Improved scintillation response of Ce doped GGAG garnet films: A pathway to high LY materials

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Recently, a new concept of Ce^{3+} doped multicomponent aluminum garnets, $(GdYLu)_3(GaAl)_5O_{12}$:Ce, has been proposed with partial substitution of Gd and Ga for (Y, Lu) and Al ions, respectively, where detrimental effect of shallow traps is significantly reduced [1]. Due to the combined substitution of Gd and Ga, the shallow traps are extensively suppressed and these systems have excellent LY, two times higher than LuAG:Ce or YAG:Ce.

The aim of this work is detailed study of shallow traps in Ce^{3+} doped multicomponent $(GdLu)_3(GaAl)_5O_{12}$:Ce garnets and their impact on the scintillation properties (scintillation efficiency and timing characteristics) of this material. Due to the technological breakthrough in the liquid phase epitaxy high purity single crystalline films were grown from the lead-free BaO-B_2O_3-BaF_2 flux with special emphasis on elimination of potential impurities coming from the flux. The thickness of films was 10 – 30 μm. Combined experimental study of photoelectron yield (under alpha excitation), decay kinetics of delayed recombination in the millisecond time range (under e-beam excitation), and cathodo- and radio-luminescence spectroscopies were used in the study.

For this research a set of 30 samples GdGa-LuAG:Ce were prepared covering full concentration ranges of Gd and Ga substitutions. In Fig. 1 the photoelectron yield (PhY) of several samples with various compositions are displayed together with a reference $Gd_3Ga_3Al_2O_{12}$:Ce (GGAG:Ce) single crystal which has the LY of 50 000 phot/MeV (under gamma excitation). While the PhY of LuAG:Ce epitaxial film is about half of that value, the PhY of heavily Gd, Ga doped epitaxial films is approaching that of the best bulk crystals measured so far. These films have also excellent timing characteristics. The prompt Ce^{3+} (5d-4f) component in the scintillation decay (under alpha-particle or e-beam excitation) is between 50-70 ns [2] and very weak slow decay components and afterglow were measured, Fig. 2. Namely, the signal decreases in 2 orders of magnitude within 0.5 μs after the 1 ms e-beam excitation and it decreases another order of magnitude within the next 10 μs. These exceptional scintillation properties were observed in samples with Gd and Ga content ≥ 2 atoms/f.u. The results are in excellent agreement with the cathodoluminescence decay kinetics of 5d-4f(Ce^{3+}) emission measured in the nano-microsecond time range [2] where it was shown that trap centers responsible for “slow light” in the scintillation emission are completely suppressed in the heavily Gd, Ga doped garnets. Above mentioned two Gd & Ga atoms/f.u. can be considered as bottom limit – namely, at lower concentrations the slow Gd emission is not completely quenched and Gd → Ce energy transfer [3] results in decreased LY and appearance of very slow components [4], see figures below; and at lower Ga content, the trap states are not completely buried in the conduction band which results in delayed recombination on Ce^{3+} centers.

In conclusion, presented results clearly show that comparable scintillation performance of epitaxial films with that of Czochralski grown bulk crystals can be indeed achieved. It was shown, that the energy transfer processes involving trap states are suppressed in heavily Gd, Ga substituted LuAG:Ce and the dominant scintillation decay is due to the prompt recombination of electrons and holes at the Ce^{3+} emission centers.

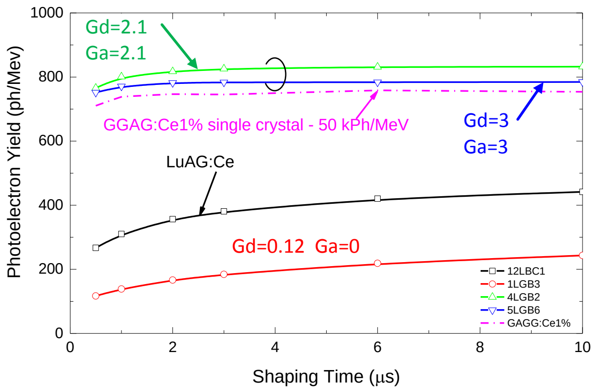
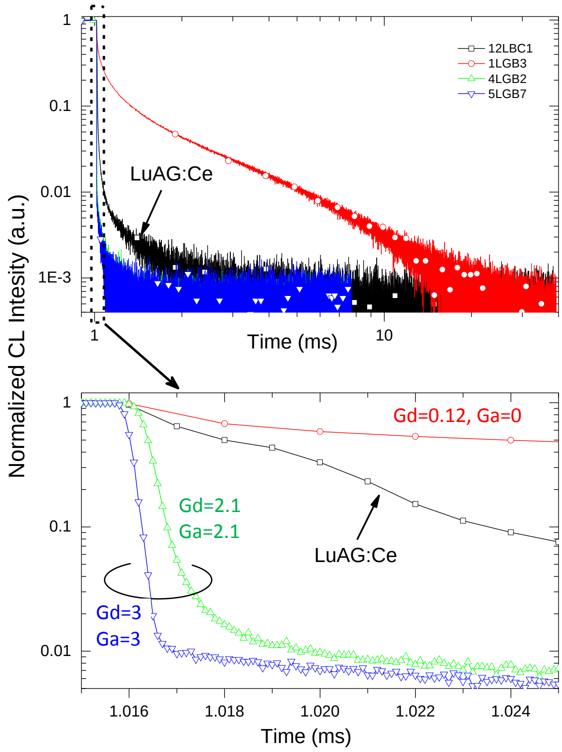


Fig. 1 (above) Photoelectron yield of GdGa-LuAG:Ce under alpha particle excitation.

Fig. 2 (right) Scintillation decay (delayed recombination at λ_{em} = 520 nm) under 1 ms electron beam excitation.



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