

Confocal microscopy of luminescence inhomogeneity in LGSO:Ce scintillator crystal

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Introduction

The yield of scintillation crystals $\text{Lu}_x\text{Gd}_{1-x}\text{SiO}_5:\text{Ce}$ (LGSO:Ce) at the proper choice of Lu/Gd ratio reaches 140 % and 300 % of that in LSO:Ce and GSO:Ce, respectively [1]. Composition dependence of light yield peaked at $x \approx 0.5$ has been observed in Ce-doped $\text{Lu}_{1-x}\text{Y}_x\text{AlO}_3:\text{Ce}$ (LuAP) [2], $\text{Lu}_{1-x}\text{Sc}_x\text{BO}_3:\text{Ce}$ [3], $\text{Y}_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}$ [4], and some other systems. The improvement of light yield might be caused by formation of nanosized regions enriched with substituting cations and, consequently, formation of potential barriers limiting diffusion of thermalized uncorrelated carriers and promoting their capture at Ce^{3+} and subsequent 5d-4f radiative transitions.

In this report, formation of such regions is studied in LSO:Ce and LGSO:Ce. It is known that LGSO:Ce luminescence spectra consists of two subbands peaked at ~420 and ~510 nm and attributed to 5d-4f transitions in Ce1 (CeO7) and Ce2 (CeO6) centres (Fig. 1) [1]. The correlation between Gd content and Ce1/Ce2 bands intensity is shown in Fig. 2. Therefore local Lu/Gd distribution can be evaluated using features of luminescence spectra.

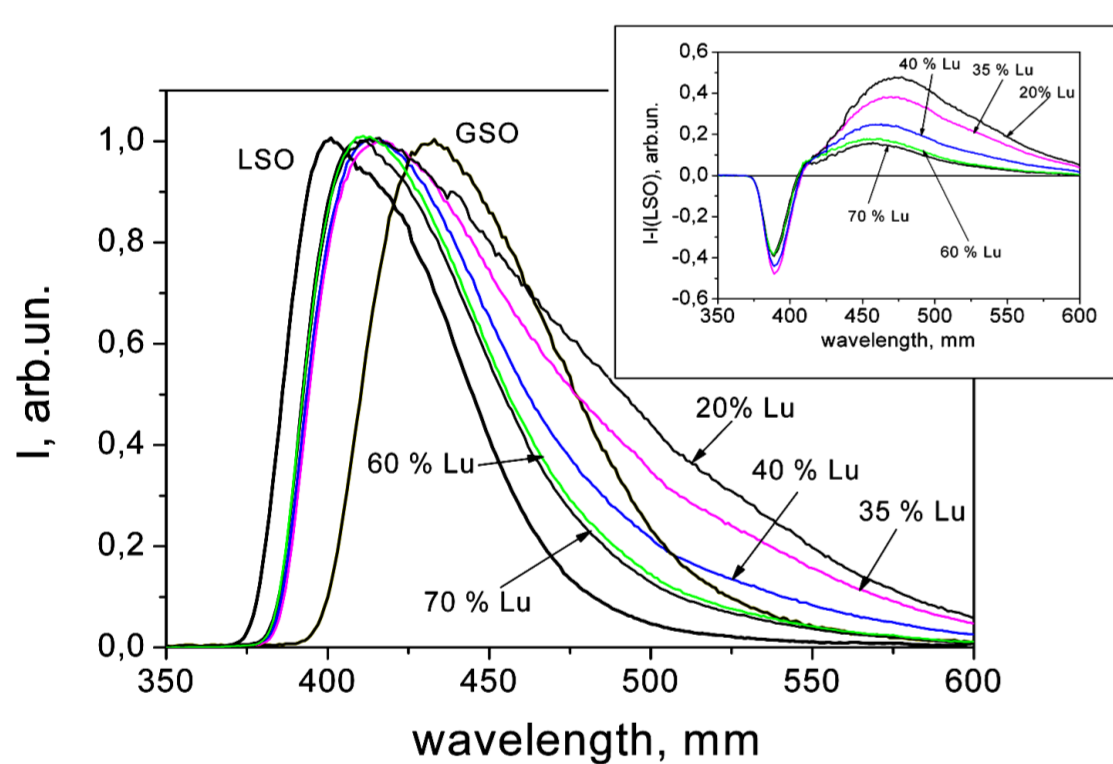


Fig. 1. X-ray luminescence spectra of LGSO:Ce with different Ce content (difference spectra on the inset)

