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A new radiation damage phenomenon of LSO:Ce scintillation crystal

Laishun Qin, Yu Pei, Sheng Lu, Huanying Li, Zhiwen Yin, Guohao Ren*

Shanghai Institute of Ceramics, Chinese Academy of Sciences (SICCAS), No. 215 Chengbei Road, Jiading, Shanghai 201800, P.R. China

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Abstract

Cerium-doped lutetium oxyorthosilicate (LSO:Ce) is an excellent scintillator with a large radiation hardness. But, in this work, some LSO:Ce crystals grown by SIC showed a new radiation damage phenomenon after exposure to ^{60}Co γ-rays. An absorption band peaking at near 430 nm was observed. Because of the absorption the emission spectra were distorted but no new emission center appeared. There is no new thermoluminescence (TL) peak in the TL curve but the relative intensity of peaks is different from that of normal samples. The peaks at 490 and 546 K may be attributed to the appearance of absorption band that was proved by annealing at different temperatures. Absorption band can be eliminated a little at 200 °C and completely at 300 °C, either in air or in N_2 atmosphere.

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Keywords: LSO:Ce; Radiation damage; Optical absorption

1. Introduction

Cerium-doped lutetium oxyorthosilicate ($Lu_2(SiO_4)O$:Ce or abbreviated to LSO:Ce) was initially investigated as a phosphor by Gomes et al. [1]. In the year 1989, LSO:Ce crystal was first successfully grown and found to be a promising inorganic scintillator [2–4]. It has a density of $\rho = 7.4\,g/cm^3$, a decay time of approximately 40 ns, light output larger than 25,000 Ph./MeV, and other good physical and chemical properties. Therefore, it is thought to be one of the best scintillators as scintillation properties are comprehensively concerned.

LSO:Ce crystals used in high-energy physics experiments are required to have high resistance to radiation. The radiation damage of LSO:Ce from CTI Company was studied by Kobayashi [5]. It was found that LSO:Ce was radiation hard at least up to 10^6 rad and showed only a small amount of damage at 10^8 rad.

*Corresponding author. Tel.: +86 21 5992 6852; fax: +86 21 5992 7184.
E-mail addresses: sicqls@mail.sic.ac.cn (L. Qin), rgh@mail.sic.ac.cn (G. Ren).

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In the present work, LSO:Ce crystals were grown in Shanghai Institute of Ceramics (SIC). Radiation damage caused by ^{60}Co γ-rays and a new radiation damage phenomenon in LSO:Ce crystal was presented.

2. Experiment

LSO:Ce crystals were grown by Czochralski technique in fluent N_2 atmosphere. The Ce concentration in initial charge is at 0.5% relative to Lu, and the Ce concentration in the crystal is expected to be about 0.1%. Most crystals were colorless and transparent but some were yellowish. For experiments, the samples with dimension of $10 \times 10 \times 2\,mm^3$ were cut from these different boules and then the two 10×10 faces were optically polished.

Irradiation of LSO:Ce was carried out at Shanghai Radiation Center with ^{60}Co γ-rays. All samples were irradiated at a dose rate of about 500 rad/h for 48 h. After irradiation the samples were kept in dark before measurement.

Optical transmission spectra for all samples were measured with a Shimadzu UV-2501PC spectrophotometer. Measurements of excitation and emission spectra were performed using a LS50B luminescence spectrometer (made by Perkin Elmer Company) before and after irradiation. Thermoluminescence spectra (TL) were recorded in air from room temperature to 300 °C at a heating rate of 1 k/s using a FJ427A1 type TL recording instrument made by Beijing Nuclear Instrument Factory (BNIF) in China. This instrument is equipped with a GDB-20 photomultiplier tube (PMT) with very low dark-current made by BNIF. TL curve data were automatically calibrated and the background was well reduced by the program provided by BNIF.

3. Results and discussion

Optical transmission spectra of all samples were measured before irradiation, and two kinds of representative curves are shown in Fig. 1. Transmission spectra at short wavelength region are similar to Melcher's result [3], and there are three absorption bands caused by doped Ce ion peaking at 357, 294 and 264 nm, respectively. Meanwhile, a weak absorption band peaking at 430 nm in the transmission spectra of the yellowish samples was found (see dotted line in Fig. 1). This absorption band, however, was not found in optical transmittance spectra of the colorless samples (solid line in Fig. 1).

