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Improvement and luminescent mechanism of $Bi_4Si_3O_{12}$ scintillation crystals by Dy^{3+} doping

Bobo Yang [a], Jiayue Xu [a,*], Yan Zhang [a], Haibo Zeng [a], Tian Tian [a], Yaoqing Chu [a], Yubai Pan [b], Qingzhi Cui [a]

[a] Institute of Crystal Growth, School of Materials Science and Engineering, Shanghai Institute of Technology, Shanghai 201418, PR China
[b] Shanghai Institute of Ceramics, Chinese Academy of Science, Shanghai 200050, PR China

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ABSTRACT

$Bi_4Si_3O_{12}$:Dy (BSO:Dy) crystals have been grown by the modified vertical Bridgeman method and doping effects on light yield have been investigated. Doped with small amount of Dy_2O_3 (0.05–0.3 mol%), the light yield and energy resolution of BSO crystals were improved significantly. However, high concentrations of Dy_2O_3 doping resulted in the decrease of light yield. Pulse height measurement under γ-ray irradiation shows that 0.1 mol% Dy_2O_3 doping can make the relative light yield of BSO from 24.6% to 35.8% of BGO crystal, with fast decay time of ~90 ns. X-ray excited radioluminescence spectra showed Dy doping has an extra emission in the host emission band (Bi^{3+} emission) and acts as a sensitizer to the Bi luminescent center. These results indicate that BSO:Dy crystal could be one of promising candidates for replacing BGO in some application such as electromagnetic calorimeter and dual readout in nuclear or high energy physics.

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1. Introduction

Scintillator crystals are used in medical diagnostics, in industrial applications and in scientific application. A spectacular application in the latter field is the use of scintillator crystals in electromagnectic calorimeters. These are used in high energy physics, nuclear physics and astrophysics to count electrons and photons. One of the most famous scintillators is bismuth germanate ($Bi_4Ge_3O_{12}$, BGO) which has many excellent characteristics. Table 1 shows the scintillation characteristics of BGO and BSO [1]. Compared with BGO, bismuth silicate ($Bi_4Si_3O_{12}$, BSO) which has attracted much attention in high energy particle detection has some advantages of faster decay time, higher radiation hardness and lower cost [1–3]. Crystal growth and optical properties of BSO crystal were reported [4–8]. Recent studies [9] show that BSO crystals are excellent materials to be used in 4π electromagnetic calorimeters in the energy region of several hundreds of MeV. What is more, BSO crystals have the better performance than BGO used for dual readout [10–12]. For BSO, the light yield is about 20% of BGO which is an unfavorable property. Doping with rare earth elements is used to improve the scintillation properties of scintillation crystals. The properties of Ce, Nd, Eu, Yb, Dy, Cr and Fe doped BSO crystals had been investigated by many researchers [13–18]. The above studies have no positive effect on improving the light yield of BSO. In this paper, we carried out to find a way for enhancing the light yield of BSO.

The growing interest in the visible luminescence of Dy^{3+} doped oxide hosts is stimulated by practical reasons. The emission of Dy^{3+} originates from the $^4F_{9/2}$ level, Dominating are the transitions to $^6H_{15/2}$ (~470 nm) and $^6H_{13/2}$ (~570 nm). Relatively high light yield and moderate decay time with ns order when coupled with PMT were observed in Dy-doped oxyorthosilicates and garnets [19,20]. BSO is a self activated scintillator, and the luminescence center is Bi^{3+}, which the emission wavelength is about 480 nm [1–4]. Due to the luminescence characteristic of Dy^{3+}, the emission spectra of BSO crystals doped with Dy will respectively differ greatly from that of pure BSO crystal. In this paper, pure and BSO:Dy single crystals were grown in the same condition using the vertical Bridgman method, X-ray excited radioluminescence and scintillation properties were investigated.

2. Experiment

Pure and BSO:Dy crystals with nominal concentrate of 0.05–4 mol% Dy_2O_3 were grown by the modified vertical Bridgman method. High purity Dy_2O_3 (>4N), SiO_2 (>4N) and Bi_2O_3 (>5N) were used as

* Corresponding author. Tel.: +86 021 60873581.
E-mail address: xujiayue@sit.edu.cn (J. Xu).

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starting materials. These starting materials were dried at 300 °C for 3 h before weighing accurately. These starting materials were weighed according to stoichiometric equation ratio of Bi_2O_3:SiO_2 and doping concentrations of Dy_2O_3 (0, 0.05, 0.1, 0.2, 0.3, 1, 2, 3 and 4 mol%) and then mixed in a ball mill coated with polyethylene for 8 h. The mixture was then held at 800 °C for 8 h. After sintering, it was ground to powder and mixed again in the ball mill for 3 h. The samples were then put into the aluminum oxide crucible and held at 850 °C for 12 h to prepare pure and BSO:Dy polycrystalline materials. The feed materials were then loaded into the Pt crucible to grow single crystal in Bridgman furnace. In this work, the polycrystalline powders were melted at 1075 °C which is about 50 °C higher than the molting point of BSO crystal for about 12 h to ensure complete melting. The lowing rate of the crucibles was 0.2–0.5 mm/h. The orientation of the seed crystal was chosen to be (001) in all our growth runs. Fig. 1 showed as-grown BSO crystals doped with 0.1 and 3 mol% Dy_2O_3, respectively.

