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Optical and scintillation characteristics of $Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce single crystals

Warut Chewpraditkul[a,*], Ongsa Sakthong[a], Nakarin Pattanaboonmee[a], Weerapong Chewpraditkul[a], Tomasz Szczesniak[b], Lukasz Swiderski[b], Marek Moszynski[b], Kei Kamada[c,d], Akira Yoshikawa[c,d], Martin Nikl[e]

[a] Department of Physics, King Mongkut's University of Technology Thonburi, Bangkok 10140, Thailand
[b] National Centre for Nuclear Research, A. Soltana 7, Swierk-Otwock PL 05-400, Poland
[c] Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
[d] New Industry Creation Hatchery Center (NICHe), Tohoku University, Sendai 980-8579, Japan
[e] Institute of Physics, ASCR, Cukrovarnicka 10, 16253 Prague, Czech Republic

A R T I C L E I N F O

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A B S T R A C T

The optical and scintillation characteristics of $Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce single crystals are investigated. At 662 keV γ-rays, light yield (LY) of 37,900 ph/MeV and energy resolution of 7.0% obtained for $Gd_2YAl_2Ga_3O_{12}$:Ce are superior to those of 18,900 ph/MeV and 11.5% obtained for $Lu_2YAl_2Ga_3O_{12}$:Ce. Scintillation decays are measured using the time-correlated single photon counting technique. A fast component decay time of 45 ns with relative intensity of 88% obtained for $Lu_2YAl_2Ga_3O_{12}$:Ce is superior to that of 50 ns (65%) for $Gd_2YAl_2Ga_3O_{12}$:Ce. The linear attenuation coefficient at 662 keV γ-rays is also determined and discussed.

1. Introduction

Cerium-doped $Y_3Al_5O_{12}$ (YAG:Ce) and $Lu_3Al_5O_{12}$ (LuAG:Ce) garnet crystals are widely used for detection of high-energy photons and particles due to their good scintillation properties: high light yield (LY) and short decay time [1,2]. Although theoretical LY value of about 60,000 ph/MeV was estimated for the aluminum garnet scintillators [3] the measured LY values for LuAG:Ce single crystals are only about 25,000 ph/MeV [4]. It is due to the presence of shallow electron traps which delay the energy delivery to Ce^{3+} centers and the scintillation pulse contains intense slow components [5,6]. Balanced admixture of Gd and Ga into the aluminum garnet strongly suppresses trapping effects and prevents unwanted thermal ionization of $5d_1$ level of Ce^{3+} emission center at the same time [7,8]. The recent studies thus focus on the Ce-doped multicomponent garnets [9,10]. The development of multicomponent garnet scintillators within last decade has been reviewed [11].

In this paper, we investigate the optical and scintillation properties of $Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce single crystals grown by the Czochralski method. The photoluminescence (PLE) excitation and emission (PL) spectra are measured. LY and energy resolution measurements are carried out using photomultiplier (PMT) readout. Scintillation decay measurements are carried out using the time-correlated single photon counting technique. The total mass attenuation coefficient at 662 keV γ-rays is also determined and compared to the calculated value. All measurements are performed at room temperature (RT).

2. Materials and experimental methods

$Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce single crystals with cerium concentration of 1 at% were grown by the Czochralski method, for details see [12]. Polished plates of 5x5×1 mm^3 cut from the parent crystals were used for all the measurements. The density of 6.34 and 6.37 g/cm^3 for $Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce, respectively, was determined by the Archimedes method.

The PLE and PL spectra were measured using a Hitachi F-2500 fluorescence spectrophotometer. The photoelectron yield was determined by means of a single photoelectron method [13]. Each studied crystal was mounted to the window of a Hamamatsu R6231 PMT using silicone grease and covered with several layers of Teflon tape. The signal from a R6231 PMT was sent to a CANBERRA 2005 preamplifier

* Corresponding author.
E-mail address: raychew.rc@gmail.com (W. Chewpraditkul).

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and then to a Tennelec TC243 spectroscopy amplifier set at 4 μs shaping time constant. The Tukan 8k MCA was used to record the pulse height spectra.

Scintillation decays were measured by the time-correlated single photon counting technique [14] 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) 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). Output signals from an ORTEC 556 TAC were sent to a Tukan 8k MCA to display the scintillation decay spectra.

A good geometry arrangement of source (15 mCi ^{137}Cs), crystal sample and NaI:Tl detector was employed to determine the total attenuation coefficient for the studied crystals at 662 keV γ-rays. A narrow beam of γ-rays with diameter of 3 mm is achieved by 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 PLE and PL spectra of $Gd_2YAl_2Ga_3O_{12}$:Ce and $Lu_2YAl_2Ga_3O_{12}$:Ce crystals are presented. Two dominant bands observed in the PLE spectra are related to the well-known 4 f→5d$_1$ (between 380 and 480 nm) and 4 f→5d$_2$ (between 325 and 375 nm) transitions of the Ce^{3+} ions. An absorption line at 274 nm due to $^8S_{7/2}\rightarrow{}^6I_J$ transition of Gd^{3+} ions is clearly seen in the PLE spectrum of $Gd_2YAl_2Ga_3O_{12}$:Ce. The presence of this line indicates the occurrence of the $Gd^{3+}\rightarrow Ce^{3+}$ energy transfer in $Gd_2YAl_2Ga_3O_{12}$:Ce crystal. Double peak shape of the PL spectra is due to the $5d_1\rightarrow{}^2F_{5/2}$ and $5d_1\rightarrow{}^2F_{7/2}$ transitions and the peak positions were determined at 501 and 535 nm for $Gd_2YAl_2Ga_3O_{12}$:Ce and at 483 and 519 nm for $Lu_2YAl_2Ga_3O_{12}$:Ce using Gaussian decomposition of the PL spectra. 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 $Gd_2YAl_2Ga_3O_{12}$:Ce with respect to $Lu_2YAl_2Ga_3O_{12}$:Ce. This result indicates that the crystal field strength around Ce^{3+} at the dodecahedral site of $Gd_2YAl_2Ga_3O_{12}$:Ce is larger than that of

