Influence of Mg²⁺ codoping on excitation relaxation and emission properties of (Lu,Gd)₃(Ga,Al)₅O₁₂:Ce,Mg scintillators

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Scintillation materials have currently a very wide application field from medical imaging and high energy physics experiments to the homeland security systems and nuclear safety monitoring. The successful growth of Ce-doped gadolinium aluminum gallium garnet (GAGG) single crystals that are prospective for the detection of ionizing radiation due to their high density, high light yield, and the absence of intrinsic radioactivity encouraged an intensive development of mixed garnet-type scintillators for efficient and fast scintillation detectors. The codoping by small amounts of divalent alkali-earth ions was proved to be a successful method to achieve faster scintillation, whereas the influence of codoping on the emission properties at the codoping levels comparable to that of the activator ions still remains a subject for investigation [1].

In this work, we focus on Ce-doped and Mg-codoped $(Lu,Gd)_3(Ga,Al)_5O_{12}$:Ce,Mg (LuGGAG) multicomponent single crystalline epitaxial layers grown using liquid phase epitaxy (LPE) method. LPE technique has an advantage in the preparation of Ce-doped garnets at lower temperatures allowing noticeable reduction of the concentration of antisite and vacancy-related defects acting as effective traps for electrons. We report our results of a study of electronic excitation relaxation and emission properties in a set of Ce-doped LuGAGG films containing different content of Mg^{2+} ions (from 0 to 2200 ppm). The linear and transient absorption, steady-state and time-resolved photoluminescence spectroscopy have been used to study the influence of Mg^{2+} ions on excitation relaxation and emission properties of Ce³⁺ activator ions in the 80-600K temperature range.

The dominant excitation relaxation pathways were examined exploiting selective photoexcitation of the first and second excited levels of Ce^{3+} ions. The study of photoluminescence temperature dependence under selective excitation of the lowest excited Ce^{3+} level 5d₁ revealed emission thermal quenching due to increasing thermal escape of electrons to the conduction band and subsequent nonradiative recombination. The slight increase of photoluminescence intensity from low-to-room temperature after the selective excitation of 5d₂ level indicate the localization of electrons during their transfer to emitting 5d₁ level due to bandgap fluctuations caused by compositional disorder of mixed garnet crystal matrix. The additional decrease of photoluminescence intensity and increased emission decay rate was found in the LuGAGG films containing more than 300 ppm of Mg. The later observation is in line with the formation of nonradiative activator-codopant related excitation relaxation route [2].

[1] D. Zhu, M. Nikl, W. Chewpraditkul, and J. Li, "Development and prospects of garnet ceramic scintillators: A review," *Journal of Advanced Ceramics*, vol. 11, no. 12, pp. 1825–1848, Nov. 2022.

[2] P. Schauer, O. Lalinský, M. Kučera, Z. Lučeničová, and M. Hanuš, "Effect of Mg codoping on cathodoluminescence properties of LuGAGG:Ce single crystalline garnet films," *Optical Materials*, vol. 72, pp. 359–366, Oct. 2017.