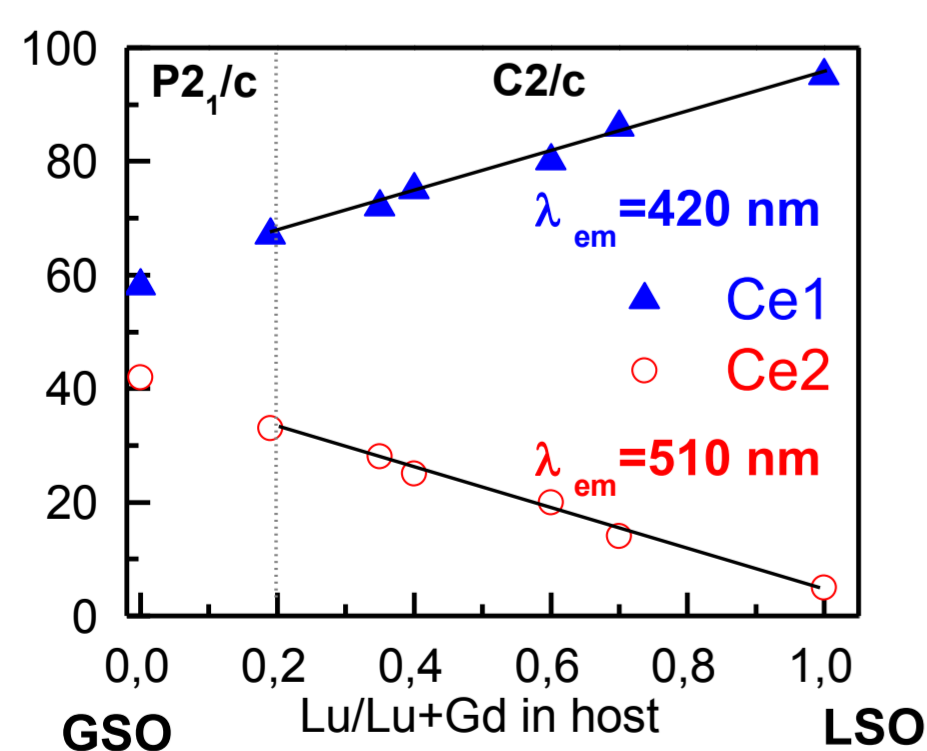


Fig. 2. Contribution of Ce1 (CeO7) and Ce2 (CeO6) luminescence bands into overall luminescence as a function of LGSO composition

Experimental and Results

The spatial distribution of PL parameters was measured using confocal microscopy. CW laser diode emitting at 405 nm was used for excitation. The objective with $NA = 0.55$ ensured the spatial resolution of 400 nm in X-Y plane and 1600 nm in Z direction perpendicular to the crystal surface. The spatial distributions of luminescence parameters were recorded across LSO:Ce and LGSO:Ce crystals grown by Cz method in the same conditions.