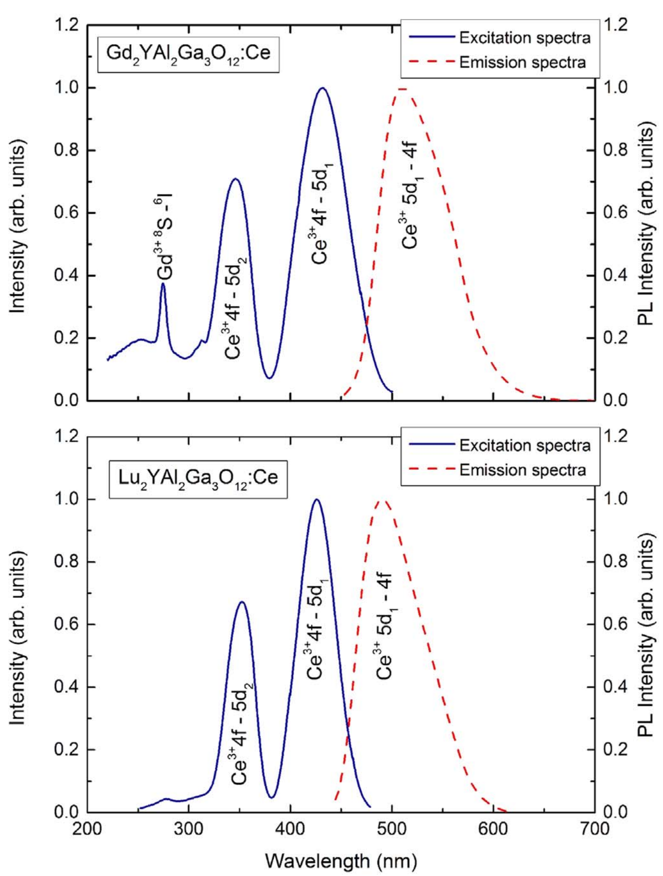

Fig. 1. Excitation and emission spectra of the studied crystals.

Table 1
Optical transitions of Ce^{3+} in the studied crystals.

	$Lu_2YAl_2Ga_3O_{12}$:Ce	$Gd_2YAl_2Ga_3O_{12}$:Ce
Absorption	427/349 nm	434/345 nm
Splitting	0.65 eV	0.75 eV
Emission	483/519 nm	501/535 nm
Stokes shift	0.34 eV	0.39 eV

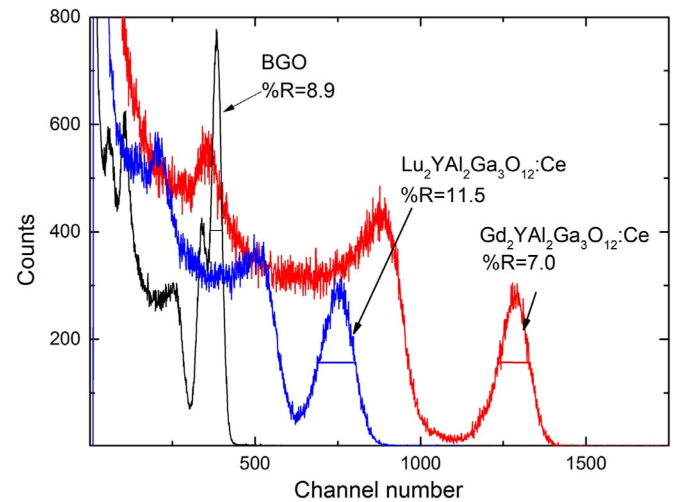

Fig. 2. Pulse height spectra of ^{137}Cs γ-rays measured with the studied crystals.

Table 2
LY and ΔE/E@662 keV γ-rays for the studied crystals.

Crystal	LY (ph/MeV)	ΔE/E (%)
$Gd_2YAl_2Ga_3O_{12}$:Ce	37,900	7.0
$Lu_2YAl_2Ga_3O_{12}$:Ce	18,900	11.5

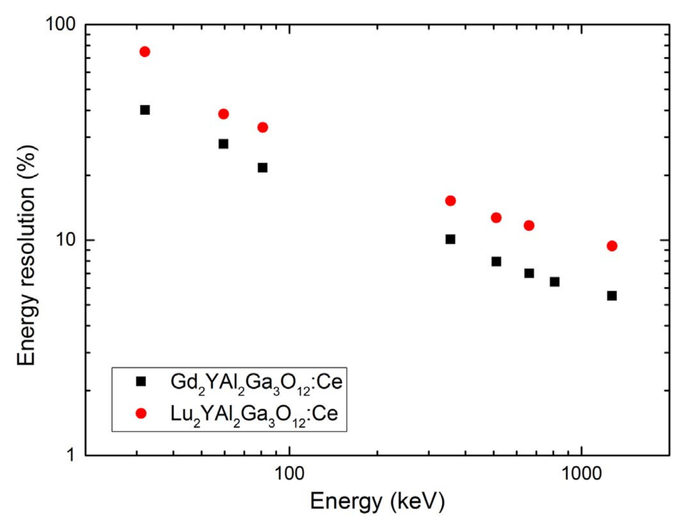

Fig. 3. Energy resolution versus energy of γ-rays for the studied crystals.

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