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Growth and characteristics of LYSO ($Lu_{2(1-x-y)}Y_{2x}SiO_5:Ce_y$) scintillation crystals

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Abstract

In this experiment, properties of LYSO crystals with different yttrium concentrations were investigated by Czochralski method. The concentrations of yttrium and cerium were analyzed by ICP-AES and the distribution coefficients of yttrium and cerium in LYSO crystal were calculated. It was found that the light output and the decay time of LYSO crystals are almost the same as that of cerium-doped lutetium oxyorthosilicate crystal (LSO). The energy resolutions of LYSO crystals, however, fluctuated in a large range.

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1. Introduction

Cerium-doped oxyorthosilicates of gadolinium, yttrium and lutetium, i.e. Gd_2SiO_5:Ce (GSO), Y_2SiO_5:Ce (YSO) and Lu_2SiO_5:Ce (LSO), respectively, have been grown by the Czochralski method and demonstrated as efficient scintillators [1–3]. The advent of above scintillators brought more and more interests in physics and crystal growth. A summary of the relevant properties of these materials along with conventional scintillation crystals such as thallium-doped sodium iodide (NaI(Tl)) and bismuth germanate (BGO) is given in Table 1.

Although LSO exhibits better scintillation properties, its melting point of 2150 °C is very close to the breakdown temperature of iridium crucible and insulating material of ZrO_2. Moreover, the Lutetium oxide is relatively expensive. Compared with LSO, both GSO and YSO have lower melting points and cheaper raw materials. As preferable

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Table 1
Scintillation properties of Ln_2SiO_5:Ce and other typical scintillators [1–6]

Property	Na(Tl)	BGO	GSO	YSO	LSO
Density (g/cm^3)	3.67	7.13	6.71	4.5	7.4
Light output (a.u)	100	15–20	25		75
Decay time (ns)	230	300	60		40
Rugged?	No	Yes	No	Yes	Yes
Melting point (°C)	924	1323	1950	1980	2150

choices, the mixed scintillators such as cerium doped $Lu_{2-x}Gd_xSiO_5$ (LGSO) and cerium doped $Lu_{2-x}Y_xSiO_5$ (LYSO) were grown by researchers [5–9]. LGSO performs worse scintillation properties. However, LYSO presents similar scintillation properties and may be a good alternative to LSO. In addition, the adding of yttrium may have an improved effect on the distribution of cerium and it is necessary to know the distribution coefficient of yttrium in LYSO crystal.

In this paper, LYSO crystals with different yttrium concentrations were grown by Czochralski method and the distribution coefficient of yttrium in LYSO crystal as well as its scintillation properties was investigated.

2. Experiment

2.1. Growth of LYSO crystals

The starting powder materials are Lu_2O_3, Y_2O_3, SiO_2 and CeO_2 of 99.99% purity. All oxides were heated at 200 °C for 10 h to ensure powders free of moisture and CO_2. The powders were weighed in stoichiometry ratio of $Lu_{2(1-x-y)}Y_{2x}SiO_5{:}Ce_y$, mixed and iso-statically pressed into tablets. In the experiments, the content of cerium was always at 0.5% (relative to RE). The doped concentrations of yttrium were selected as at 3.91%, 4.47%, and 10.09%. Then the tablets were sintered at 1500 °C for 6 h in order to obtain compact charge of LYSO.

Crystal growth was performed in the iridium crucible with the diameter of 80 mm. Flowing nitrogen (N_2) was used as protective atmosphere and LSO crystals were used as seed. The typical growth rate was 2 mm/h. The whole growth process was automatically controlled by computer. Fig. 1 shows the photo of the as-grown LYSO crystal boule with the diameter of 30 mm.

Fig. 1. Photo of the as-grown LYSO crystal boule.

2.2. Measurements of distribution coefficient and scintillation properties

Samples were cut from the top, middle and bottom of the above crystal boule for measurement of distribution coefficient. The corresponding position is shown in Fig. 2. Then they were ground into powders. After the powders were dissolved in high temperature alkali solution, the concentrations of yttrium and cerium in LYSO crystals were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

Samples with dimension of $10 \times 10 \times 2\,mm^3$ were cut from the as-grown crystals, and six sides were all optically polished. Test on the scintillation properties of LYSO were performed in Institute of High Energy Physics, Chinese Academy of Sciences. Fig. 3 shows the schematic diagram of the equipment for measurement of light yield and decay time. In this equipment, a calibrated XP2020 photomultiplier (PMT) was used to detect the scintillation light. A conically optical wave guide was put between PMT and LYSO crystal. To achieve good light collection, the wave guide was optically coupled with PMT and crystal by the silicone, and Teflon was used as a light reflector. The radioactive source was the standard ^{22}Na.