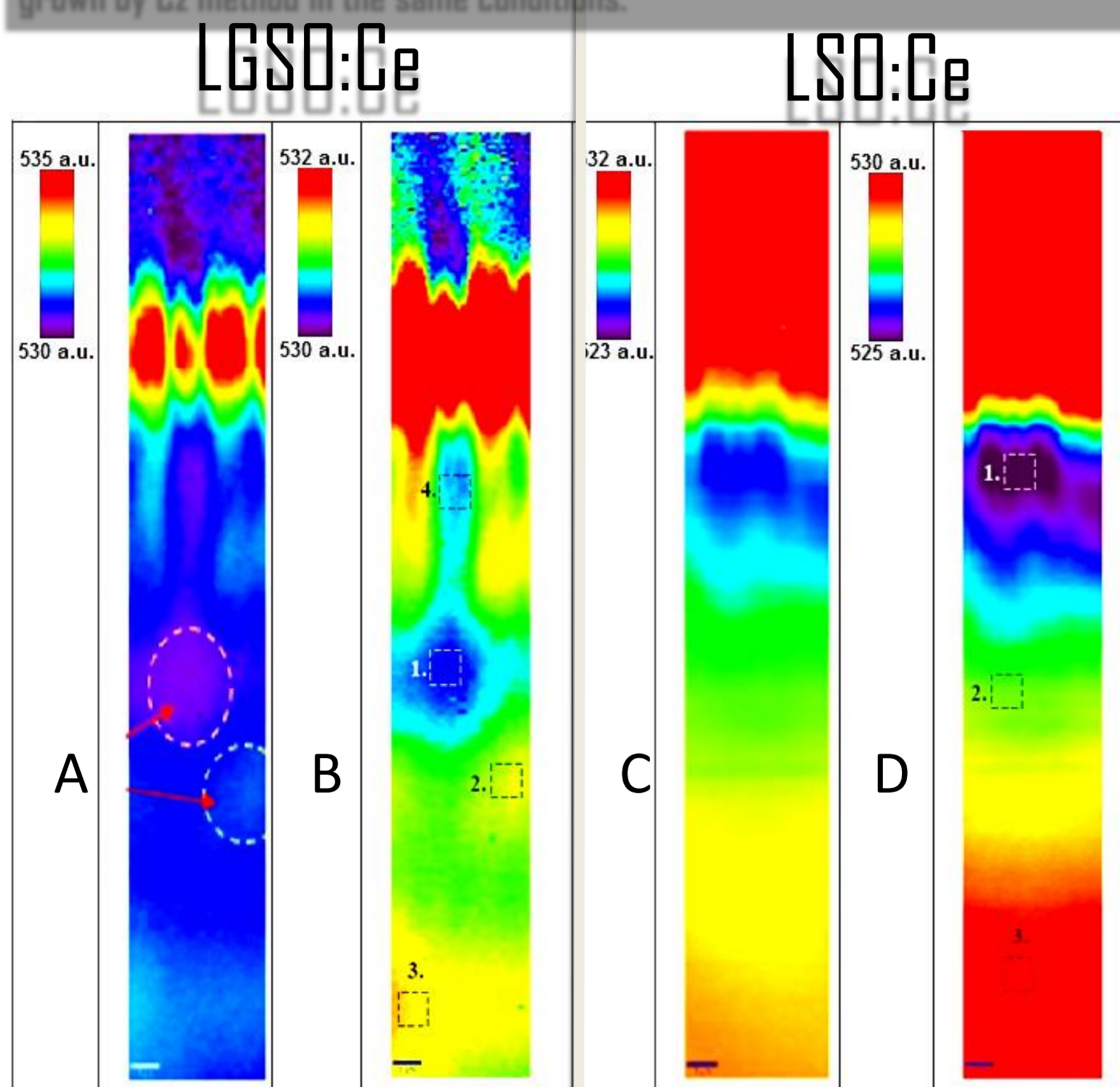


Fig. 3. Spatial distribution of PL band center of mass in LGSO:Ce (A, B), LSO:Ce (C, D) samples.

