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Tm Concentration Dependence on Scintillation Properties of $Y_3Al_5O_{12}$ Garnet Single Crystals

Toshiaki Kunikata,[1*] Prom Kantuptim,[2] Daiki Shiratori,[3] Takumi Kato,[1] Daisuke Nakauchi,[1] Noriaki Kawaguchi,[1] and Takayuki Yanagida[1]

[1]Nara Institute of Science and Technology, 8916-5 Takayama, Ikoma, Nara 630-0192, Japan
[2]Department of Physics, Faculty of Science, King Mongkut's University of Technology Thonburi, Bangkok 10140, Thailand
[3]Tokyo University of Science (TUS) 6-3-1 Niijuku, Katsushika-ku, Tokyo 125-8585, Japan

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Tm-doped $Y_3Al_5O_{12}$ (YAG) single crystals with different Tm concentrations (0.1, 0.5, 1.0, and 2.0%) were grown using the floating zone method. X-ray-induced scintillation spectra, scintillation decay curves, and γ-ray-induced pulse height spectra of the obtained Tm-doped YAG single crystals were investigated. All the samples showed emission peaks due to the 4f–4f transitions of Tm^{3+} ions, and the decay times were around 40–20 μs under X-ray excitation. The 0.5% Tm-doped YAG sample showed the highest light yield under 662 keV γ-ray excitation among the samples. The optimal dopant concentration of Tm-doped YAG single crystals was confirmed to be 0.5%.

1. Introduction

Radiation detectors play an important role in various applications, such as medical diagnoses,[1,2] security screenings,[3,4] and industrial inspections.[5] A high proportion of radiation detectors are equipped with scintillators that convert ionizing radiation into a lot of low-energy photons. Scintillators are coupled with photodetectors including a photomultiplier tube (PMT) and a Si photodiode. Therefore, the performance of scintillators strongly affects that of radiation detectors. Various scintillators, such as films,[6] ceramics,[7–12] glasses,[13–22] single crystals,[23–32] and nanoparticles,[33] have been investigated to date. Single crystal materials tend to show excellent properties and are widely used as scintillators.

Yttrium aluminum garnet (YAG, $Y_3Al_5O_{12}$) single crystals are one of the good candidates for scintillators because they have high radiation resistance and acceptable density (4.57 g/cm^3) for X-ray detection, and can form a substitutional solid solution with rare-earth ions. In particular, there are many reports on the scintillation properties of Ce-doped YAG single crystals with fast decay time and high light yield.[34–37] Although other rare-earth ions can also show attractive luminescence properties, there were relatively few studies on those of other rare-earth-ion-doped YAG single crystals.

*Corresponding author: e-mail: kunikata.toshiaki.kt1@ms.naist.jp
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In this study, the scintillation properties of Tm-doped YAG single crystals were investigated. Tm^{3+} ions can show bright blue-green luminescence due to 4f-4f transitions, and emission wavelengths match a wavelength range at which typical PMTs show high sensitivities. In a previous study, the scintillation properties of 0.5% Tm-doped YAG and 0.5% Tm-doped $Lu_3Al_5O_{12}$ (LuAG) single crystals, which were grown by the micro-pulling down method, were evaluated, and photoabsorption peaks were observed under γ-ray excitation.[38] However, the Tm concentration dependence on the scintillation properties of Tm-doped YAG single crystals has not yet been investigated. In this paper, we show the Tm concentration dependence on the scintillation properties of Tm-doped YAG single crystals.

2. Materials and Methods

Tm-doped (0.1, 0.5, 1.0, and 2.0%) YAG single crystals were grown by the floating zone (FZ) method. The chemical composition is represented by $(Tm_x,Y_{1-x})_3Al_5O_{12}$ (x = 0.001, 0.005, 0.01, and 0.02); 0.1, 0.5, 1.0, and 2.0% correspond to x = 0.001, 0.005, 0.01, and 0.02, respectively. The raw powders of Y_2O_3 (99.99%; Furuuchi Chemical), Al_2O_3 (99.99%; Kojundo Chemical Laboratory), and Tm_2O_3 (99.99%; Furuuchi Chemical) were mixed and ground using an agate mortar and pestle. The mixed powders became rod-shaped by applying hydrostatic pressure and were sintered at 1400 °C for 10 h using an electric furnace. The sintered rods were partially melted and gradually solidified using an optical FZ furnace (Canon Machinery, FZD0192). The pull-down rate was set to 3 mm/h during crystal growth. The obtained Tm-doped YAG single crystals were cut and polished using a polishing machine (Buehler, MetaServ 250).

