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Comparative Study of Optical and Scintillation Responses of $Sr_3NbGa_3Si_2O_{14}$ (SNGS) and $La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA) Crystals

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Optical and scintillation properties of $Sr_3NbGa_3Si_2O_{14}$ (SNGS) single crystal were evaluated for the first time and compared with those of $La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA), which was previously reported to exhibit such properties. SNGS showed ~80% optical transmittance at wavelengths longer than 250 nm. Under 250 nm excitation, a strong photoluminescence (PL) appeared at 400 nm with a primary decay time of 1.4 ns. In an X-ray-induced radioluminescence spectrum, an intense emission peak at 400 nm was observed, and the primary scintillation decay time was ~1.37 μs. Finally, the absolute scintillation light yield of SNGS turned out to be 850 photons/MeV, which was higher than that of LTGA.

1. Introduction

Piezoelectric materials are one of the functional materials that convert stress energy to electric energy and are widely used for sensors. Up to now, many materials have been investigated for this application. Among them, $La_3Ta_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LTGA) and Sr3NbGa$_3$Si$_2$O$_{14}$ (SNGS) single crystals have langasite-type structures, and they are famous for their piezoelectric characteristics even at temperatures higher than 1000 °C.[(1)] Recently, we have studied optical properties, such as photoluminescence (PL) and radioluminescence (RL, also called scintillation), of other langasite crystals including $Ca_3NbGa_3Si_2O_{14}$ (CNGS), $La_3Ga_5SiO_{14}$ (LGS), $La_3Nb_{0.5}Ga_{5.3}Al_{0.2}O_{14}$ (LNGA), and LTGA.[(2,3)] In previous studies, we found that these langasite crystals exhibited an interesting feature of very fast sub-ns decay in the visible wavelength range and a detectable scintillation

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signal.[2,3] Therefore, it was proved that langasite crystals can be potentially suitable for scintillator applications.

Scintillators are one of the phosphors that convert a high-energy ionizing radiation to hundreds of visible-ultraviolet photons immediately via large scale quantum cutting.[4] Scintillation detectors, usually consisting of scintillator materials and photodetectors, have played a major role in ionizing radiation detectors for use in medical,[5] security,[6] well-logging,[7] astrophysics,[8] and particle physics applications.[9] Although most commercial scintillators are Ce^{3+}-doped,[10] since Ce^{3+} 5d-4f transition is parity- and spin-allowed, nondoped materials (*e.g.*, ZnO[11]) have also attracted much attention in terms of reducing the cost of rare earths that are generally doped into scintillators as emission centers. Generally, nondoped materials exhibit scintillation due to some defects or excitons. In addition, langasite crystals are attractive for scintillation detectors since they are relatively heavy and easy to fabricate in a bulk shape using conventional melt-growth techniques.

In the present study, we focused on investigating one of the langasite-type materials, SNGS, in comparison with LTGA, which was the brightest among the langasite materials previously investigated.[2,3] Up to now, the transmittance of SNGS has been reported several times,[12–14] but no study about emission properties has been carried out.

2. Experimental Procedure

Samples had an area of 4 × 4−7 mm^2 and a thickness of 1 mm. Wide surfaces of samples were optically polished. Figure 1 shows a picture of the samples used in this study. LTGA was orange, while SNGS was visibly colorless and transparent.

Transmittance and PL spectra were evaluated using JASCO V670 and Hamamatsu Quantaurus-QY, respectively. We used Quantaurus-τ (Hamamatsu) for PL decay time investigation with an excitation wavelength of 280 nm and a monitoring wavelength of 400 nm for SNGS. In the case of LTGA, the monitoring wavelength was 500 nm under 280 nm excitation. Although the excitation peak of SNGS was around 250 nm, 280 nm was the limit of this instrument.

