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Temperature behavior of Ce^{3+} emission in $(Lu, Y)_2SiO_5$ single crystals excited by vacuum ultraviolet synchrotron light

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ABSTRACT

Cerium doped (Lu,Y)₂SiO₅ (or LYSO) single crystals have been studied by means of luminescence excitation spectroscopy in the temperature range from 7 up to 300 K. Vacuum ultraviolet excitations in 4.5–8 eV energy range from synchrotron radiation of 1.5 GeV storage ring of MAX IV synchrotron facility. It is shown that both type of Ce^{3+} emission centers (seven and six coordinated centers) are excited under any excitation energy used. It was concluded that the same energy transfer processes from host lattice to impurity ions are involved independently on coordination of Ce^{3+} . It is also demonstrated that excitonic mechanism of energy transfer is dominant under chosen excitation and intrinsic and bound excitons are included in excitation of Ce^{3+} luminescence in LYSO.

1. Introduction

Cerium doped solid solution of lutetium and yttrium orthosilicates $Lu_{2(1-x)}Y_{2x}SiO_5$ (LYSO) is one of the most perspective scintillator due to a nice combination of properties needed for modern applications. LYSO is a non-hygroscopic material, it has a high density (7.1 g/cm³), it reveals a quick decay time (30–50 ns), high energy resolution (8–9% at 662 keV) and light yield (approximately 30,000 photons/MeV) [1–3].

However, although LYSO:Ce has been identified as a possible scintillator material for medical and nuclear applications, there is still a lack of fundamental knowledge about the final stage of energy relaxation processes [4-6], which is crucial for the evaluation of scintillator performance in relevant modern applications, such as the new CMS detector in CERN [14]. One of the most crucial factors influencing scintillator performance is the efficiency of energy transfer from the crystal lattice to the luminescence center (Ce^{3+}). As a rule energy transfer processes include migration processes of charge carriers to luminescence centers and, therefore, they are temperature dependent. Thus, the purpose of this study is a detailed study of temperature dependencies of luminescence properties of Ce³⁺ centers in LYSO single crystals. A special attention should be paid to a temperature range from liquid helium temperature to room temperature where delocalization of charge carriers from localized states and shallow traps take place. Considering that LYSO ($E_g = 6.8-7.4$ eV [7]) belongs to the class of wide-bandgap materials, vacuum ultraviolet (VUV) excitation should be applied for the successful excitation of excitonic states in this crystal. Hence, VUV excitation spectroscopy under synchrotron radiation excitation has been utilized in current research. This technique is a powerful method for the study of optical and luminescence properties of wide bandgap materials [8–14] as well as of wide bandgap semiconductors and semiconductor nanocrystals in some cases [15–17].

It is worth noting that Ce³⁺ ions has two nonequivalent positions in the LYSO crystal lattice. In LYSO Ce³⁺ ion preferentially substitutes Lu³⁺ due to similar ionic radii [18]. Depending on the number of oxygen ligands around these Ce^{3+} ions, two optically active cerium sites can be established: Ce1 and Ce2, with seven and six oxygen ligands, respectively. In other words LYSO has two different Ce³⁺ ion sites of C1 point group symmetry, i.e., with a coordination number of seven and six, each with two non-silicon bound oxygen atoms. Luminescence properties of both centers under photo excitation are summarized in Table 1. Temperature behavior of luminescence of both emission centers Ce1 and Ce2 have been reported in Refs. [19,20], where Ce³⁺ ions have been excited by low-energy photons which allow to realize intracenter 4f-5d transitions within Ce³⁺ ions. However, for scintillators it is important to know an energy transfer mechanism from host lattice to luminescence centers and, in the case of LYSO matrix, excitations in VUV energy range needed. Therefore, in current research we focused on the study of temperature behavior of luminescence properties of Ce³⁺ in LYSO under

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Table 1

Main luminescence characteristics of seven coordinated and six coordinated Ce^{3+} centers in LYSO.

Center Type	Emission wavelength, nm	Excitation wavelength, nm	Decay time, ns	References
Ce1	394	264	35–38	[19-22]
	427	294		
		364		
Ce2	460	325	48	[19,20]
			56 50	[22]



Fig. 1. 3D plot of the temperature dependence of the emission spectra in Ce^{3+} doped LYSO single crystals under excitation of 7.0 eV.

excitations allowing excite excitonic states and electron-hole pairs.

2. Materials and methods

Cerium-doped (LuY)₂SiO₅ (LYSO:Ce) single crystals were grown by the Czochralski method (Crystal Photonics, Inc.). According to the information provided by the manufacturer, the yttrium content in the crystal is about 5 %, while the nominal cerium doping level is 0.2 % [23, 24]. The crystals have been cut into 10×10 mm square with a thickness of 1 mm for luminescence experiments.