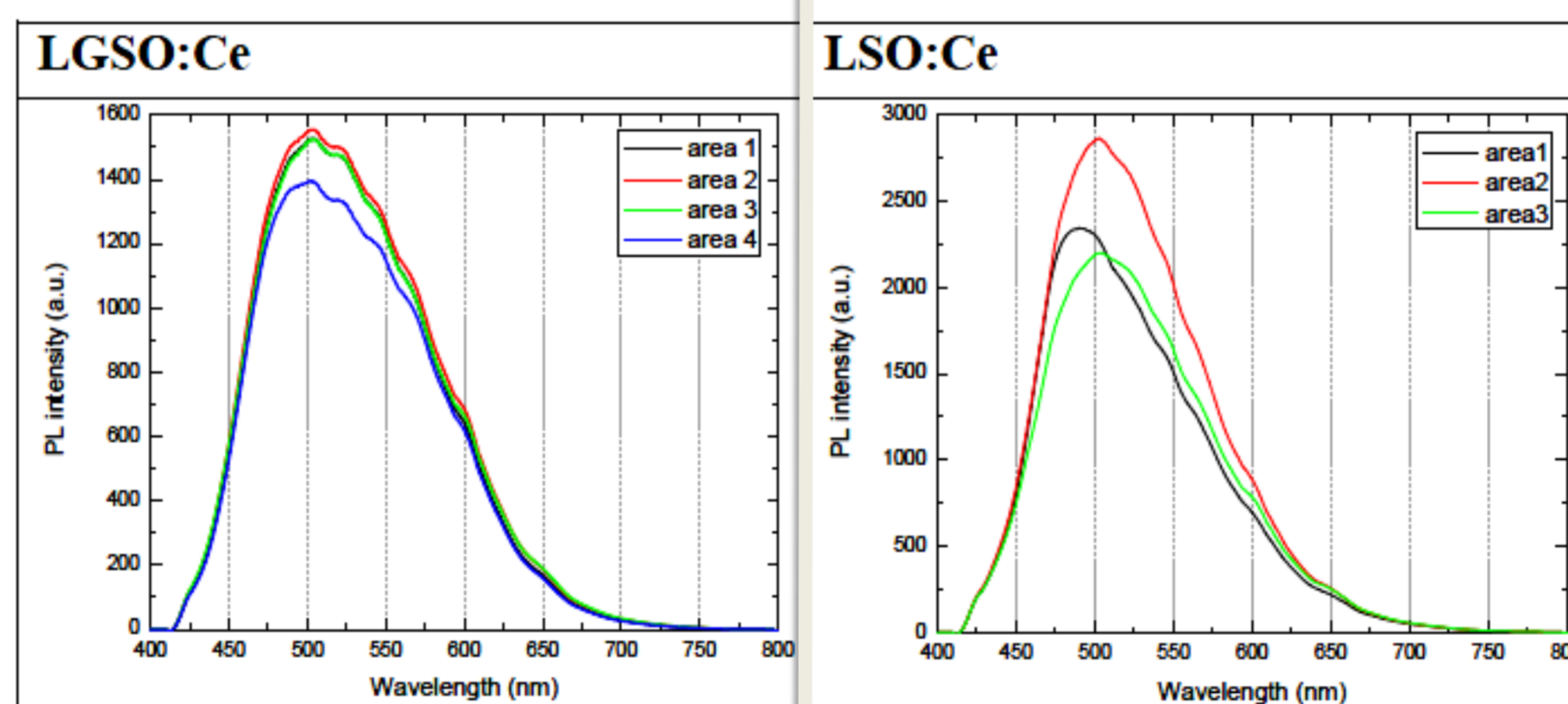


Fig. 4. Spatially averaged PL spectra from different areas in LGSO:Ce (left), LSO:Ce (right) samples.