To study the influence of annealing on the 430 nm absorption band, yellowish samples were annealed at 1000 °C in N_2 atmosphere for 2 h. It is interesting that all yellowish samples turn colorless after annealing. For comparison, the optical transmission spectra of one of the yellowish samples before and after annealing are shown in Fig. 2. Difference between transmission spectra before and after annealing is shown in Fig. 3. An absorption band peaking at 430 nm was found in optical transmittance before annealing.

All samples including the originally colorless and the annealed were exposed to ^{60}Co source. The total dose was calculated up to 2.4×10^4 rad. Fig. 4 shows three kinds of transmission curves after the irradiation. Curve *c* shows a typical transmission spectrum of the annealed or the originally yellowish LSO:Ce crystals after the irradiation. Curve *a* and *b* reveal two different conditions of colorless samples after γ-rays

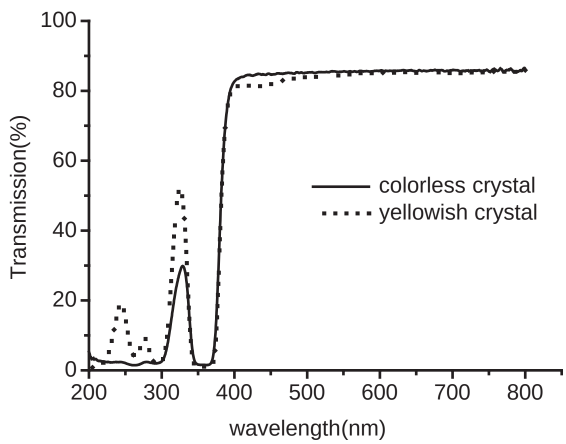

Fig. 1. Transmission spectra of the as-grown colorless and yellowish LSO:Ce crystals.

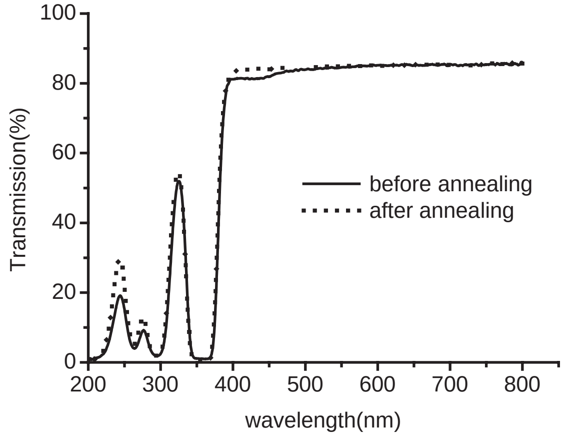

Fig. 2. Optical transmission spectra of the yellowish LSO:Ce before and after annealing.

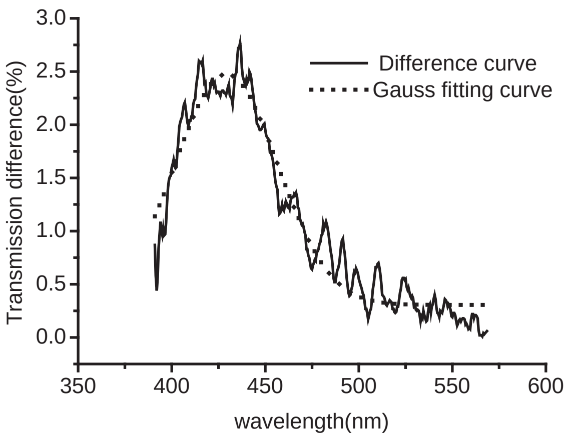

Fig. 3. Difference between transmission spectra before and after annealing and Gauss fitting for yellowish LSO:Ce sample.

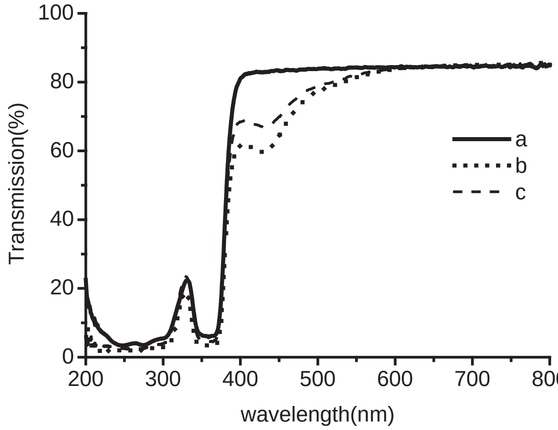

Fig. 4. Optical transmission spectra of LSO:Ce crystals after γ-rays irradiation. Curves *a* and *b* are for two different conditions of colorless crystals. Curve *c* is for the originally yellowish crystal.