Luminescence spectroscopy measurements have been carried out under synchrotron radiation excitation. Luminescence excitation and emission experiments have been performed on the FinEstBeAMS [25,26] undulator beamline, which was constructed at the 1.5 GeV storage ring of the MAX IV [27] synchrotron facility in Lund, Sweden. In current research the excitation energy range was varied in 4.5–11.0 eV energy range. Luminescence detection in the UV–visible spectral range (200–700 nm) was performed by means of the Andor Shamrock (SR-303i) 0.3 m spectrometer, which was equipped with the CCD camera (Newton DU970P-BVF) covering the spectral range from 200 to 900 nm. LYSO crystals have been placed on the sample holder of the cold finger of a closed-cycle cryostat (Advanced Research Systems) and placed in an ultra-high vacuum chamber $(10^{-9} mbar)$. The experiments have been carried out in the temperature range from 10 K to 300 K. The excitation spectra have been corrected with respect to the synchrotron



Fig. 2. The emission spectra in Ce^{3+} doped LYSO single crystals under excitation of 7.0 eV in wide temperature range. The temperature dependencies taken at 394 nm and 520 nm emissions are normalized and shown inset.

light flux controlled by the AXUV-100G diode. More technical details of the endstation have been published elsewhere [28,29].

3. Results and discussion

The emission spectra of Ce^{3+} doped LYSO single crystals measured at wide temperature range are shown in 3D view in Fig. 1. In order to avoid a contribution of termostimulated luminescence signal into intensity of the emission spectra the emission spectra have been measured in cooling regime. The spectra have been measured by means of CCD camera capturing the spectral range of Ce1 and Ce2 emission bands. The emission spectra in Fig. 1 have been excited by 7 eV photons. This energy exceeds band gap energy in LYSO and Ce³⁺ have been excited after energy transfer from host lattice.

The emission spectra from Fig. 1 are plotted in standard 2D view in Fig. 2 for better visualization and their shape analysis. The emission spectra have a main peak at 427 nm with a well-resolved high-energy shoulder at about 394 nm, which transforms to a separate peak at low temperatures. Such double peak structure is typical for 5d-4f radiative transition in Ce³⁺ ion due to splitting of ground state. Based on literature data double peak emission (394 and 427 nm) belongs to the seven coordinated Ce³⁺ center in LYSO (see Table 1). Each emission spectrum depicted in Fig. 2 has a significant long wavelength tail which extends to 650 nm. Based on literature data six coordinated Ce³⁺ center (Ce2) is responsible for it (Table 1). It worth noting that in Ref. [30] authors explained the long wavelength emission in Ce³⁺ doped Lu₂SiO₅ (LSO) and Y₂SiO₅ (YSO) by dimer {Ce³⁺ - Ce³⁺} centers.

Temperature dependencies of intensities of 394 nm (Ce1 center) and 520 nm (Ce2 or dimer center) emissions are shown in Fig. 2 (inset). For better comparison both curves in Fig. 2 (inset) are normalized. One can see that these two temperature are almost identical. It is worth noting that the similarity of temperature dependencies of 394 nm and 520 nm depicted in Fig. 2 (inset) indicates also that the emission center responsible for the long wavelength tail in the emission spectra cannot be dimer {Ce³⁺ - Ce³⁺} center. Indeed, Ce1 and dimer {Ce³⁺ - Ce³⁺} center discussed in Ref. [30] have different temperature behavior of luminescence because of energy transfer from Ce1 to dimer {Ce³⁺ - Ce³⁺} center. Therefore, we assume that Ce1 and Ce2 centers are responsible for emission spectra shown in Figs. 1 and 2.

One can see that the intensities of both emission bands (Fig. 2 inset) are stable at low temperature (up to \sim 30 K). They grow up at



Fig. 3. The temperature dependence of the excitation spectra in Ce^{3+} doped LYSO single crystals. The emission was taken from total spectral range of Ce^{3+} emission. The arrows meaning see in the text.

temperature higher than 40 K reaching their maximum value at ~140–150 K. Furthermore, a slight intensities degradation takes place if temperature is higher than 150 K. The intensities growth in 40–150 K temperature range well correlates with temperature quenching of intrinsic emissions (emission bands of self-trapped exciton and self-trapped hole) observed and discussed in Ref. [31]. The degradation of intensities of both emissions in Fig. 2 (inset) at temperature higher than 150 K is due to standard thermal quenching of Ce1 and Ce2 centers [19, 20].