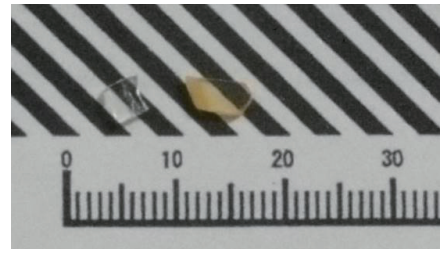


Fig. 1. (Color online) Picture of SNGS (right) and LTGA (left).

As a scintillation response, RL spectra were corrected using an X-ray generator equipped with a Cu target supplied with 80 kV bias voltage and 1 mA tube current. The emission was measured using an Andor DU-420-BU2 CCD spectrometer, and a detailed explanation for the setup was previously reported.[15] X-ray-induced decay time kinetics were evaluated using a pulse X-ray streak system[16] to observe the rise part and an afterglow characterization system with the fast mode[17] for the entire decay time profile. Finally, scintillators were coupled with photomultiplier tube (PMT) R7600-200 (Hamamatsu) with optical grease, and a ^{137}Cs γ-ray or ^{241}Am α-ray were irradiated. The setup and data flow were also reported previously[18]

3. Results and Discussion

Transmittance spectra of SNGS and LTGA are displayed in Fig. 2. The transmittance reached 80% at a wavelength longer than 250 nm in SNGS, while some absorption bands due to defects appeared around 250–500 nm in LTGA. The observed result of LTGA was similar to that previously reported.[2] The absorption edge of SNGS appeared around 250 nm. Previously, another langasite material, LGS, was investigated. LGS also showed an absorption edge around 250 nm.[19]

In Fig. 3, the PL emission map of SNGS is shown. A strong emission was observed around 400 nm under 280 nm excitation. The quantum yield (QY) of SNGS was not high (~1%). In LTGA, an emission peak appeared around 500 nm under 280 nm excitation (not shown here), and this was consistent with the previously reported result.[2] Compared with LTGA, the QY of which was < 0.1%, SNGS was brighter in PL.

The PL decay time profile of SNGS monitored at 400 nm under 280 nm excitation is shown in Fig. 4. The decay time of SNGS was long (1.4 μs). On the other hand, that of LTGA was 8.9 ns, which is consistent with that in the previous work.[2] The decay time

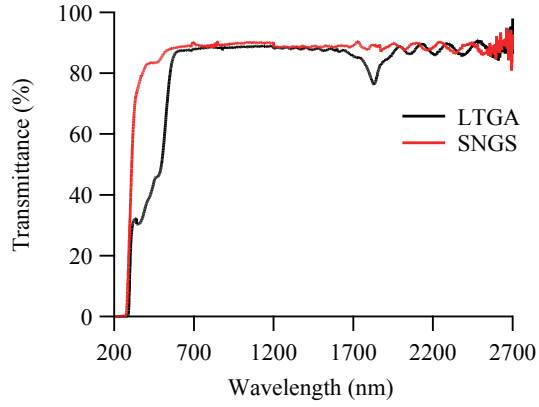
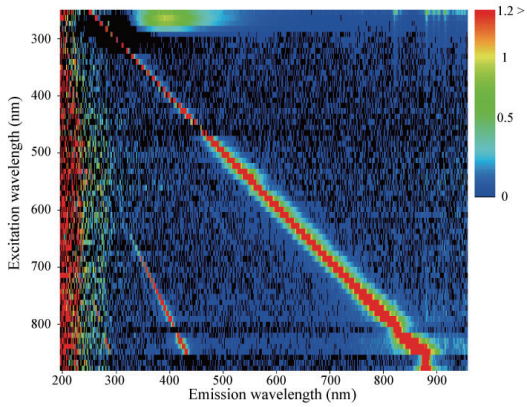


Fig. 2. (Color online) Transmittances of SNGS and LTGA.