Photoluminescence quantum yields (PL QYs) were evaluated using Quantaurus-QY (C11347, Hamamatsu Photonics). The X-ray diffraction (XRD) patterns and scintillation properties of the obtained samples were investigated. The XRD patterns of the samples were measured in the range of 10–70° with an X-ray diffractometer (Rigaku, MiniFlex 600). The X-ray-induced scintillation spectra of the samples were evaluated using our original setup equipped with an X-ray generator (Spellman, XRB80N100/CB).[39] In this measurement, the tube voltage and operation current of the X-ray generator were set to 40 kV and 1.2 mA, respectively. The X-ray-induced scintillation decay curves of the samples were measured using our other original setup.[40] Pulse height spectra were measured under γ-ray irradiation from a ^{137}Cs sealed source (662 keV). The samples were optically coupled with a PMT (Hamamatsu Photonics, R7600U-200) whose applied voltage is −700 V. The shaping time was set to 10 μs. Signals from the PMT were analyzed using a preamplifier, a shaping amplifier, and an analog-to-digital converter, which are shown in a previous report.[41] The light yield of the 0.5% Tm-doped YAG sample was estimated by comparison with that of the commercially available Ce-doped Gd_2SiO_5 single crystal (7000 photons/MeV).

3. Results and Discussion

Figure 1 shows the Tm-doped YAG single crystals obtained in this study. The thicknesses of the samples were approximately 1.0 mm. All the samples were transparent and the 1.0 and

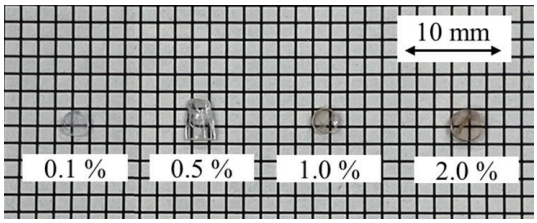

Fig. 1. (Color online) Appearance of Tm-doped YAG samples.

2.0% Tm-doped YAG samples showed brown color. In general, the colors of single crystals can be changed by the presense of oxygen vacancies. There is a possibility that the brown colors of the 1.0 and 2.0% Tm-doped YAG samples were caused by the effect of oxygen vacancies. Figure 2 shows the XRD patterns of the Tm-doped YAG samples and the reference data of YAG (Crystallography Open Database, No. 2003066). All the XRD peaks of the Tm-doped YAG samples were ascribed to that of the reference data of YAG. No undefined phases were observed in this measurement.

The PL QYs of the 0.1, 0.5, 1.0, and 2.0% Tm-doped YAG samples were 17, 31, 9, and 7%, respectively. In the 1.0–2.0% Tm-doped samples, the PL QY decreased with increasing dopant concentration owing to the concentration quenching phenomenon.

Figure 3 shows the X-ray-induced scintillation spectra of the Tm-doped YAG samples. The multiple emission peaks due to the 4f-4f transitions of Tm^{3+} ions were observed at around 250–550 nm. The observed emission peaks are consistent with those of Tm-doped materials in the previous reports.[38,42,43] The peaks at around 280, 350, 460, and 520 nm are ascribed to $^1I_6$-$^3H_6$, $^1D_2 \rightarrow {}^3H_6$, $^1G_4 \rightarrow {}^3H_6$, and $^1D_2 \rightarrow {}^3H_5$ transitions, respectively. The 0.1 and 0.5% Tm-doped samples showed the broad emission in the wavelength range from 300 to 450 nm. This can be ascribed to the intrinsic luminescence of YAG, which is explained by emissions due to defects[38] or self-trapped excitons.[44]

Figure 4 shows the X-ray induced scintillation decay curves of the Tm-doped YAG samples and instrument response function (IRF). The scintillation decay curves were fitted to single exponential functions in the time ranges that did not overlap with that of the IRF component. The estimated scintillation decay times of the 0.1, 0.5, 1.0, and 2.0% Tm-doped YAG samples were 43, 44, 34, and 23 μs, respectively. The scintillation decay times are roughly consistent with that of Tm-doped YAG shown in a previous report.[38] When the Tm concentration was 0.5–2.0%, the scintillation decay time decreased with increasing dopant concentration. This seems to be due to the concentration quenching phenomenon, similar to the results of PL QY.

Figure 5 shows the pulse height spectra of the Tm-doped YAG samples under γ-ray irradiation from the ^{137}Cs sealed source. The photoabsorption peaks of the 0.1, 0.5, 1.0, and 2.0% Tm-doped YAG samples were observed at 585, 705, 410 and 240 channels, respectively. The light yield of the 0.5% Tm-doped YAG sample is the highest among the samples according to the peak positions. The estimated light yield of the 0.5% Tm-doped YAG sample was 3500 photons/MeV. This value was lower than those of Tm-doped YAG (11000

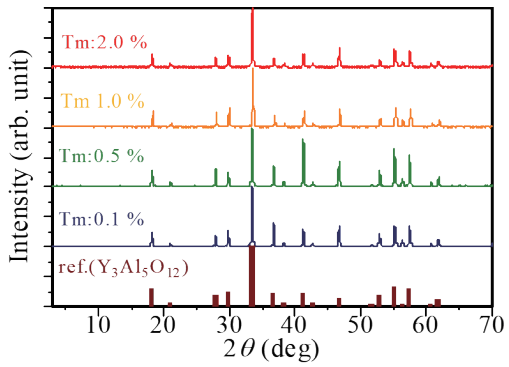

Fig. 2. (Color online) XRD patterns of Tm-doped YAG samples and reference data of YAG.

