $Gd_3Al_{2.6}Ga_{2.4}O_{12}$:Ce crystals.