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Scintillation properties of Er-doped $Y_3Al_5O_{12}$ single crystals

Akihiro Yamaji [a,*], Hiraku Ogino [b], Yutaka Fujimoto [a], Akira Suzuki [a], Takayuki Yanagida [c], Yuui Yokota [a], Shunsuke Kurosawa [b,c], Akira Yoshikawa [a,c]

[a] Institute of Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan
[b] University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
[c] New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aza Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan

HIGHLIGHTS

- Er doped $Y_3Al_5O_{12}$ single crystal scintillators were grown with different Er concentrations.
- Optical properties associated with 4f-4f transition were evaluated.
- Radio luminescence spectra measurements were performed under 5.5 MeV alpha-ray irradiation.
- The highest light yield was estimated to be 63% of that of $Bi_4Ge_3O_{12}$ under 5.5 MeV alpha-ray irradiation.

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ABSTRACT

Er-doped $Y_3Al_5O_{12}$ single crystals with different Er concentrations of 0.1, 1.0, 10, 30, and 50% were grown by the micro-pulling down method. There were several absorption lines due to the Er^{3+} 4f-4f transitions in the transmittance spectra and these lines correspond to the transitions from the ground state of $^4I_{15/2}$ to the excited states. The photo- and radio-luminescence spectra showed Er^{3+} 4f-4f emissions. Relative light yield under 5.5 MeV alpha-ray irradiation of Er 0.1%:$Y_3Al_5O_{12}$ was estimated to be 63% of that of $Bi_4Ge_3O_{12}$.

1. Introduction

Rare-earth-doped oxide crystals have been used for scintillator applications in the wide fields such as detection for high-energy photons and particles, medical imaging and security applications. For the application of Positron Emission Tomography (PET), the scintillators are required to have fast decay times and the fast 5d-4f emissions of rare-earth ions such as Ce^{3+} and Pr^{3+} are promising. However, for X-ray radiology systems, the decay times of tens of microseconds are acceptable because these application equipped the integrated-type radiation detectors with phosphors that decay more slowly. Then, the emissions of 4f-4f forbidden transitions of trivalent rare-earth ions which decays up to few microseconds are sufficient for these applications. In our previous study, those of rare-earth-doped oxide crystals (e. g., Nd:$Y_3Al_5O_{12}$ (YAG), Tm:$YAlO_3$, Nd:$Lu_3Al_5O_{12}$ (LuAG), Tm:LuAG and Er:LuAG) have been investigated and demonstrate the interesting scintillation performance (Danevich et al., 2005; Sugiyama et al., 2011a,b; 2012; Totsuka et al., 2012). The other oxide crystals are also expected to demonstrate the promising properties as scintillation materials.

Er:YAG crystals have been widely used for solid-state laser materials which has an emission at 2.94 μs wavelength (Zhekov et al., 1989). They are useful for surgery applications because water has strong absorption in this wavelength (Majaron et al., 1998; Gutknecht et al., 2011). Thus, Er:YAG crystals have been investigated as laser emitting substrate. However, the scintillation properties of that have not been studied well. In this report, Er:YAG single crystals have been prepared by the micro-pulling down method (μ-PD) (Yoshikawa et al., 2004), and their scintillation and luminescent properties with 4f-4f transitions of Er^{3+} ions are evaluated and discussed.

2. Experimental procedure

Crystal growth experiments were performed by the μ-PD method with a radio-frequency heating system and an iridium

* Corresponding author. Tel.: +81 22 215 2214; fax: +81 22 215 2215.
E-mail address: yamaji-a@imr.tohoku.ac.jp (A. Yamaji).

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crucible. Stoichiometric mixture of 4N Y_2O_3, α-Al_2O_3 and Er_2O_3 powders was used as starting materials. Nominally, Y^{3+} was substituted by Er^{3+} according to the formula of $(Er_x, Y_{1-x})_3Al_5O_{12}$ ($x = 0.001, 0.010. 0.10, 0.30, 0.50$). The crystals were grown in argon atmosphere to avoid oxidation of the iridium crucible. The typical growth rate was 6 mm/h and an undoped YAG single crystal was used as a seed. The as-grown crystals were cut and mechanically polished for optical and scintillation measurements.

