Cerium doped Lithium Gadolinium Borate: A neutron Scintillator

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Introduction

The cerium doped lithium gadolinium borate Li₆Gd(BO₃)₃ [LGBO] single crystals are of current interest due to their potential application as scintillators for the neutron detection. The unit cell of LGBO contains a large number of boron atoms (10B isotope) which are useful to capture neutrons that yield energy of approximately 2.8MeV per absorbed neutron through the ¹⁰B (n, a) ⁷Li reaction. In addition, ⁶Li also has a large capture cross section for thermal neutrons through ⁶Li (n, a) ³H reaction [1]. The large band gap of $\sim 8-9$ eV of the host LGBO can incorporate many rare earth ions to alter its luminescent properties for applications including lasers, scintillators etc [1]. Among these Ce doped LGBO (LGBO:Ce) has received more attention as a scintillator for the thermal neutrons due to its short decay time (~27 ns) and high light output yield which is approximately six fold that of 6Li-glass scintillators currently used as neutron detectors [2].

Experiment

Synthesis

Single crystals of 0.1, 0.5 and 1 at% cerium doped LGBO were grown using the Czochralski technique. A typical photograph of as-grown single crystals is shown in Fig. 1. Initial material was prepared by mixing 4N pure (99.99%) constituent oxides (Li₂O₃, Gd₂O₃, CeO₂, B₂O₃) and using the high temperature solid state reaction at 750° C with intermediate mixing. Asprepared material was taken in a platinum crucible and heated to 900°C (about 50°C more

than its melting temperature) to homogenise the melt. Pull rates of 0.5 mm/h and 0.3 mm/h were used respectively for 0.1 % and 0.5%/1% cerium doped crystals. The rotation rate was kept at 10 rpm during the entire growth. The grown crystals were then cooled down to room temperature at 30°C/h to avoid the cracking.



Fig. 1 As-grown single crystal of LGBO:Ce crystals.

Characterization

Radioluminescence spectra were measured by exciting the samples with X-rays (Cu-anode) at an exciting voltage of 40kV and a tube current of 30mA. Emission spectra were recorded from Avantes 3648 spectrometer by using UV600 optical fiber.

Crystals with different Ce concentrations were irradiated using ²⁴³Am alpha source having alfa energy of 5.27 MeV and 5.5 MeV, ²⁵²Cf thermal neutron source and ⁶⁰Co as a gamma source to the scintillation response. The emission

was measured using a PMT and spectra were recorded with the help of preamplifier, spectroscopic amplifier and MCA.

Results and discussion

Interaction of neutrons with LGBO crystals takes place via following reactions: $\underline{\text{With}}^{10}\underline{\text{B}}$

 $\frac{\text{With }^{10}\text{B}}{^{10}\text{B}_5 + \text{n}}$ → $^{7}\text{Li}_3 + \alpha$ (Q value = 2.792 MeV, ground state) $^{10}\text{B}_5 + \text{n}$ → Li₃ (excited) +α + γ (0.48 MeV) (Q value = 2.310 MeV. $\frac{\text{With }^{6}\text{Li}}{^{6}\text{Li}_3 + _{0}\text{n}^{1}}$ → $^{4}\text{He}_2$ (2.05 MeV) + $^{3}\text{T}_1$ (2.75 MeV and $\frac{\text{With }^{157}\text{Gd}}{^{157}\text{Gd}_{64} + _{0}\text{n}^{1}}$ → $^{158}\text{Gd}_{64} + \gamma$ (8 MeV Cascade) + Conversion electrons + X rays (29-182 keV)

The charged particles emitting from the reactions mentioned above consequently excite Ce ions and result into the characteristic Ce³⁺ emission peaking at 420 nm that corresponds to the transition of electrons from 5d to 4f energy levels. The radioluminescence emission spectrum of 0.1% Ce doped crystals is shown in figure 2. It shows the Ce³⁺ emission band at 420 nm besides an emission band in UV (300 nm) and visible regions (above 550 nm) due to Gd³⁺ emissions. The emission band at 420 nm matches well with most of the commercially available PMTs and may be used for the detection of neutrons.

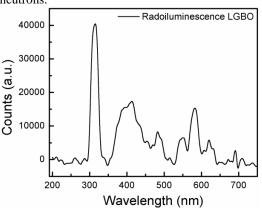


Fig. 2 Radioluminescence spectra of LGBO:0.1%Ce crystals.

Figure 3 shows the response of LGBO crystals having different Ce concentrations with various radiation sources. Channel numbers (X-axis) represent the scintillation light yield of the crystals. Different size and geometry of the crystals cause different counts on the Y-axis. The light yield was observed to increase with an increase in the Ce concentration from 0.1 to 0.5%, but no further improvement was observed for 1% Ce doped crystals. The absolute light yield for 0.5 % Ce doped crystals was measured to be 4500 ph/MeV. The lower value of light output can be assigned to the presence of Ce⁺⁴ ions as the crystals were grown in air environment. Presence of Gd also leads to the shower of lower energy radiations that consequently causes lower light output and poor energy resolution.

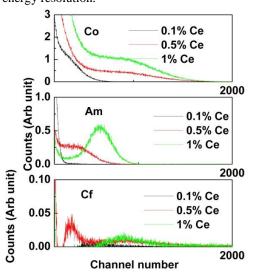


Fig. 3 Response for different radiation sources from LGBO crystals having different Ce concentrations.

Crystal growth in reducing atmosphere with enriched Li and depleted Gd is expected to increase the light output significantly and will be a topic of future investigation.

References

- [1] Ogorodnikov et al., Physics of the Solid State, **53**, 263-270 (2011).
- [2] Hiroshi Nishimura et al., Japanese Journal of Appl Phys, **45**(2A), 909-911, (2006).