Radiation Measurements 90 (2016) 153–156

Contents lists available at ScienceDirect

Radiation Measurements

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

Luminescence and scintillation response of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce scintillators

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HIGHLIGHTS

- Luminescence of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce were studied.
- High LY value above 35,000 ph/MeV was obtained for both crystals.
- Dependence of light yield on integrating time was examined.
- The LY ratio under α- and γ-excitation (α/γ ratio) was determined.
- Photofraction and attenuation coefficient at 662 keV were evaluated.

ARTICLE INFO

Article history:
Received 20 October 2015
Received in revised form
15 December 2015
Accepted 28 December 2015
Available online 30 December 2015

Keywords:
$YGd_2Al_2Ga_3O_{12}$:Ce
$LuGd_2Al_2Ga_3O_{12}$:Ce
Light yield
Luminescence
Scintillation

ABSTRACT

The luminescence and scintillation properties of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce single crystal scintillators were compared. The light yield (LY) and energy resolution were measured using R6231 photomultiplier. At 662 keV γ-rays, high LY value of 37,900 ph/MeV and energy resolution of 7.0% obtained for $YGd_2Al_2Ga_3O_{12}$:Ce are slightly better than the values of 35,400 ph/MeV and 7.6% obtained for $LuGd_2Al_2Ga_3O_{12}$:Ce. The ratio of LY values under excitation with α- and γ-rays (α/γ ratio) was also determined. The LY dependence on amplifier shaping time was measured in order to investigate the timing characteristics in the scintillation response. The estimated photofraction in pulse height spectrum of 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.

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1. Introduction

$Y_3Al_5O_{12}$:Ce and $Lu_3Al_5O_{12}$:Ce single crystals are good scintillator materials used for detection of X-/γ-rays and high energy charged particles due to a fast scintillation response and good light yield (LY) (Moszynski et al., 1994; Nikl et al., 2000). Recently, high LY values up to 25,000 photons/MeV for LuAG:Ce single crystals were reported (Dujardin et al., 2010; Sakthong et al., 2014) although the theoretical value of about 60,000 photons/MeV was estimated for the aluminum garnet scintillators (Dorenbos, 2010). It is due to the presence of antisite-defects related electron traps which delay the energy delivery to Ce^{3+} centers and the scintillation pulse contains intense slow components (Nikl et al., 2007; Chewpraditkul et al., 2009). While the admixture of Ga into the aluminum garnet strongly reduces trapping effects (Nikl et al., 2006; Fasoli et al., 2011), the thermal ionization of $5d_1$ excited state of Ce^{3+} center at room temperature can be made negligible due to the Gd admixture (Kamada et al., 2011a; Nikl et al., 2013a). The recent studies thus focus on a material concept based on multicomponent garnet $(Gd,Y,Lu)_3(Ga,Al)_5O_{12}$ single crystal (Kamada et al., 2011a, 2011b; Ogiegło et al., 2013). An improvement of the scintillator performance was achieved in $Gd_3Al_2Ga_3O_{12}$:Ce single crystal grown by the Czochralski method which show high LY of 46,000–50,600 ph/MeV (Prusa et al., 2013; Kamada et al., 2012a).

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http://dx.doi.org/10.1016/j.radmeas.2015.12.036
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The development of multicomponent garnet scintillators within last decade has been reviewed in (Nikl et al., 2013b).

In this paper, we investigated the luminescence and scintillation properties of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce single crystals grown by the Czochralski method. The absorption and photoluminescence spectra were measured. The LY and its ratio under excitation with α- and γ-rays (α/γ ratio) were measured. The dependence of LY on amplifier shaping time was measured in order to investigate the timing characteristics of scintillation response. The photofraction in pulse height spectrum and total mass attenuation coefficient at 662 keV γ-rays were also determined. All measurements were done at room temperature (RT).

2. Experimental

$YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce crystals with cerium concentration of 1 at % were grown by the Czochralski method from an iridium crucible under Ar + 1.5% of O_2 atmosphere, for details see (Kamada et al., 2012b). Polished plates of 5 × 5 × 1 mm^3 cut from the parent crystals were used for all the measurements. The density of 6.34 and 6.81 g/cm^3, respectively, for $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce crystals, was determined by Archimedes method.

Photoluminescence (PL) emission spectra were measured using a Hitachi F-2500 fluorescence spectrophotometer. Absorption spectra were recorded using a Perkin Elmer Lambda 35, UV–Vis spectrophotometer. LY measurements were done using a R6231 photomultiplier (PMT) under excitation with α- and γ-rays. The photoelectron yield was determined by relating the full-energy peak position with that of the single photoelectron peak from the PMT photocathode (Moszynski et al., 1997).

To determine the total mass attenuation coefficient at 662 keV γ-rays for the studied crystals, a good geometry arrangement of a 15 mCi ^{137}Cs source, crystal sample and NaI:Tl detector was employed. A narrow beam of γ-rays is defined by circular apertures (⌀3 mm) in the Pb-collimators of source and detector, placed at a distance of 40 cm.

