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Can self-trapped excitons deliver energy to impurities in tungstates?

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Tungstates are widely used as scintillator materials in security, medicine and particle physics devices (e.g., $CaWO_4$, $CdWO_4$, $PbWO_4$). Their emission is represented by a broad band in the visible spectral region, which is usually ascribed to the radiative decay of self-trapped excitons (STEs) localized at oxyanion complexes. Due to the large energy of optical phonons and strong electron-phonon interaction in these systems hot electrons and holes created by high-energy quanta of ionizing radiation are believed to relax and form STEs very quickly, thus preventing energy from being transferred to defects and impurities. Despite tungstates of divalent metals have been popular research objects for decades, the basic physics features of their excitations, capable of confirming their excitonic nature, have not been revealed. No papers have been systematically dealing with this question except Ref. [1]. It is not known whether these excitations can be mobile or transfer energy to impurity ions. It is not even clear what is the reason of the thermal quenching of STE emission, i.e. is it thermal ionization, non-radiative recombination or hopping diffusion and energy transport. Here, we present the results of the studies on the STEs capability of transferring energy to impurity ions in tungstates.

The measurements were performed mainly on $CdWO_4$:Sm, which seems to be especially suitable for such study due to the availability of bridging oxygens connecting neighboring oxyanions in the crystals structure. The experiments were conducted at the I3 beamline of the MAX III storage ring, Lund, Sweden and laboratory spectroscopy setups at the Institute of Physics, University of Tartu. The dependence of excitation spectra of the STE and impurity emissions on temperature was measured in a wide energy region from 3 to 40 eV. A special attention was paid to the energy regions of intra-centre impurity excitation, direct exciton creation in the Urbach tail and the creation of excitons via electron-hole recombination in the fundamental absorption region. The measurements of emission decay kinetics were performed in a wide range of 4-600 K in order to reveal the mechanisms responsible for the energy transfer to impurity ions.

It is shown that while the intensity of the Sm^{3+} emission remains practically unchanged under the intra-centre excitation in the whole temperature range studied, it undergoes a remarkable evolution with temperature under excitation in the excitonic and fundamental absorption regions. In these energy regions, the intensity of the Sm^{3+} emission remains modest at low temperatures due to the competitive process of STE formation, grows drastically in the region of thermal quenching of the STE emission and thereafter the impurity emission disappears almost completely. The former effect is ascribed to the enhanced mobility of STEs due to hopping diffusion at elevated temperatures, while the latter - to the ionization of excitons at temperature near 500 K, which exceeds the STE emission quenching temperature by 100 K. The temperature dependences are different in the excitonic and fundamental absorption regions. A detailed analysis of the mechanism of energy transfer by STEs will be presented based on the results of time-resolved spectroscopy studies.

[1] K.W. Meert et al., J. Lumin. 173 (2016) 263.

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