Kinetic of the power-dependent up-conversion luminescence in Er^{3+}/Yb^{3+}-doped single nanocrystal

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This work analyzes the dynamics of the up-conversion process occurring in the single $NaYF_4$ nanocrystal (NC) doped with Er^{3+} and Yb^{3+} ions under varying excitation laser power. A confocal microscope was used to collect the time-dependent red emission of erbium ($^4F_{9/2}\rightarrow{}^4I_{15/2}$) activated by a laser operating at 980 nm with power changing from hundreds of μW to a few mW. A strong correlation between the laser power and the shape of the decay profile was observed, including changes in both the rising part and the decay tail. Furthermore, we noticed significant decrease in the average decay time with the increasing power of the excitation laser.

In order to explain this phenomenon, observed exclusively for small nanocrystals, we developed a new semi-empirical model of kinetic rate equations of the erbium-ytterbium system. Starting from the standard approach used for bulk crystals, we additionally assumed the presence of two independent erbium populations: internal, simulating ions isolated from the interaction with the environment, and external, attributed to the ions localized close to the NC's surface. For the latter, we introduced an additional nonradiative decay channel depopulating the red-emitting level through the interaction with surfactants. Moreover, an extended $Yb^{3+}\rightarrow Er^{3+}$ energy transfer model, including phonon statistics, was considered.

Eventually, we found that laser radiation prominently changes the temperature of the NC and tunes the up-conversion kinetics. Rising temperature helps to overcome a potential barrier for nonradiative decay and modifies the phonon occupation number in the case of energy transfer. Interestingly, the efficiency of these processes can be reduced by coating NCs with an optically passive shell, insulating external ions from the environment. The proposed kinetic model allowed us to estimate correct fits describing experimental luminescence decay profiles for various excitation powers. We found the presented approach valuable for the analysis of the up-conversion processes within a single nanocrystal and temperature sensing at the nanoscale.

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