Influence of terbium content on the photoluminescence properties of Ce-doped $Tb_xGd_{3-x}Al_5O_{12}$

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Rare-earth-activated scintillating materials are used in a wide range of applications including white light-emitting diodes, medical imaging devices, and high-energy physics experiments. In high-energy physics applications, scintillators are required to have a high density to ensure strong stopping power and a short scintillation decay time. Rare-earth-activated garnets with a flexible chemical composition are prospective as such scintillators and can be grown by a variety of growth techniques. Cerium-doped yttrium aluminum garnet (YAG:Ce) can be considered a potential candidate, however, electron trapping limits its performance. The influence of electron trapping can be reduced by partially substituting aluminum with gallium ions in the lattice, but this also lowers the energy barrier for thermal depopulation of the emitting Ce level, resulting in scintillation quenching. The depopulation barrier can be increased by replacing yttrium ions with gadolinium, resulting in an increased light yield. However, the introduction of Gd ions into the host lattice slows the scintillation decay due to excitation transfer through the gadolinium sublattice. Recently, fast excitation transfer has been reported in garnet scintillators containing terbium.

In this work, we study the influence of terbium introduction on the emission properties of Ce-doped Ga-free gadolinium aluminum garnet with a fraction of Gd substituted by Tb. We focus on the excitation transfer between the matrix-building ions and the activator ion Ce^{3+}. To investigate the influence of Tb content on the emission intensity and decay rate, we studied a set of three layers of Ce-doped $Tb_xGd_{3-x}Al_5O_{12}$ with Tb content $x = 0.5$, 1.3, and 1.5 grown by the substrate melt-dipping liquid phase epitaxy method. Steady-state and time-resolved photoluminescence and cathodoluminescence spectroscopies were used to study the excitation relaxation following the resonant excitation to the specific energy levels of Ce^{3+}, Tb^{3+}, and Gd^{3+} in photoluminescence experiments, as well as after the generation of high-energy nonequilibrium carriers by accelerated electrons in cathodoluminescence experiments.

The comparison of the photoluminescence decay transients under the excitation of $5d_1$ and $5d_2$ levels of the activator ion Ce^{3+} and the Tb^{3+} $5d_1$ level shows that the introduction of terbium into the lattice accelerates the initial decay of Ce^{3+} luminescence. The photoluminescence decay of Ce^{3+} has three components: fast, intermediate, and slow. The fast component is attributed to the excitation transfer from the activator ion Ce^{3+} to Tb^{3+}, the intermediate component corresponds to the radiative recombination of isolated Ce^{3+} ions, whereas the slow component is associated with the delayed return of trapped carriers to the emitting Ce^{3+} centers. The Tb^{3+} emission is relatively weak and exhibits a fast sub-nanosecond decay component related to cross-relaxation between $^5D_3$ and $^5D_4$ of the Tb^{3+} ion.