The grown single crystals were cut along the growth axis and polished into wafers of 2/8 mm thickness for the measurements of optical and scintillation properties. The transmittance spectra were measured by a double-beam ultraviolet-infrared spectrophotometer (Agilent Cary5000). Then, radioluminescence spectra under X-ray excitation were collected using the spectrofluorometer (FLS920, Edinburgh Instrument). The pulse height spectra of γ-ray (0.662 MeV) from ^{137}Cs obtained in BSO and BSO:Dy crystals at room temperature. The crystals were wrapped with Teflon tape and viewed on one end by a photomultiplier tube (CR105) with optical grease. The yield of the photomultiplier was analyzed by a multichannel analyzer (DV4096). The scintillation light decay kinetics was measured using a Photonis XP2254b PMT, which has a multi-alkali photo cathode and a quartz window. In all these measurements one end of the sample was coupled to the PMT with Dow Corning 200 fluid. A ^{137}Cs source was employed to excite the samples.

3. Results and discussion

3.1. Relative light yield and energy resolution

The transmittance spectra of pure and BSO:Dy crystal samples have been measured in the range of 200–900 nm, as shown in Fig. 2. Obvious absorption peaks were observed, which were attributed to transitions of 4f electrons of Dy^{3+}. 4 nm red-shift of the absorption edge in BSO:Dy crystal, as the doping concentration of Dy_2O_3 increased to 4 mol%, is observed (inset of Fig. 2). As the concentration of Dy^{3+} increased, the absorptions became stronger.

Table 1
Scintillation characteristics of BGO and BSO [1].

Crystal	Density (g/cm^3)	Hardness (Mohs)	Radiation length (cm)	Peak emission (nm)	Light yield (relatively)	Decay time (ns)
BGO	7.13	5	1.12	480	100	~300
BSO	6.80	5	1.15	480	20	~100

Anyway, all the samples keep near 80% transmittance in the range from 350 to 900 nm.

Pulse height measurement of BSO and BSO:Dy is performed at room temperature. The samples are optically coupled with Hamamatsu CR105 photomultiplier tube (PMT) and irradiated with 662 keV γ-rays from a ^{137}Cs source. The analog signals from the PMT are shaped by a preamplifier using 2 μs shaping time and analyzed by a pulse height analyzer (PHA) operated in the peak voltage mode. To compare the light output of BSO:Dy with that of BGO, a standard BGO sample was also measured under the same experimental conditions. The pulse height spectrum is shown in Fig. 3. The obtained photo peak of the pulse height spectrum is fitted to a Gaussian (not shown in Fig. 3) to evaluate the peak position and full-width at half maximum (FWHM) to obtain the relative light yield and energy resolution. The relative light yield and FWHM energy resolutions of BSO and BSO:Dy crystals are given in Table 2. As can be seen in Table 1, with small doses of Dy^{3+} doping, the light yield increases dramatically by about 50%. When the doping concentration of Dy_2O_3 is more than 0.1 mol%, the light yield decreases as the doping concentration increased. While the doping concentration is increased more than 1 mol%, the light yield of BSO:Dy crystal degrades as lower than pure BSO crystal. The doping effects on the FWHM energy resolution are similar as that on the light yield.

3.2. Decay time curve

The scintillation decay was measured by a PMT with fast response time and recorded by an Agilent 6052A digital scope using ^{137}Cs excitation. Fig. 4 shows the decay time curve of the BSO single crystal and 0.05 mol% Dy_2O_3 doped BSO crystal excited by 662 keV γ-rays photons from a ^{137}Cs source. As the size of samples is small, the signal from PMT were weak, coupled with some noise, the decay time curve is not very well. The numerical values of their decay time listed in the figure were extracted by an exponential fit. The average decay time of BSO:Dy crystal calculated based on the corresponding decay time curve are summarized in Table 1.

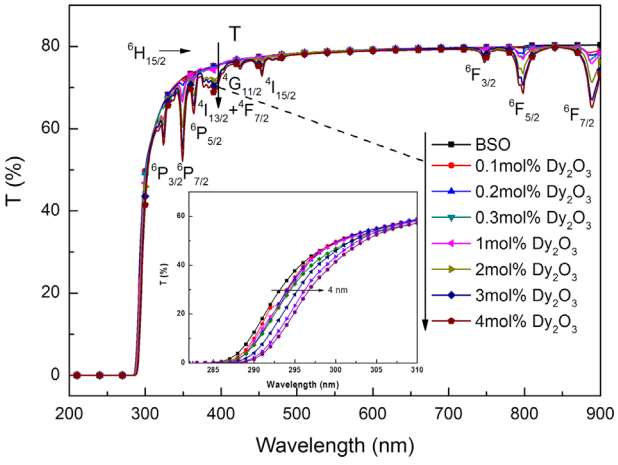

Fig. 2. Transmittance spectra of pure and BSO:Dy crystal samples.

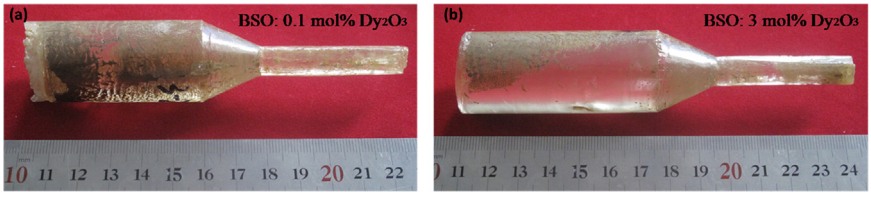

Fig. 1. As grown crystals of BSO:Dy with 0.1 and 3 mol% Dy_2O_3.

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