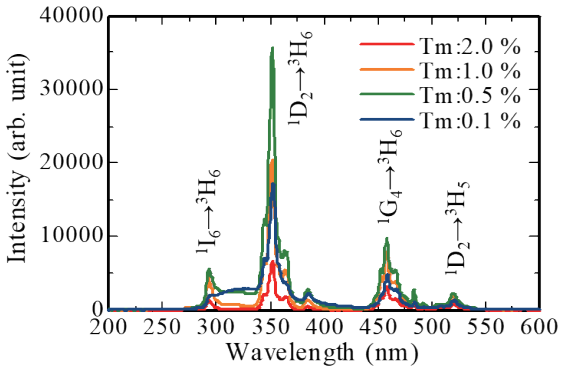

Fig. 3. (Color online) X-ray-induced scintillation spectra of Tm-doped YAG samples.

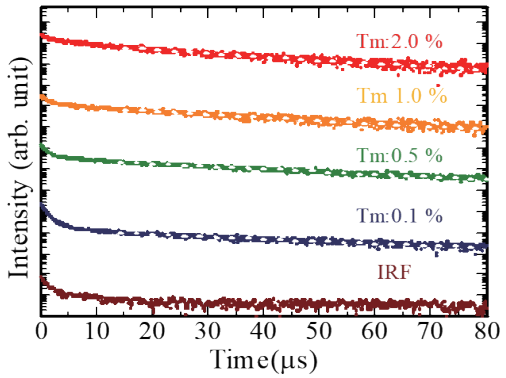

Fig. 4. (Color online) X-ray-induced scintillation decay curves of Tm-doped YAG samples and IRF of the equipment.

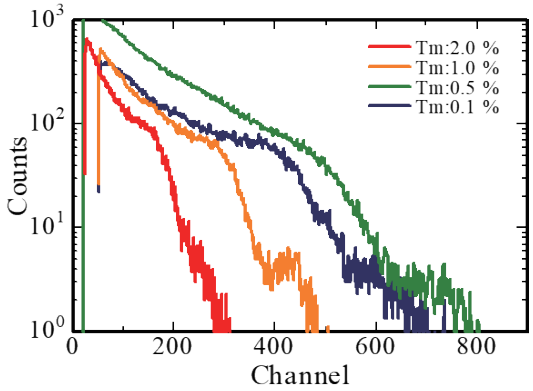

Fig. 5. (Color online) Pulse height spectra of Tm-doped YAG samples under γ-ray irradiation.

photons/MeV) and Tm-doped LuAG (6000 photons/MeV).[38] There are two possible reasons for this result. One is the difference in shaping time. The shaping time in our measurement was 10 μs (the maximum value of our setup), whereas that time in the previous study was 30 μs.[38] The obtained light yields could be underestimated. The other possible reason is the difference in the single crystal growth method used. The FZ method was used in this study, whereas the micro-pulling down method was used in the previous study.[38] In general, the light yields of single crystals were strongly affected by defects. Defect formation during single crystal growth depends on the growth method used. Figure 6 shows the normalized light yields of the Tm-doped YAG samples under γ-ray irradiation. In the 1.0–2.0% Tm-doped YAG samples, the light yield tended to decrease with increasing dopant concentration, similar to the results of PL QY. This result can be explained by the concentration quenching phenomenon.

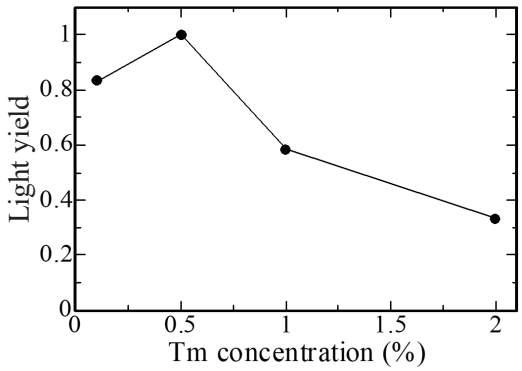

Fig. 6. Normalized light yields of Tm-doped YAG samples.

4. Conclusions

We grew Tm-doped YAG single crystals with different Tm concentrations (0.1, 0.5, 1.0, and 2.0%) by the FZ method and evaluated their XRD patterns and scintillation properties. The XRD patterns of the Tm-doped YAG samples were ascribed to the reference data of YAG. The obtained scintillation spectra and scintillation decay times were consistent with those in a previous report[38] on Tm-doped YAG. In the pulse height spectra under γ-ray irradiation, the 0.5% Tm-doped YAG sample showed the highest light yield. The scintillation decay times and light yields of the 1.0 and 2.0% Tm-doped YAG samples were lower than those of the 0.5 % Tm-doped YAG sample, which can be explained by the concentration quenching phenomenon.

Acknowledgments

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