The transmittance spectra of the crystals were measured in the spectral range from 190 to 900 nm by using a spectrophotometer (JASCO V-550). The photoluminescence spectra and decays were recorded by using a spectrofluorometer (Edinburgh Instrument FLS-920) with a Xe arc lamp (Edinburgh Instrument Xe900) and a steady-state hydrogen microsecond flash lamp (Edinburgh Instrument μF920H) as excitation sources, respectively. The radioluminescence spectra were also obtained by the similar setting under 5.5 MeV α-rays irradiation from an ^{241}Am radio-isotope. To determine the light yields, the pulse height spectra of the samples were measured under 5.5 MeV α-rays irradiation. For the pulse height measurements, the samples were mounted on a photomultiplier (Hamamatsu R7600) with optical grease (OKEN 6262A) and the side of those were covered by a Teflon tape to prevent the scintillation luminescence from escaping. The as-produced signals from the photomultiplier were fed into a preamplifier (ORTEC 113), a shaping amplifier (ORTEC 572) with a shaping time of 10 μs and a multichannel analyzer (Amptek Pocket MCA 8000A) in the pulse height mode.

3. Results

Crystals of nominally 0.1, 1.0, 10, 30 and 50% Er doped YAG crystals were prepared according to the μ-PD method. The fiber-shaped crystals were grown with diameters of 1–3 mm, and the fabricated measurements samples were $1 \times 2 \times (1–3)$ mm^3.

Transmittance spectra of Er:YAG in the wavelength range from 190 to 900 nm are shown in Fig. 1. Several sharp absorption bands due to the Er^{3+} 4f-4f transitions are observed and the dominant absorption bands at 256, 381, 524 and 646 nm are attributed the transition from the ground state of $^4I_{15/2}$ to the $^4D_{7/2}$, $^4G_{11/2}$, $^2H_{11/2}$ and $^4F_{9/2}$, respectively (Dieke and Crosswhite, 1963). The absorption intensities increase with the increase of Er concentrations.

Photoluminescence spectra of Er:YAG crystals under 256 nm excitation exhibited the intense peaks at 404 and 473 nm, as shown in Fig. 2. These peaks can be assigned to the $^2H_{9/2} \rightarrow {}^4I_{15/2}$ and $^2H_{9/2} \rightarrow {}^4F_{7/2}$ transitions from the energy level diagram for the Er^{3+} ion in the garnet crystals (Dieke and Crosswhite, 1963; Sugiyama et al., 2012). The photoluminescence decay curve for $^4D_{7/2} \rightarrow {}^4I_{15/2}$ of Er 0.1%:YAG is provided in Fig. 3 and the excitation and emission spectra were 256 and 404 nm, respectively. The decay time was deduced by single exponential fitting and the value was 52.6 μs. The other samples were also recorded and the relationship between decay time and nominal Er concentration is plotted in Fig. 4. The values of Er 1%, 10% and 30% were 37.2, 13.1 and 12.5 μs, respectively. That of Er 50%:YAG was not able to measured due to the low luminescent intensity. The decay times were exponentially decrease with the increase of Er concentration, and which would be due to the concentration quenching and this result was similar to that of previous study (Xu et al., 2002).

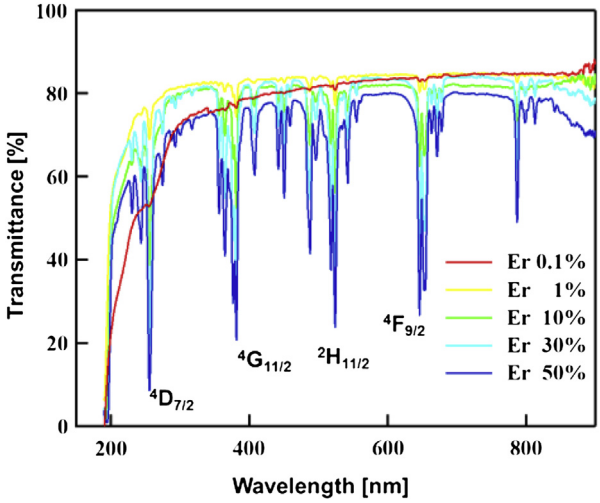

Fig. 1. Transmittance spectra of Er:YAG in the spectral range from 190 to 900 nm.