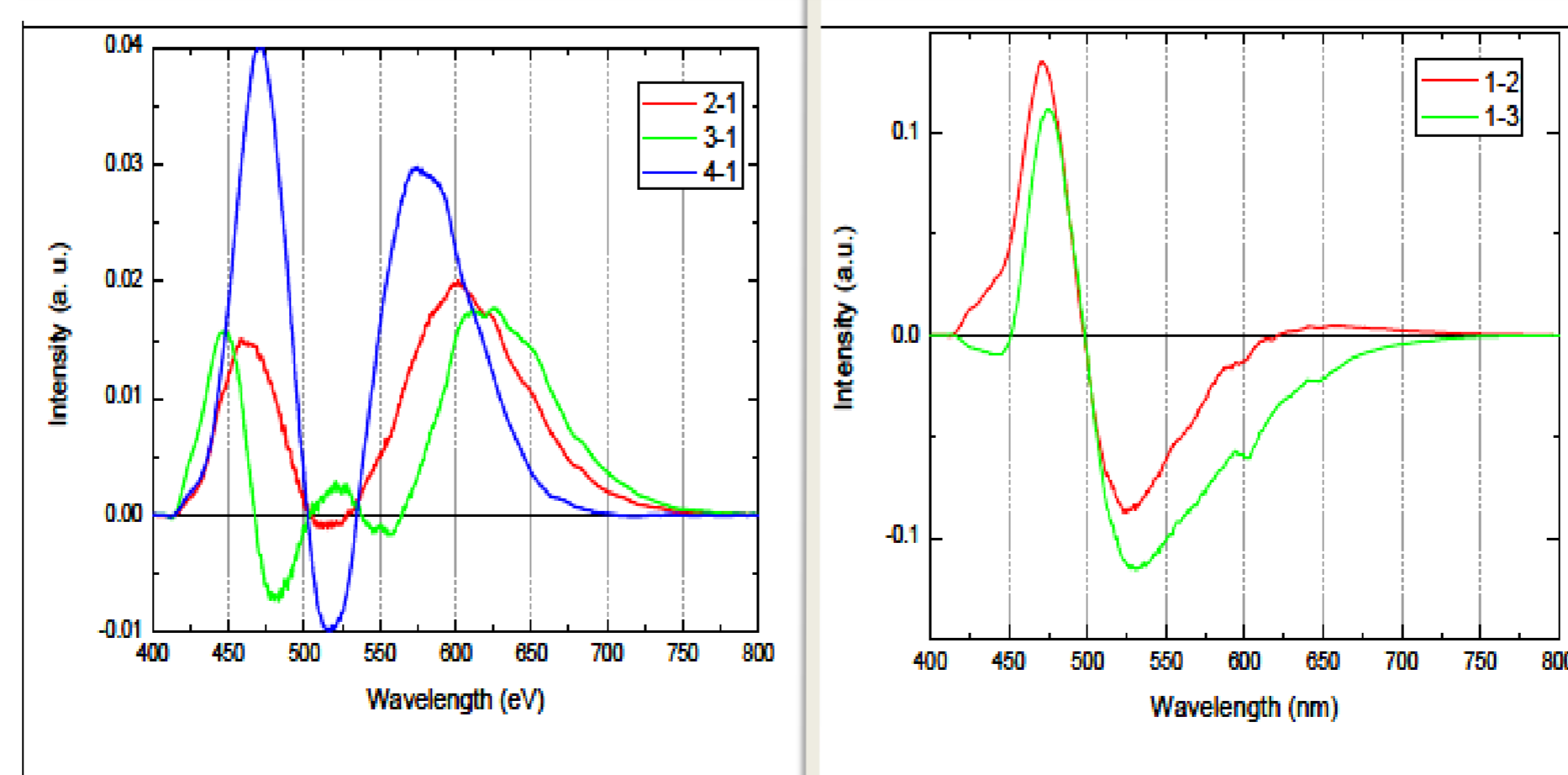


Fig. 5. Difference between the PL spectra in LGSO (left) and LSO (right) samples. PL spectra were normalized by their maxima before subtraction.

Discussion

With increasing distance from the surface the PL band in LSO:Ce gradually shifts to longer wavelength due to reabsorption. Meanwhile, spatial inhomogeneities of the order of 1-3 μm micron in diameter are observed for PL center of mass in LGSO:Ce (Fig. 3).

Comparison of the spectra (Fig. 4) recorded in the areas with different band position reveals a "zigzag" at 400-600 nm, which is probably linked to reabsorption of PL due to different distances from surface to the scanned areas (Fig. 5.)

The spectral component peaked in the vicinity of 600 nm was observed only in LGSO:Ce. This component can be attributed to variation of Ce1/Ce2 distribution caused by irregularities in Gd concentration across the sample (compare with the inset in Fig. 1). This peak is not observed in LSO:Ce, because of very low concentration of Ce2 centres. The different peak positions in the spectra in Fig. 1 (inset) and Fig. 5 are possibly due to different excitation type (in confocal measurements mainly Ce2 luminescence is excited).

Conclusions:

- The comparison of spatial distributions of PL parameters in LSO:Ce and LGSO:Ce crystals shows that the spatial inhomogeneity of the spectral component observed in the red region (>550 nm) of LGSO:Ce reflects fluctuations in Lu/Gd ratio across the crystal.
- Experimental observation of microscale inhomogeneities certifies the existence of smaller size regions as well with dimensions less than carrier capture length. Such fluctuations might influence distribution of carriers and should be taken into account at consideration of scintillation process in mixed crystals.

Acknowledgements

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