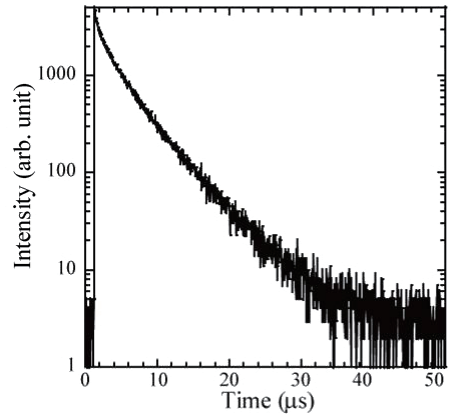


Fig. 3 (left). (Color online) PL emission map of SNGS. The horizontal and vertical axes show the emission and excitation wavelengths, respectively.
Fig. 4 (right). PL decay time profile of SNGS.

of SNGS was similar to that of the defect emission of ZnO.[11] In our previous studies, we examined the PL decay of some langasite materials, and only LTGA exhibited a detectable signal; thus SNGS was a relatively bright material compared with other langasite crystals.

X-ray-induced RL spectra of SNGS and LTGA at room temperature are shown in Fig. 5. Compared with LTGA, SNGS showed a high luminescence intensity. Since the excitation band (280 nm) of SNGS had a lower energy than the bandgap (see Fig. 2), the origin of the emission was not the self-trapped exciton. In LTGA, the origin of the emission around 425 nm was considered to be the F^+ center;[2] thus a similar emission mechanism due to lattice defects could be expected in SNGS. As observed in the case of PL, SNGS was brighter than LTGA in scintillation.

Figure 6 shows the ^{137}Cs 662 keV γ-ray- and ^{241}Am 5.5 MeV α-ray-irradiated pulse height spectra. Although the photoabsorption peak was not very sharp, the peak and Compton edge were observed. In a comparison with the light yield of the calibrated Sn-doped glass scintillator with a similar emission wavelength,[20] that of SNGS turned out to be 850 ph/MeV. In other langasite-type materials, the pulse height spectrum could not be observed;[2,3] thus the light yield of SNGS was considerably higher than those of the other langasite-type scintillators studied so far. At the same time, the ^{241}Am α-ray-induced pulse height spectrum was also evaluated, and the light yield under 5.5 MeV α-ray was 640 ph/5.5 MeV-α. Such a difference under γ-ray and α-ray excitations sometimes occurs owing to the Linear Energy Transfer (LET) effect caused by the difference in excitation density. In an extreme case, completely different emission centers are excited by the change in emission wavelength or decay time. Clear experimental results of the LET effect can be found in the RL spectrum of Ag-doped phosphate glass[21] for dosimeter

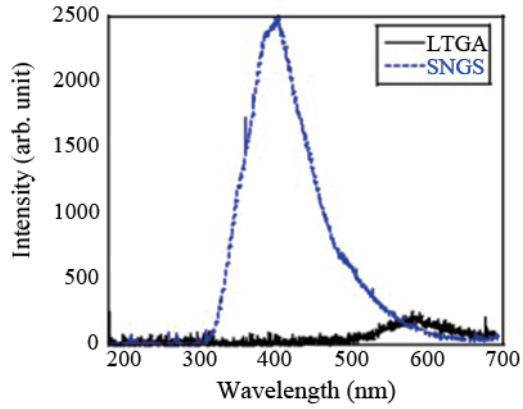
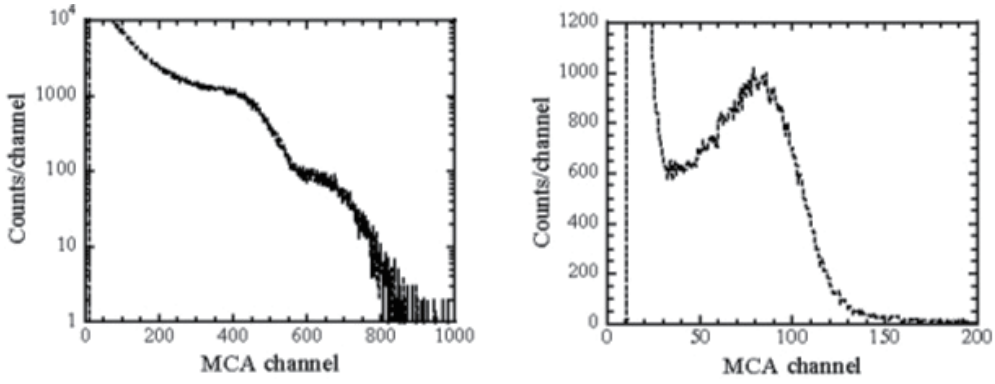


