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Trapping and self-trapping in ytterbium-doped oxides with charge transfer luminescence

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ABSTRACT

Temperature dependence of the charge transfer luminescence (CTL) of Yb-doped yttrium aluminum garnet $Y_3Al_5O_{12}$-Yb (YAG–Yb) and Yb-doped lutetium aluminum perovskite $LuAlO_3$-Yb (LuAP–Yb) crystals under X-ray excitation and their thermostimulated luminescence are investigated in the temperature range 30–350 K and compared to those of undoped crystals. Simulation using a set of kinetic equations describing the processes of creation of excitons, electron–hole pairs, their trapping and self-trapping, radiative relaxation and quenching is presented for the systems under investigation to analyze qualitatively two different types of experimentally observed temperature dependences: CTL yield decline with the temperature decrease below 110 K as in case of YAG–Yb and constant yield in the same temperature range as in case of LuAP–Yb.

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1. Introduction

Charge transfer luminescence (CTL) of ytterbium-doped compounds for the first time was identified in Ref. [1] and shown to be manifested in a large series of matrices in Ref. [2]. It receives further attention as a relatively unexplored variety of rare-earth luminescence as well as a potential basis for the creation of a new class of scintillating materials. For scintillator performance efficient energy transfer from the matrix to luminescence centers is of key importance. CTL temperature dependence studied earlier under the excitation in the charge transfer absorption band [3] here it is extended to the X-ray excitation. The crystals of Yb-doped yttrium aluminum garnet $Y_3Al_5O_{12}$–Yb (YAG–Yb) and Yb-doped lutetium aluminum perovskite $LuAlO_3$–Yb (LuAP–Yb), the most elaborate crystals (the former as a laser material and the latter as a scintillator doped by Ce), manifest two different types of the CTL temperature dependence at high-energy excitation: in YAG–Yb with temperature decrease below ~110 K the CTL yield drops substantially, while in LuAP–Yb it stays at a constant level. Since both materials demonstrate substantial thermostimulated luminescence, the processes of trapping and self-trapping of electronic excitations (EEs) need to be taken into account. To investigate the role of intrinsic luminescence of the matrices in the energy transfer processes similar measurements were performed for undoped YAG and LuAP crystals.

2. Experimental

YAG–Yb and undoped YAG as well as LuAP–Yb and undoped LuAP crystals were grown in the Laboratory of Crystal Growth of the Institute for Physical Research, Armenia by Bridgman process, transparent optical ceramics YAG–Yb was grown by Konoshima Chemicals Co., Ltd. The measurements under X-ray excitation were performed in the LPCML using the X-ray source XRG3000 INEL with tungsten anode operating at 35 kV. The luminescence from the illuminated face of the crystal was collected by a PMT through an interference filter, density filter or without any filter in case of X-ray luminescence temperature dependence and thermostimulated luminescence (TSL) measurements, or guided by an UV optical fiber coupled to the Jobin-Yvon Triax 320 monochromator and detected by a CCD camera for X-ray luminescence spectra measurements. The samples were mounted on a cold finger of a closed cycle helium cryostat. During TSL measurements, the rate of heating was 10 K/min. The flux of X-ray photons of ~24 keV on the sample was about 6.5×10^7 photons $mm^{-2} s^{-1}$. Taking into account the attenuation length of LuAP ($\rho = 8.34\,g/cm^3$), which equals to ~45 μm, the density of absorbed photons is $\sim 1.4 \times 10^{12}$ $cm^{-3} s^{-1}$; $3E_g$ for this material being ~24 eV, allows each photon to create $\sim 10^3$ EEs, thus their density can be evaluated as 1.4×10^{15} $cm^{-3} s^{-1}$.

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Measurements in the UV–VUV range have been done using synchrotron radiation at the Superlumi station of HASYLAB, DESY [4].

3. Results and discussion

In Fig. 1, a typical temperature dependence of the CTL of YAG–Yb crystals excited by X-rays is presented. On high temperature side of the curve a Mott-type quenching is observed. At the low temperature side the CTL yield also drops almost to the level observed at RT (more than an order of magnitude relative to the maximum corresponding to 110–120 K). A similar tendency was reported in Ref. [5], trapping of electronic excitations evidenced by the TSL glow curves was proposed as a competitive relaxation channel. In Fig. 1, the curves for single crystals are compared to ceramics. Different geometry of the samples (ceramics was substantially thinner and smaller) does not allow to compare the absolute yields either of the CTL or of TSL, however the ratio of the X-ray luminescence intensity to the TSL intensity was of the order of 10 for single crystals and of the order of 100 for the ceramics sample. Nonetheless in all three samples we observe a similar decrease of the CTL yield at low temperatures. Thus, not only traps cause the decline of the CTL yield at low temperature. Analogous behavior is observed for the CTL of $Lu_3Al_5O_{12}$–Yb, in case of $YAlO_3$–Yb the yield drops with the temperature decrease but to somewhat higher level.