3. Results and discussion

3.1. Photoluminescence characteristics

In Fig. 1 the absorption and emission spectra of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce crystals are presented. Two dominant bands observed in the absorption spectra are related to the well-known 4f→5d_1 (between 380 and 500 nm) and 4f→5d_2 (between 325 and 375 nm) transitions of the Ce^{3+} ions, while absorption line at 274 nm is due to $^8S_{7/2} \rightarrow {}^6I_J$ transitions of Gd^{3+} ions. The PL emission maxima in $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce are located at 510 and 512 nm, respectively. Double peak shape of the PL spectra is due to the 5$d_1 \rightarrow {}^2F_{5/2}$ and 5$d_1 \rightarrow {}^2F_{7/2}$ transitions and the peak positions of doublet were determined at 501 and 535 nm for $YGd_2Al_2Ga_3O_{12}$:Ce and at 505 and 538 nm for $LuGd_2Al_2Ga_3O_{12}$:Ce using Gaussian decomposition of the PL spectra plotted on an energy scale which corrected the intensity value on the vertical axis to photon flux per constant energy interval. The main optical properties deduced from these experiments are collected in Table 1. Note somewhat larger energy difference (splitting) between 5d_1 and 5d_2 levels for $YGd_2Al_2Ga_3O_{12}$:Ce with respect to $LuGd_2Al_2Ga_3O_{12}$:Ce. It can be attributed to higher crystal field strength around Ce^{3+} at the dodecahedral site when a Lu^{3+} is replaced by a larger Y^{3+} ion (Wu et al., 2007; Kottaisamy et al., 2008). A Stokes shift in the studied samples estimated from the 4f→5d_1 absorption and 5$d_1 \rightarrow {}^2F_{5/2}$ emission peaks is also collected in Table 1.

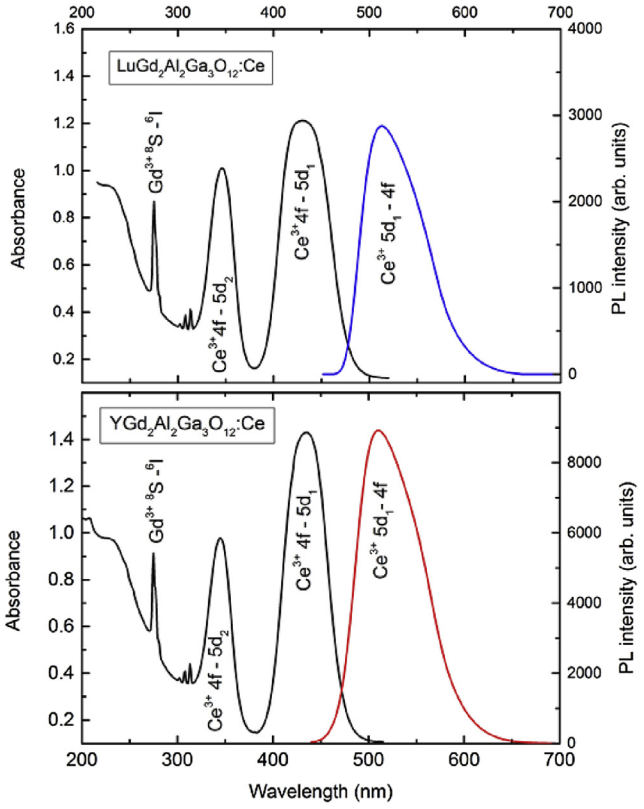

Fig. 1. Absorption and emission spectra of $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce crystals.

Table 1
Optical transitions of Ce^{3+} in $YGd_2Al_2Ga_3O_{12}$:Ce and $LuGd_2Al_2Ga_3O_{12}$:Ce.

	$LuGd_2Al_2Ga_3O_{12}$:Ce	$YGd_2Al_2Ga_3O_{12}$:Ce
Absorption	433/346 nm	434/345 nm
Splitting	0.73 eV	0.75 eV
Emission	505/538 nm	501/535 nm
Stokes shift	0.40 eV	0.38 eV

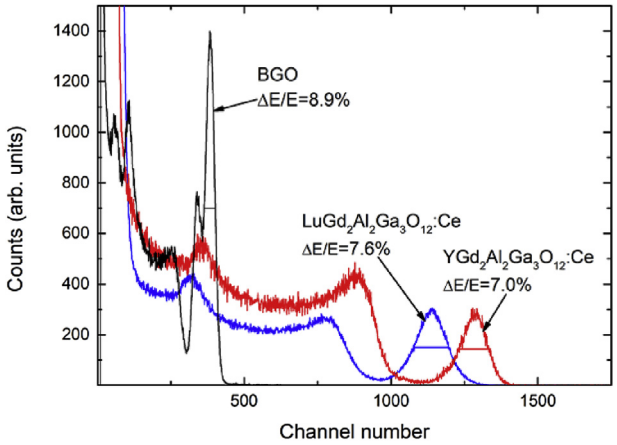

Fig. 2. Pulse height spectra of 662 keV γ-rays measured with $YGd_2Al_2Ga_3O_{12}$:Ce, $LuGd_2Al_2Ga_3O_{12}$:Ce and BGO crystals.

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