Influence of Ga/Al Ratio on Luminescence and Scintillation in Ce^{3+}/Tb^{3+} Co-Doped $Gd_3(Ga,Al)_5O_{12}$ Scintillators

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In recent years, the integration of advanced X-ray imaging methodologies with monochromatic X-ray sources has enabled imaging at submicron to micron scales, thereby garnering significant interest in high-resolution synchrotron radiation X-ray imaging research [1]. Oxide single-crystal scintillators represent a critical class of functional materials distinguished by their high density, large effective atomic number, and superior physical and chemical stability—properties that render them particularly well-suited for X-ray imaging applications [2]. Among these materials, Ce^{3+}-doped $Gd_3(Ga,Al)_5O_{12}$ (GGAG:Ce) exhibits the highest light yield reported to date among oxide-based single-crystal scintillators, positioning it as one of the most promising candidates for X-ray detection [3]. In recent investigations, we have focused on co-doping Ce^{3+} and Tb^{3+} ions within the garnet crystal structure. Co-doped Ce^{3+},Tb^{3+} scintillators have demonstrated enhanced luminescence and scintillation performance attributed to efficient energy transfer between the two ions [4]. Specifically, Ce^{3+} emission, originating from the 5d–4f transition, is highly sensitive to the crystal field due to the spatial extension of the 5d orbital. In contrast, Tb^{3+} emission, governed by 4f–4f transitions, is largely unaffected by crystal field variations due to the localization of 4f electrons near the nucleus. Consequently, while the luminescence characteristics of Ce^{3+} are influenced by the host composition, those of Tb^{3+} remain relatively stable.

In this study, we investigated the bidirectional energy transfer dynamics between Ce^{3+} and Tb^{3+} as a function of Ga-to-Al ratio ($x = 1, 2, 3, 4$) in $Gd_3Ga_xAl_{5-x}O_{12}$. Crystals with the composition $Gd_3Ga_xAl_{5-x}O_{12}$:0.5%Ce, 15%Tb ($x = 1, 2, 3, 4$) were grown using the micro-pulling-down method, and their photoluminescence, photoluminescence excitation, and radioluminescence characteristics were evaluated. The results revealed that the efficiency of bidirectional energy transfer between Ce^{3+} and Tb^{3+} was dependent on the Ga:Al ratio, with the highest total radioluminescence intensity observed at a Ga:Al ratio of 3:2. In addition to a discussion on luminescence behavior, this study also presents the successful growth of large single crystals via the Czochralski method and demonstrates their practical applicability through X-ray imaging experiments.

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