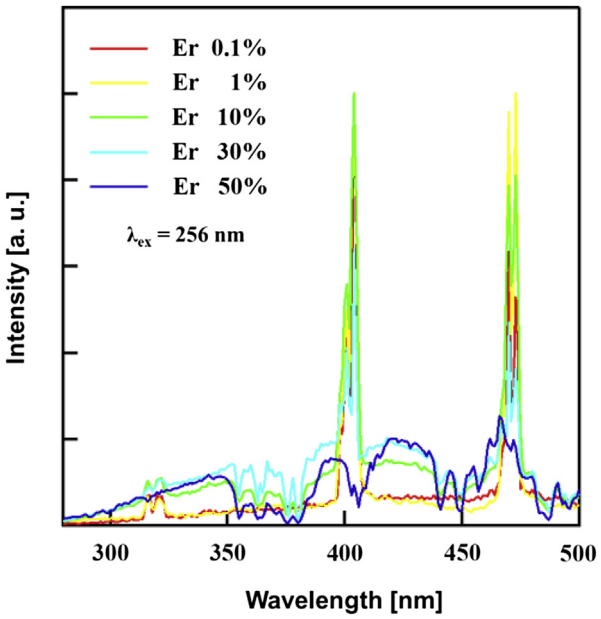

Fig. 2. Photoluminescence spectra of Er:YAG under 256 nm excitation.

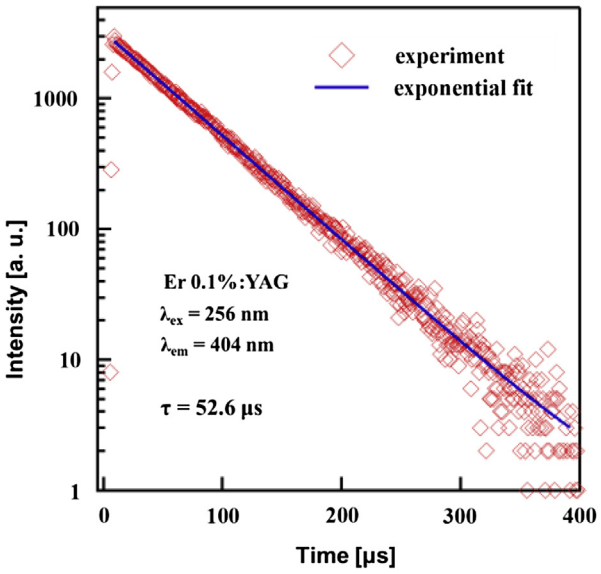

Fig. 3. Photoluminescence decay profile of Er 0.1%:YAG, and the excitation and emission wavelength were $\lambda_{ex} = 256$ nm and $\lambda_{em} = 404$ nm, respectively.

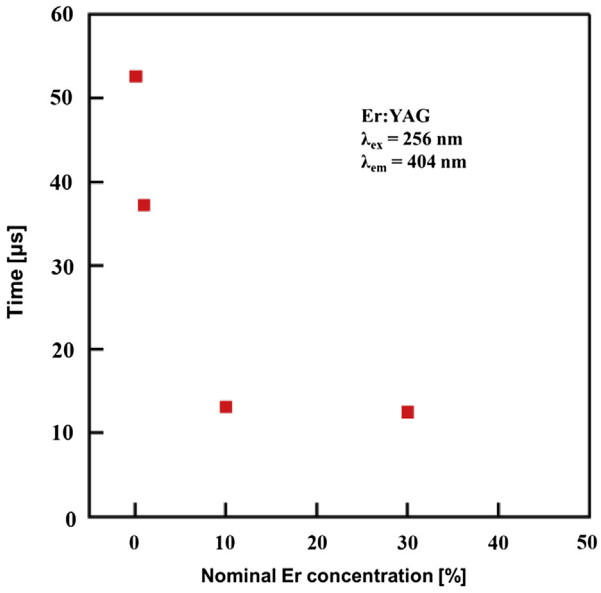

Fig. 4. Photoluminescence decay of Er:YAG plotted against the nominal Er concentrations.