Recently it was shown that LuAP–Yb crystals demonstrate efficient CTL (to be published elsewhere, preliminary results presented in Ref. [6]), its yield under X-ray excitation being about 60% of that of BGO. Temperature dependence of X-ray excited CTL is shown in Fig. 2 together with respective TSL glow curves for Yb concentrations of 2%, 5% 10%, and 50%. There is no decrease of the X-ray luminescence yield in the low-temperature region for Yb concentration up to 50%, the main glow peaks are located at temperatures above 130 K. The peaks at 174 K and 222 K, well-known from the study of LuAP–Ce scintillator ([7] and references therein), are most pronounced for LuAP–Yb 2% sample. With the increase of Yb concentration, their intensity drops as well as that of the peak at 138 K. This fact might indicate that these are electronic traps competing for the electrons with Yb^{3+}-centers. With larger concentration of the latter, which consecutively capture first an electron and then a hole, we have a smaller number of electrons left for other traps. The yield of the CTL is inversely correlated with the TSL yield, thus in this compound, as well as in YAG–Yb, we have a competition between trapping of free carriers and their energy transfer to the CTL centers. However, different conditions for such competition (note log scale of y-axis in TSL of Fig. 2) do not manifest themselves as substantial changes of the profile of the X-ray excited CTL temperature dependence, affecting mainly its overall yield.

Undoped crystals of YAG and LuAP were studied to investigate the role of intrinsic excitations in the energy transfer to the CTL centers.

In Figs. 3a and 4a luminescence spectra of undoped YAG and LuAP are presented together with the excitation spectra of the CTL of respective compounds. In both compounds at low temperature ($T<150$ K), intrinsic luminescence consists of two bands peaking

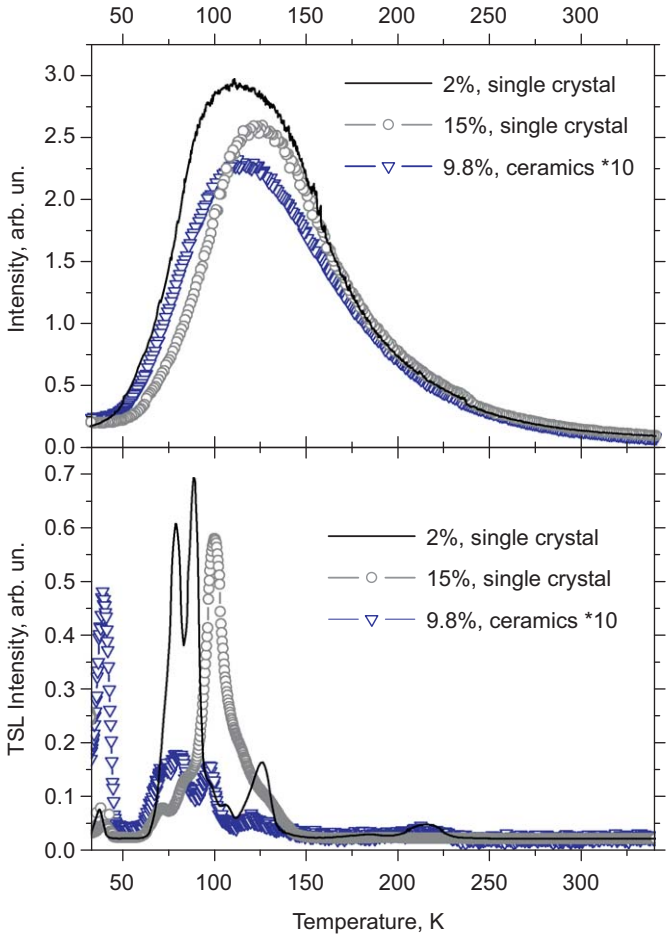

Fig. 1. YAG–Yb, upper panel: temperature dependence of X-ray luminescence; bottom panel: TSL glow curves.

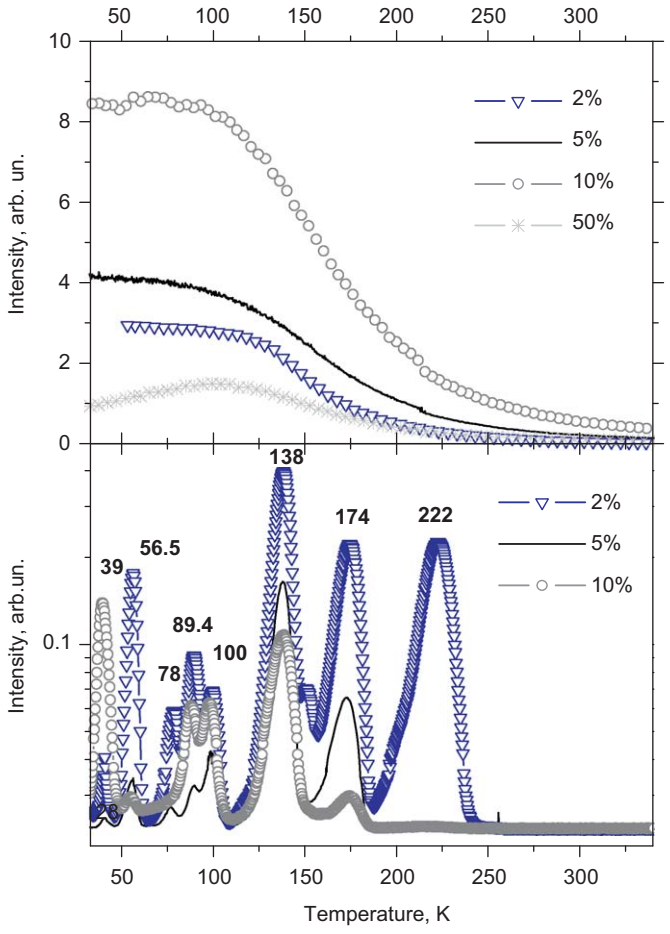

Fig. 2. LuAP–Yb, upper panel: temperature dependence of X-ray luminescence; bottom panel: TSL glow curves.

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