

LUMINESCENCE OF $\text{LiLa}_9(\text{SiO}_4)_6\text{O}_2$ SILICATE CRYSTALS DOPED WITH Ce^{3+} and Pr^{3+} IONS

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Crystals doped with rare earth ions find numerous applications as scintillators in various fields of technology including nuclear physics, medical imaging, dosimetry, etc. In this paper, we report on luminescence and optical properties of a new class of $\text{LiLa}_9(\text{SiO}_4)_6\text{O}_2$ (LLSO) crystals doped with Ce^{3+} and Pr^{3+} ions which we found potentially attractive for scintillator applications.

The LLSO crystals belong to the nesosilicate family and are characterized by a partially disordered apatite structure that can be doped with optically active rare-earth ions substituting for two La^{3+} sites with different oxygen coordination and symmetry. The crystals for this study were synthesized and XRD verified in the Luminescent Materials Laboratory, University of Verona (Italy).

The spectroscopic study included measurements of photoluminescence (PL) and X-ray excited luminescence (XRL) spectra in the 1.5-5.5 eV energy range as well as UV-VUV excitation spectra in the 3.5-10 eV range at temperatures of 20, 90 and 300 K. Luminescence decay kinetics were recorded upon cathode and UV-pulse laser excitation. In addition, thermally stimulated luminescence of the crystals was studied by recording glow curves in the temperature range from 90 to 500 K.

PL spectra of $\text{LLSO}:\text{Ce}^{3+}$ (1%) recorded upon UV-, VUV-, X-ray and e-beam excitation are composed of two emission bands centered near 2.4 and 2.94 eV with different Stokes shifts that correspond to interconfigurational $5d \rightarrow 4f$ transitions in Ce^{3+} ions substituting for La^{3+} ions in two cationic sites with different coordination numbers. The Ce^{3+} sites demonstrate different thermal quenching that results in significant temperature dependence of PL quantum yield and shape of the PL spectra. The decay time of $5d \rightarrow 4f$ transitions is 19 ns under UV laser excitation and $\tau \approx 70$ ns under e-beam excitation at $T=295$ K. The latter is explained by delayed host-to-impurity energy transfer.

Emission of $\text{LLSO}:\text{Pr}^{3+}$ is presented by intraconfigurational $4f \rightarrow 4f$ transitions in the visible region and interconfigurational $4d \rightarrow 5f$ transitions in the UV region (two broad bands) observed at 90 and 295 K. Positions of $4f \rightarrow 4f$ bands in PL spectra are independent on excitation energy, while features related to $5d \rightarrow 4f$ emission are dependent on excitation energy. As in the case of the Ce^{3+} crystal, this is explained by the existence of two Pr^{3+} sites with different symmetry. Two types of $4d \rightarrow 5f$ emission are observed at $T=295$ K: type I is excited near 5.25 eV yielding a doublet emission feature peaked at 4.0 and 4.45 eV, while type II is excited near 5.0 eV PL yielding similar emission with maxima at 3.55 and 3.85 eV. The decay time of $5d \rightarrow 4f$ emission is about 10 ns upon UV laser excitation in the region of $4f \rightarrow 5d$ absorption at room temperature. It is worth noting that $\text{LLSO}:\text{Pr}^{3+}$ excitation and emission spectra do not show any features which could indicate population of Pr^{3+} $1S_0$ level and enhancement of the $4f \rightarrow 4f$ transitions. This suggests that the $1S_0$ level is above the lowest $4f$ - $15d$ state that prevents observation of cascade photon emission in the sample.

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