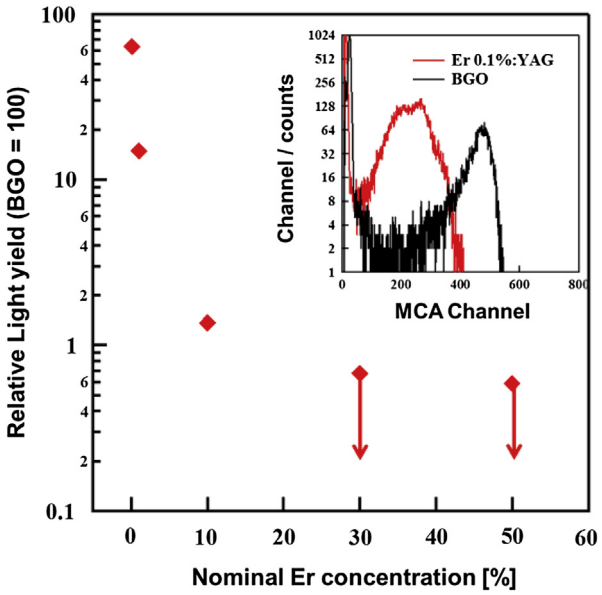

Fig. 6. Pulse height spectra of Er 0.1%:YAG and BGO under 5.5 MeV alpha-ray irradiation (inset). Relative light yields of Er:YAG under 5.5 MeV alpha-ray plotted against nominal Er concentrations.

The radioluminescence spectra of Er:YAG crystals in spectra range from 200 to 700 nm under 5.5 MeV alpha-ray irradiation are presented in Fig. 5. There were two intense peaks at 408 and 475 nm and these peaks could be assigned to the $^2H_{9/2} \rightarrow {}^4I_{15/2}$ and $^2H_{9/2} \rightarrow {}^4F_{7/2}$ transitions like those of photoluminescence spectra. However, in the case of Er 50%:YAG, the luminescence peak at 561 nm was the most intense in the measured spectra range and which was corresponding to the $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transition. The excess Er^{3+} would also function as the host structure similar to that of $Er_3Al_5O_{12}$ (Tanner et al., 2004) and the band structure would be changed.

The example of the pulse height spectra of Er 0.1%:YAG and BGO under 5.5 MeV alpha-ray irradiation are shown in Fig. 6 (inset). The relative light yields were calculated from the position of the total absorption peak compared with that of $Bi_4Ge_3O_{12}$ (BGO) which is a commercial scintillator crystal (Holl et al., 1988). To calculate simply, it is assumed that the peak channels of Er:YAG are dominated by the emission at 410 nm. The quantum efficiency of the photomultiplier were 33% at 410 nm (Er:YAG) and 28% at 480 nm (BGO), respectively. Here, the relative light yield of BGO was normalized to the value of 100%. The values of the relative light yields of Er 0.1, 1.0, 10, 30 and 50% doped YAG were determined to be 63, 15, 1.3, 0.68 and 0.59%, respectively, as shown in Fig. 6. The light yields decreased with an increase of Er concentration because the excess Er doping would cause the concentration quenching.

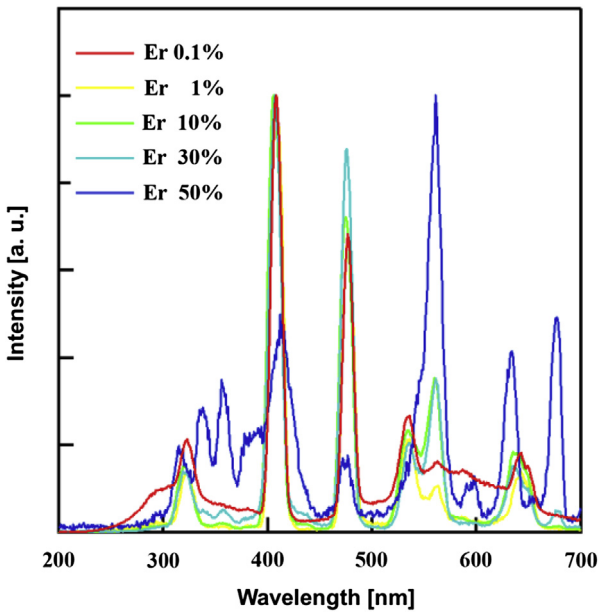

Fig. 5. Radioluminescence spectra of Er:YAG under 5.5 MeV alpha-ray irradiation.

4. Conclusions

Nominally $(Er_x\ Y_{1-x})_3Al_5O_{12}$ (x = 0.001, 0.010, 0.10, 0.30, 0.50) single crystals were grown by the μ-PD method. The absorption lines due to the Er^{3+} 4f-4f transitions were observed and these intensities increased with the increase of Er concentrations. In the photo- and radio-luminescence spectra measurements, Er^{3+} 4f-4f emission peaks were detected under 256 nm excitation and 5.5 MeV alpha-ray irradiation, respectively. According to the pulse height spectra under 5.5 MeV alpha-ray irradiation, Er 0.1% doped YAG demonstrated the highest light yield among all samples, and the value of this sample was estimated to be 63% of that of BGO.

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