Cathodoluminescence decay kinetics of Ce^{3+} doped LuAG:GdGa multicomponent garnets

Z. Onderisinova[1], M. Kucera[1], M. Hanus[1], O. Lalinsky[2], J. Bok[2], P. Schauer[2], M. Nikl[3]

[1]*Charles University, Faculty of Mathematics and Physics, Ke Karlovu 5, 12116 Prague 2, Czech Republic*
[2]*Institute of Scientific Instruments, AS CR, Kralovopolska 147, 61264 Brno, Czech Republic*
[3]*Institute of Physics, AS CR, Cukrovarnicka 10, 16000 Prague 6, Czech Republic*

Ce^{3+} doped single crystal aluminum garnet $Lu_3Al_5O_{12}$ (LuAG) belongs to the group of high figure of merit scintillators. However, the main disadvantage of this material, having a promising light yield of 26 kPh/MeV [1], is presence of slow components in the scintillation response. Undesirable structural defects, identified as antisite defects, are responsible for slow component in scintillation decays and degrade this compound in applications where fast response is required. Therefore, a new material $Gd_3Ga_3Al_2O_{12}$:Ce multicomponent garnet has been recently designed [2] and demonstrated positive influence of Gd^{3+} and Ga^{3+} substitution on the light yield.

In this work we study the scintillation properties of Ce^{3+} doped $(GdLu)_3(AlGa)_5O_{12}$:Ce garnets using electron beam excited decay kinetics. The aim of this research is study of shallow electron traps and their role in the scintillation efficiency of multicomponent garnets. High purity single crystalline films were prepared by liquid phase epitaxy with Gd content in the range from 0 to 3, and Ga content from 0 to 3.5. Scintillation properties of these films are comparable to single crystals having light yield of 50-60 kPh/MeV [3]. The cathodoluminescence (CL) decay kinetics was studied in the temperature range 100 - 480 K under 50 ns electron beam excitation. The CL experiments enable simultaneously detect both the prompt electron-hole 5d-4f recombination of Ce^{3+} and delayed recombination originated from shallow traps.

The CL decay curves of two samples with the different Gd, Ga content are displayed in log-log scale in Fig. 1. The panel (a) shows decay curves which are typical for the LuAG:Ce and GdGaLuAG:Ce samples with lower Gd and Ga content x_{Gd}, x_{Ga} < 2. The decays are evidently composed of two contributions - the fast one which dominates at t < 300 ns and represents fundamental decay of 5d-4f (Ce^{3+}) with τ_{Ce} ~ 50 - 70 ns. The second slow component dominates at t > 300 ns, Fig. 1(a). This slow component can be described by power-law function t^{-p}, where p is a constant. According to Huntley [4] this component can be explain as tunneling of trapped electrons through the potential barrier to the nearby recombination centres. Thus, the entire decay curve is described by the following model, which is combined of exponential and power law functions as we have proposed in [5]:

$$I = I_1 \exp\left(-t / \tau_{Ce}\right) + I_2 \left(t_0 - t\right)^{-p} + I_0, \qquad (1)$$

where τ_{Ce} represents intrinsic Ce decay time, t_0 is included for nonzero time of the excitation pulse, parameter p is a constant which is related to the material, and I_0 is a background signal. The fit using eq. (1) provides $\tau_{Ce} \approx 50$ ns and $p \approx 0.87$.

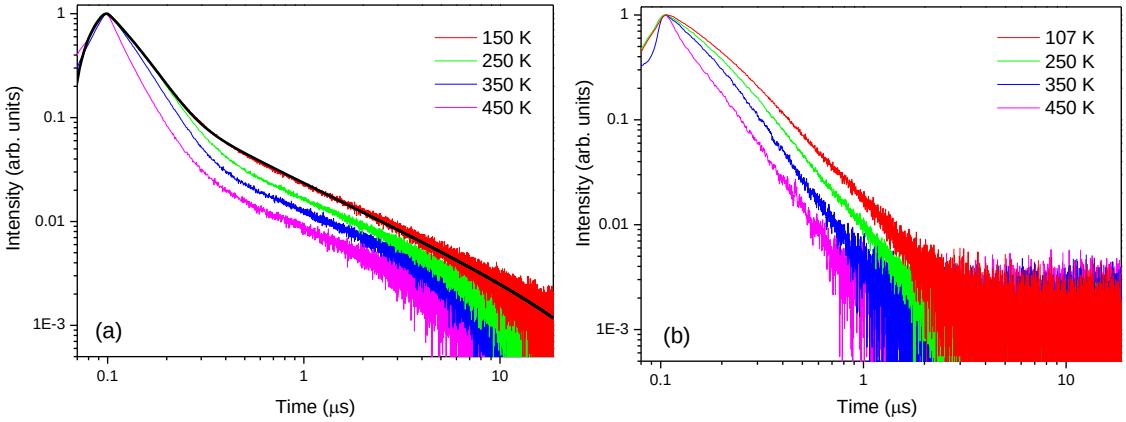

Fig. 1. Electron beam exited scintillation decays of Ce^{3+} doped $(Ce_xGd_yLu_{3-x-y})(Ga_zAl_{5-z})O_{12}$ measured at λ_{em} = 520 nm under 50 ns excitation pulse at various temperatures. Concentrations in samples are (a) x = 0.02, y = 1.17, z = 1.94 and (b) x = 0.02, y = 2.08, z = 2.10. The solid line in (a) represents convolution of decay function eq. (1) with the excitation pulse.

In LuAG:Ce and in samples with low Gd, Ga content < 1 (figures not shown here) these values depend only little on composition as well as on temperature and any thermal ionization was not observed up to 480 K. However, in LuAG:Ce the fast 5d-4f emission carries only ~ 30 % of energy while rest is emitted as slow light as assessed by integration of decay curves, cf. [5]. At higher content the intensity of the slow component, described by a power-law function, gradually decreases and at x_{Gd}, x_{Ga} > 2 it completely disappears and slow light is practically suppressed in emission, Fig. 1(b). This indicates that shallow traps are buried into the conduction band. These results are supported by the CL decays measured in the millisecond time range (afterglow) [3].

In conclusion, we have observed that e-beam excited scintillation decays can be decomposed into 2 components - a fast exponential term with the decay time 50 – 70 ns and a slow power-law term $\propto t^{-p}$. Convenient combination of Gd, Ga dopants leads to removing of shallow traps which are responsible for tunneling effect and delayed recombination. The best results were obtained for samples with x_{Gd}, x_{Ga} > 2. The CL decays do not exhibit any significant thermal ionization of Ce^{3+} excites states up to RT even at high Gd, Ga doping.

[1] A.G. Petrosyan, K.L. Ovanesyan, R.V. Sargsyan, et al., J. Crystal Growth, **312**, 3136 (2010).
[2] K. Kamada, T. Endo, K. Tsutumi, et al., Crystal Growth & Design, **11**, 4484 (2011).
[3] M. Kucera, Z. Onderisinova, M. Hanus, et al., submitted to SCINT 2015.
[4] D.J. Huntley, Journal of Physics: Condensed Matter, **18**, 1359 (2006).
[5] M. Kucera, Z. Onderisinova, J. Bok, et al., J. Lumin., doi:10.1016/j.jlumin.2015.01.034 (2015).