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Luminescence properties of solid solutions Lu_xY_{1-x}PO₄:Eu³⁺

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1. Introduction

Phosphates with tetragonal structure possess not only excellent luminescence properties, but also a high chemical, thermal, and radiation stability [1–9], that allows to use them in scintillation detectors - LnPO₄:Re³⁺ (Ln = Y, Lu; Re= Nd³⁺, Yb³⁺) [10–12], in radiography [13], in plasma panels - YPO₄:Tb³⁺ [14] and in luminescent lamps - LaPO₄:Ce³⁺ [15]. Synthesis of solid solutions is one of the possible ways to tailor luminescent and scintillation properties of compounds. For example, the suppression of the detrimental effect of defects on the luminescent properties by the bandgap modification in solid solutions has been shown for some garnets [16,17]. Another effect is the increase of light output in solid solutions in comparison to their constituents [18–25]. The effect is ascribed to the limitation of the mean distance between the thermalized geminate electrons and holes due to random cation distribution. As a result, the enhancement of the efficiency of excitation energy transfer to the emission centers occurs in case of consecutive capture of charge carriers by the luminescence center.

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ABSTRACT

Solid solutions $Lu_xY_{1-x}PO_4$ doped with Eu^{3+} were synthesized by the sol-gel method. Luminescence spectra under UV and X-ray excitation and luminescence excitation spectra in the UV-VUV energy range are presented. Observed variations of the structure of 4f-4f Eu^{3+} lines with the composition of the solid solution are explained by the fluctuations in the distribution of substitutional cations. The role of electronic structure modification in the fundamental absorption region with changing Y/Lu ratio is discussed and is suggested as the origin of different efficiency of energy transfer to Eu^{3+} in this region. Temperature dependence of the luminescence yield is related to the competition between emission centers and traps. © 2017 Elsevier B.V. All rights reserved.

However, there are several effects, which influence the processes of excitation energy transfer from the host to the activator, resulting in a complicated dependence of the light output on the relative concentration of the substitutional cations [24]. One of them is the modification of the band structure. The band structure modification with the relative concentration of substitutional cations was shown for the Lu_xY_{1-x}PO₄ solid solutions in our previous study [26]. In particular, it is manifested in the increase of the bandgap, modification of the lower part of the conduction band, formed by the states of substitutable cations and appearance of 4f Lu states in the upper part of the valence band with the increase of **x**.

The aim of this work is to determine the factors, which affect the energy transfer from the host to the luminescence centers in $Lu_xY_{1-x}PO_4:Eu^{3+}$ solid solutions.

2. Experimental

The Lu_xY_{1-x}PO₄ solid solutions doped with 0.5 mol % Eu³⁺ ($\mathbf{x} = 0$, 0.1, 0.3, 0.5, 0.7, 0.9, 1.0), were synthesized by the sol-gel method. In all compounds \mathbf{x} value is the relative concentration of Lu cations, $\mathbf{1}-\mathbf{x}$ value is the relative concentration of Y cations. According to the granulometric analysis, predominant size of the obtained







crystalline particles was about 350–600 nm. To monitor completeness of the reaction and phase purity of the final product, the X-ray diffraction (XRD) analysis of the phosphate solid solutions was performed using a Rigaku Ultima IV X-ray diffractometer. Fig. 1 shows the XRD patterns of the Lu_xY_{1-x}PO₄ 0.5 mol % Eu³⁺. The XRD analysis revealed that the prepared samples are homogenous and crystallize in the tetragonal system, I4₁/amd space group, the unit cell parameters follow Vegard's law.

The excitation spectra of phosphates under excitation in the ultraviolet-vacuum ultraviolet (UV-VUV) spectral range were measured at the SUPERLUMI experimental station using synchrotron radiation (DESY, Hamburg, Germany) [27]. The samples were placed in a helium flow optical cryostat providing measurements over a wide temperature range from 4.2 to 400 K. The normal incidence McPherson primary monochromator with Al grating (1200 grooves/mm) allows to vary the energy of incident radiation in the region 3.7-22 eV. The luminescence has been registered using a secondary monochromator SpectraPro300i equipped with a Hamamatsu PMT R6358P. The luminescence and luminescence excitation spectra in the UV and visible ranges were measured using experimental arrangement based on the LOT-Oriel MS-257 spectrograph. This setup allows to carry out the measurements of luminescence spectra in the range of 200-1050 nm and luminescence excitation spectra in the range of 200-500 nm. The measurements of the luminescence spectra and thermally stimulated luminescence (TSL) curves were also carried out under irradiation with X-rays generated by tube INEL XRG 3000 with a tungsten anode operating at 30 keV and 20 mA. The spectra were registered using a Shamrock 500i spectrograph equipped with Newton EMCCD DU970P. The luminescence spectra under UV and X-ray irradiation were recorded in the temperature range 80-500 K. The temperature dependences of emission intensity were obtained from the measured luminescence spectra.

3. Results and discussion

The luminescence spectra of Lu_xY_{1-x}PO₄:Eu³⁺ solid solutions are presented in Fig. 2. Sharp lines in the range 580–750 nm attributed to the ${}^{5}D_{0} - {}^{7}F_{1-4}$ transitions within 4f shell of the Eu³⁺ ions demonstrate different behavior with changes in **x**. Each of the

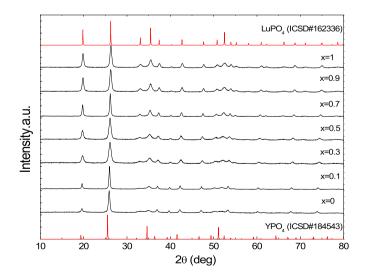


Fig. 1. XRD patterns of Lu_xY_{1-x}PO₄:Eu³⁺. The red line spectra represent literature data for LuPO₄ according to ICSD#162336 and YPO₄ according to ICSD#184543. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

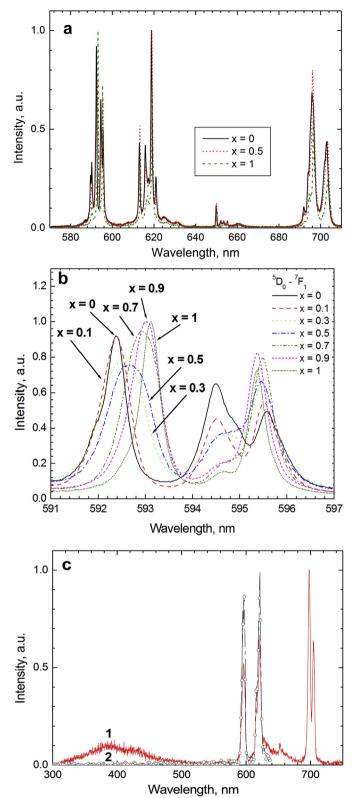


Fig. 2. (a) The luminescence spectra of YPO₄:Eu³⁺ (line), Lu_{0.5}Y_{0.5}PO₄:Eu³⁺ (dashed line), LuPO₄:Eu³⁺ (dots) at $\lambda_{ex} = 395$ nm, T = 300 K. (b) Modification of the emission lines at 591–597 nm with the change of relative concentration of substitutable cations, T = 300 K. (c) The luminescence spectra of Lu_{0.5}Y_{0.5}PO₄:Eu³⁺ under X-ray excitation at T = 300 K (1) and in TSL peak at T = 220 K (2).

transitions ${}^5D_0 - {}^7F_{1,2,3,4}$ is manifested as a series of lines due to low (tetragonal) symmetry of the lattice. With the substitution of Y^{3+} by

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