irradiation. It was found that there was a very remarkable absorption band near 430 nm not only in the spectra of the yellowish samples (curve *c*), but also in the spectra of some originally colorless samples (curve *b*). This absorption band is similar to the above-mentioned one in the spectra of the yellowish samples before annealing. But this phenomenon does not exist in some other originally colorless samples (curve *a*). The absorption band leads to the decrease by, at least 15%, in transmittance at the emission peak of LSO:Ce, sometimes even up to 25%. After a spontaneous recovery of 50 days, the transmission spectra of the irradiated LSO:Ce crystals were measured again. It was found that the absorption in curve *b* and curve *c* was lowered by 9% and 10% at 430 nm, respectively, but the absorption band still existed and could influence the scintillation light emission. It is distinct that the appearance of the absorption band was brought about after γ-ray radiation. To our knowledge, this radiation effect has not been reported yet.

To investigate the origin of the irradiation-induced absorption band, optical transmission, photoluminescence (PL) and TL spectra of irradiation-damaged LSO:Ce samples before and after irradiation were all measured. Fig. 5 shows difference between transmission spectra before and after irradiation and its Gauss fitting. In this figure, the square scattered line represents the real difference, while the solid line is its Gauss fitting. The transmission difference curve can be decomposed into two curves with the peaks of 390 and 430 nm, respectively. The absorption peak at 430 nm was similar to the peak in the fitting curve of difference spectrum in Fig. 3. The absorption peak at 390 nm is ascribed to the radiation damage reported by Kobayashi [5]. From Fig. 6, the excitation spectra are basically the same as the typical spectra. The peaks of excitation spectrum

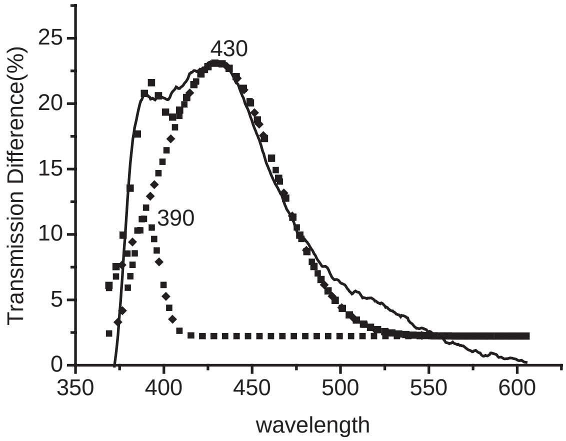

Fig. 5. Difference spectrum between the transmission spectra of the damaged LSO:Ce crystal before and after irradiation and its Gauss fitting.

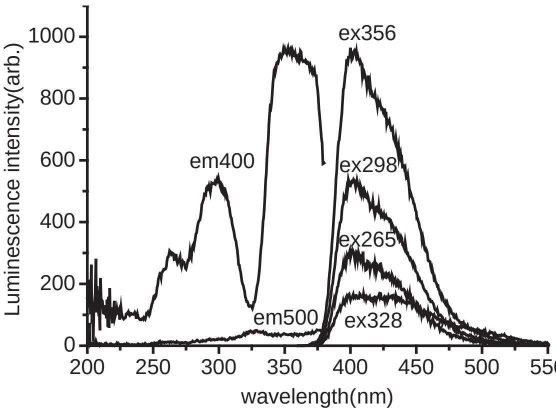

Fig. 6. Excitation and emission spectra of the damaged LSO:Ce crystal after γ-ray irradiation.

of 400 nm are at 265, 298, 356 nm, respectively, and the peaks of 500 nm are at 328 and 374 nm. The emission peak at near 400 nm is also similar to the normal LSO:Ce crystal. However, due to the occurrence of the absorption band leading to a dramatic decrease of photoluminescence intensity near the absorption band, the emission spectra are clearly distorted. In Fig. 7 TL spectrum of the damaged LSO:Ce crystal is juxtaposed with its TL spectrum before irradiation for comparison. Four TL peaks can be observed in both TL curves, approximately situated at 351, 413, 490, and

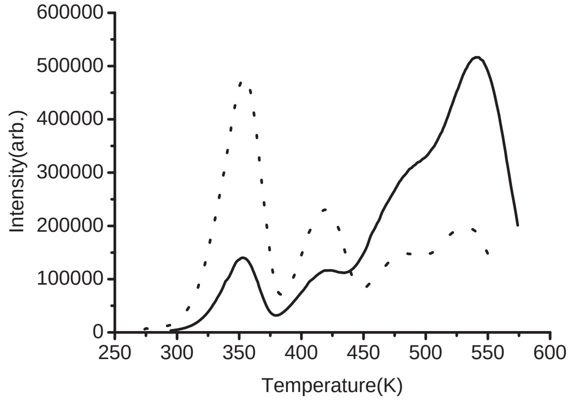

Fig. 7. Thermoluminescence spectra of the radiation-damaged LSO:Ce crystal before (dotted line) and after γ-ray irradiation (solid line).