Fig. 5. (Color online) X-ray induced RL spectra of SNGS (dotted line) and LTGA (solid line).



(a) (b)

Fig. 6. (a) ^{137}Cs γ-ray- and (b) ^{241}Am α-ray-induced pulse height spectra of SNGS.

application and Ce-doped $LiCaAlF_6$ in scintillator usage.[22] Up to now, langasite crystals have not shown a detectable pulse height spectrum, and SNGS has shown the brightest scintillation among the langasite crystals introduced so far.

Figure 7 shows the scintillation decay time profiles of SNGS and LTGA. In our previous study,[2] we could not measure the decay time profile of LTGA but, owing to the development of the new characterization system,[17] we could observe it in this study. The scintillation decay time of LTGA was 8 ns, which is similar to that in PL decay. On the other hand, the scintillation decay time of SNGS was relatively long (1.4 μs), which is consistent with the increase in pulse height observed when the shaping time was monotonically increased up to 10 μs (maximum of the instrument). In SNGS, we also observed the rise part. The rise part of the scintillation mainly depends on the energy

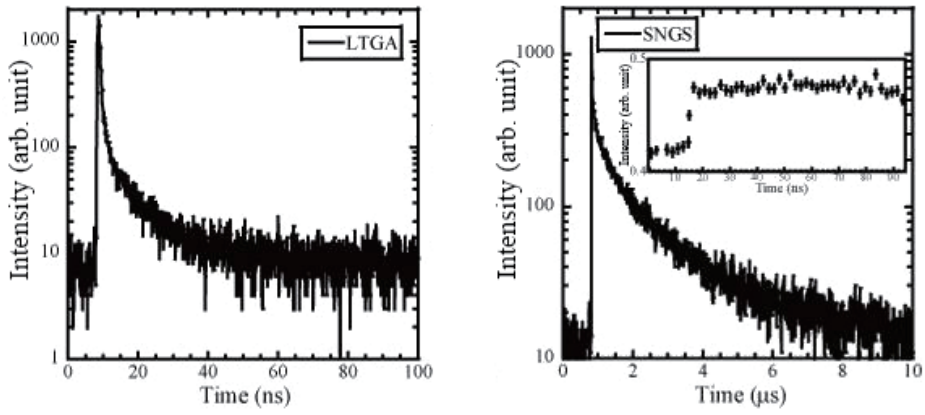


Fig. 7. X-ray-induced scintillation decay times of (a) LTGA and (b) SNGS. The inset of SNGS shows the rise part.

migration from the host to emission centers. The rise level of SNGS was similar to the instrumental response (80 ps), indicating the smooth energy migration from the host to emission centers.

4. Conclusion

Optical and scintillation properties of SNGS single crystal were tested and compared with those of LTGA crystal. SNGS exhibited a high transmittance of 80% in the wavelength range longer than 250 nm. In PL and RL, one intense emission peak at 400 nm was observed. The PL decay was 1.37 μs, which is similar to the scintillation decay (1.4 μs). By irradiating with ^{137}Cs γ-rays and ^{241}Am α-rays, the absolute scintillation light yield was determined to be 850 ph/MeV and 640 ph/5.5 MeV-α, respectively.

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