Scintillators based on the Ce^{3+} Doped of Gd$_{1-x}$Y$_x$AP:Ce Mixed Perovskites: Comparative Study Crystals and Single Crystalline Films under Synchrotron Radiation Excitation

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Solid solutions of Ce^{3+}-doped mixed perovskites Gd$_{1-x}$Y$_x$AP:Ce (x=0-1) have attracted considerable attention as promising materials for scintillation applications due to their tenable structural and luminescent properties [1]. In this work, a comparative study of these perovskites prepared in the form of single crystals (SC) and single-crystalline films (SCF) with varying Gd/Y content x is presented, focusing on their luminescent behaviour under synchrotron radiation (SR) excitation. The SCs were grown from the melt using the micro-pulling-down (MPD) method, whereas the SCFs were crystalized by liquid-phase epitaxy (LPE) from BaO-based (YAP) and PbO-based (GdAP) fluxes on YAP substrates.

The SCs and SCFs under study were initially characterized using conventional absorption, cathodo- and photoluminescence spectroscopy, as well as scintillation light yield and decay kinetics measurements under α-particle excitation. Further investigations were performed at 7–300 K using SR excitation with energies in the 3.7–25 eV range at the VUV spectroscopy station located at the P66 beamline of the PETRA III storage ring at DESY (Hamburg, Germany).

These experiments were employed to investigate the influence of Gd^{3+}/Y^{3+} substitution on the local environment of Ce^{3+} ions, energy transfer from the host to the activator, and the peculiarities of the Ce^{3+} luminescence response in SCs and SCFs. Changes in lattice parameters and crystal-field strength induced by Y/Gd compositional variation strongly correlated with modifications in the Ce^{3+} 5d–4f emission characteristics, including the spectral position and intensity of Ce^{3+} emission bands. Namely, the energy required for the creation of excitons bound to Ce^{3+} ions (BSE(Ce)), which represent the final stage of the scintillation process, decreases from 7.58 eV in YAP:Ce to 7.04 eV in GdAP:Ce SCs (Fig.1a).

Other important peculiarities of Ce^{3+} luminescence include significant differences in the positions of BSE(Ce) peaks in YAP:Ce and GdAP:Ce SCFs (7.9 and 7.04 eV) and bulk crystals (7.58 and 7.11 eV) (Fig.1a and 1b, respectively). This feature can be attributed to substantial differences in the concentration of Y$_{Al}$ and Gd$_{Al}$ antisite defects (AD) in SCs and SCFs arising from their high- and low-temperature growth using the MPD and LPE methods, respectively. As a consequence, the formation of Ce^{3+}–AD centers is more probable in bulk crystals, leading to lower-energy BSE(Ce) states compared to those predominantly associated with isolated Ce^{3+} centers in SCF scintillators, where the concentration of AD-related centers is significantly reduced.

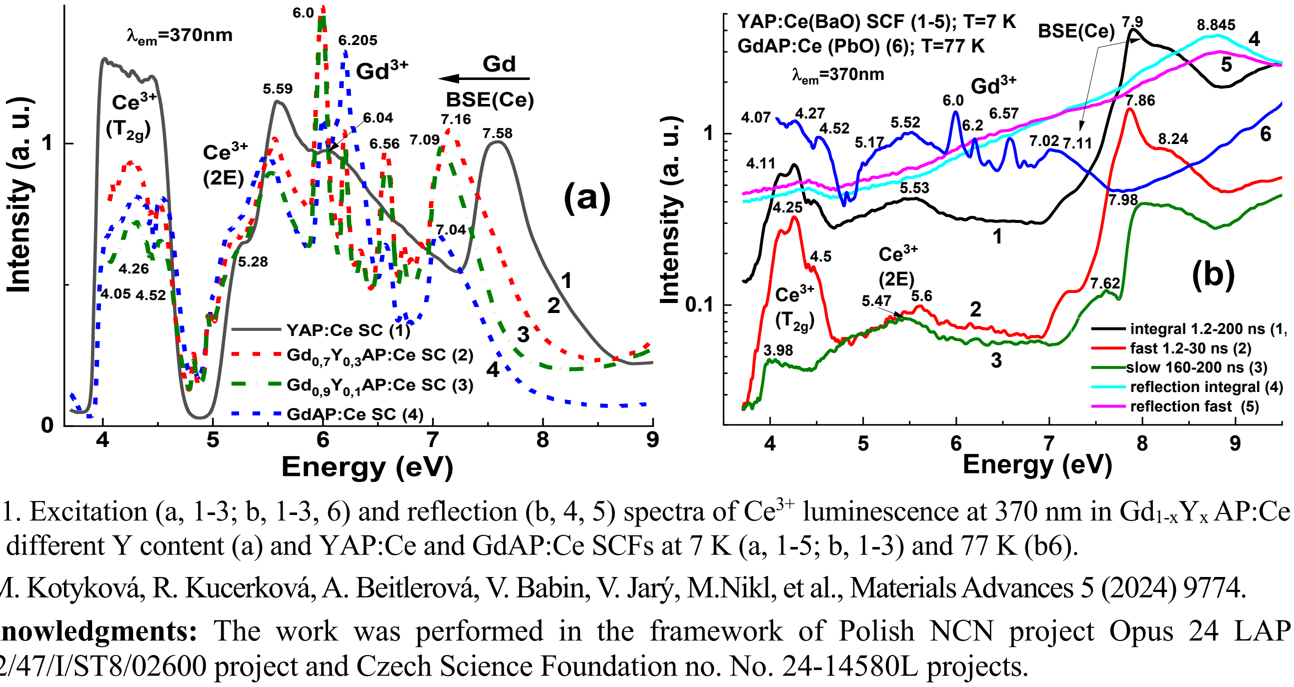

Fig. 1. Excitation (a, 1-3; b, 1-3, 6) and reflection (b, 4, 5) spectra of Ce^{3+} luminescence at 370 nm in Gd$_{1-x}$Y$_x$ AP:Ce SCs with different Y content (a) and YAP:Ce and GdAP:Ce SCFs at 7 K (a, 1-5; b, 1-3) and 77 K (b6).

[1] M. Kotyková, R. Kucerková, A. Beitlerová, V. Babin, V. Jarý, M.Nikl, et al., Materials Advances 5 (2024) 9774.

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