Fig. 3 shows the temperature dependence of excitation spectra of total emission within 350 nm–650 nm spectral range. Taking into account that intensity of Ce1 emission is much higher than emission of Ce2 centers we can assume that the excitation spectra exhibited in Fig. 3 are, in fact, the excitation spectra of Ce1 emission. Furthermore, we have measured the same temperature dependency of the excitation spectra for both emissions (Ce1 and Ce2) as well as their temperature dependencies are very similar. Therefore, we can conclude that energy transfer processes, which lead to the excitation of Ce1 and Ce2 enters are the same.

Let us to discuss the excitation spectra depicted in Fig. 3 in details. Each excitation spectrum shown in Fig. 3 has several similar features. First feature is the excitation bands in the low energy region of 4.5–5 eV and of 5.7–6 eV. This first excitation band results from 4f-5d transition in Ce^{3+} ion. This excitation band is well known in literature (Table 1) while the second excitation band is poorly studied so far and it can be related to the next 4f-5d band or to some intrinsic defects in LYSO lattice.

The next remarkable feature in Fig. 3 is the region above 6.0 eV, where excitonic transitions presumably appear. It can be seen that the excitonic band peaking at 6.55 at room temperature gradually shifts towards higher energies as the temperature decreases to 120 K. In this case, the intensity of the exciton band increases slightly. In temperature range below 120 K down to liquid helium temperature the intensity of the excitonic band significantly decreases. Simultaneously, the broad excitonic band splits forming the narrow band at about 6.5 eV with the well resolved shoulder at about 6.6 eV. The second values is very close to the energy position of the excitation peaks of intrinsic (self-trapped exciton or STE) emissions peaking at 256 nm in LYSO shown in Fig. 4. Detailed consideration of these two intrinsic emission bands as well as their temperature behavior we have reported separately in Ref. [31]. However here we would like to emphasize one's attention on the fact that excitation of the Ce³⁺ emission is shifted towards to low-energy side on 0.1-0.12 eV comparing with the excitations of intrinsic emissions



Fig. 4. Emission-excitation mapping of intrinsic as well as Ce^{3+} luminescence in LYSO single crystals at liquid helium temperature (7 K). Emission spectra at three different excitations (shown on the left side of the mapping) are depicted on the top graph, while three excitation spectra of three main emissions (denoted at the bottom of the mapping) are exhibited on the left graph.

(Fig. 4 right). Intrinsic emission has a maximum excitation at about 6.6 eV (Fig. 4 left) and it coincides with the excitation shoulder of Ce^{3+} emission at low temperature shown in Fig. 3. The arrows in Fig. 3 shows the main excitation peaks of intrinsic emission and Ce^{3+} emission observed in Fig. 4. The narrow energy region where Ce^{3+} emission is effectively excited but intrinsic emissions cannot is the range of excitation of bound exciton (or near-activator exciton). Thus, we suggest that two excitation peaks at about 6.6 eV and 6.5 eV at low temperature (Fig. 3) correspond to the excitation bands of STE and bound exciton respectively.

It is worth noting that the intensity of Ce^{3+} emission reaches its maximum in the temperature range of 120–150 K (Fig. 3). It is assumed that at this temperature the STE becomes mobile and is effectively localized near the luminescence center forming a bound exciton. At higher temperatures, a long-wavelength shift is observed in the excitation spectra, which is typical for temperature behavior of bound exciton. It is important also to note that, in contrast to the excitonic peaks, the spectral position of the excitation peak at 7.0 eV is stable over the entire temperature range. We assume that at energies above 7.0 eV, the excitation of luminescence centers occurs after the relaxation of electrons and holes excited through interband transitions, which is weakly dependent on temperature.

4. Conclusions

Luminescence characteristics of Ce^{3+} doped LYSO single crystals have been obtained in wide temperature range. It is shown that both type of Ce^{3+} emission centers (Ce1 and Ce2) are excited under any excitation energy used. We assumed that the same energy transfer processes from host lattice to Ce1 and Ce2 ions are involved. It is also demonstrated that excitonic mechanism of energy transfer is dominant under chosen excitation. The correlation between excitation spectra of self-trapped and bound excitons from one side and the excitation spectra of Ce^{3+} emission from another one is demonstrated and discussed. It is also suggested that low energy edge of the excitation curve at temperature higher than 120 K mostly formed by bound exciton states.

CRediT authorship contribution statement

V. Pankratova: Writing – original draft, Investigation, Formal analysis. K. Chernenko: Software, Methodology, Investigation. A.I. Popov: Resources, Funding acquisition, Formal analysis. V. Pankratov: Writing – original draft, Resources, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Vladimir Pankratov reports financial support was provided by University of Latvia Institute of Solid State Physics. Viktorija Pankratova reports financial support was provided by University of Latvia Institute of Solid State Physics. Anatoli I Popov reports financial support was provided by University of Latvia Institute of Solid State Physics. Kirill Chernenko reports financial support was provided by MAX IV Laboratory. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.omx.2024.100322.

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V. Pankratova et al.

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