546 K, respectively. According to the number, the position and the relatively intensity of the peaks, the TL spectrum of the undamaged LSO:Ce crystal are similar to those reported in the literatures [6–8]. However, the TL spectrum of the radiation-damaged LSO:Ce is different from that of undamaged crystal (Fig. 7, dotted line) and that in the past literatures [6–8]. In the TL spectra of the damaged LSO:Ce, the intensity of the two peaks at lower temperatures are relatively weaker than that of the peaks at higher temperatures. If thermal quenching effect is considered, the relative intensity of the two peaks at 490 and 546 K will be higher.

Annealing behavior of the radiation-damaged samples was also investigated. The annealing experiments were carried out in a sealed tube furnace heated by a SiC heater. After annealing, transmission spectra were measured to ascertain the influence of annealing temperature and atmosphere. The results indicated that the absorption in the transmission spectra disappeared completely when the annealing temperature was higher than 300 °C. However, it will not disappear if the annealing temperature is lower than 200 °C. Fig. 8 presents the transmission spectra of the radiation-damaged LSO:Ce before and after annealing in air at 200 and 300 °C, respectively. Annealing experiment was also performed in N_2 atmosphere. Similar to the behavior in air, the absorption also

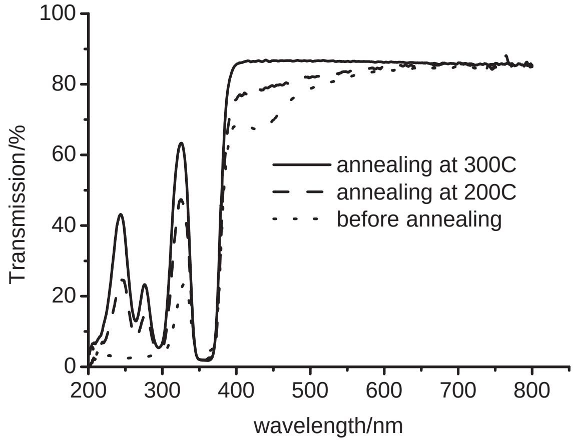

Fig. 8. Transmission spectra of the radiation-damaged LSO:Ce crystal before (dotted line) and after annealing at 200 °C (dashed line)and 300 °C (solid line).

disappeared completely when annealing at 300 °C in N_2, though a longer time like 20 h was needed.

As the 430 nm absorption band would appear in as-grown crystal and after γ-ray irradiation, the absorption would be considered from some kinds of defects in the LSO:Ce crystal. According to TL curves of the radiation-damaged LSO:Ce samples before and after irradiation, we note that although no new TL peak appears, the relative intensity of peaks at 490 and 546 K in the TL spectra of damaged LSC:Ce increases dramatically. This indicates that the defects corresponding to the TL peaks at 490 and 546 K remarkably increased after γ-ray irradiation leading to the appearance of absorption at 430 nm in the transmission spectra. Annealing behavior was consistent with the result of TL investigation. Annealing experiments in air or N_2 atmosphere at different temperatures prove that the absorption is sensitive to annealing temperature only. The absorption at 430 nm can be eliminated after annealing at 300 °C but not at 200 °C. Thus, the absorption should be caused by the defect corresponding to the TL peaks of 490 and 545 K. The physical mechanism of radiation damage is still under investigation.

4. Summary

A new radiation damage phenomenon of LSO:Ce crystal is presented. Some LSO:Ce crystals grown in SIC were damaged by γ-ray irradiation. The absorption band near 430 nm appeared in the transmission spectra of the damaged LSO:Ce. Due to the absorption the photoluminescence spectra of the damaged LSO:Ce were distorted. It is shown in the TL curve of the damaged LSO:Ce that the relative intensity of TL peaks is different from that of the undamaged crystal and that by other authors, too. The peaks at 490 and 546 K should be responsible for the absorption induced by the γ-ray irradiation.

Acknowledgments

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