Upconversion of Photoluminescence in II-VI Nanocrystals: Feasibility of Anti-Stokes Cooling

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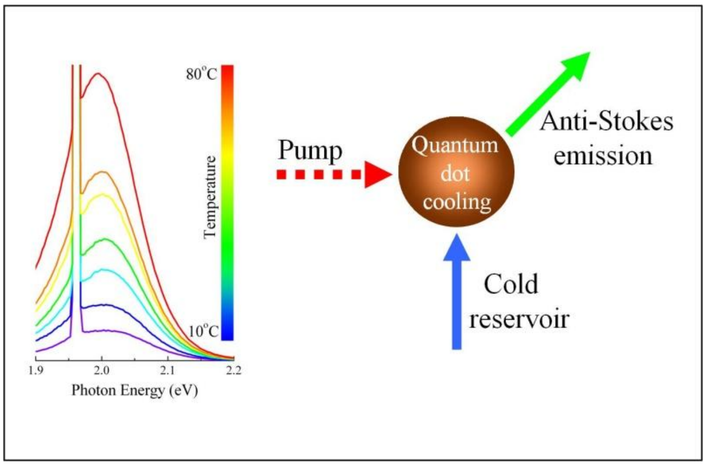
Highly luminescent semiconductor nanocrystals (NCs) possess a number of interesting and important properties that are tunable thanks to their size-dependent discrete electronic spectra characteristic of quantum dots (QDs). One of such properties is the up-converted photoluminescence (PL), i.e. the emission of photons with energy higher than that of the exciting photons. It is of great interest for applications in multi-colour displays, bio-imaging systems, unconventional lasers and solid-state optical refrigeration devices.
In this work, we consider the thermally activated photoluminescence up-conversion, also known as anti-Stokes photoluminescence (ASPL), where the additional energy comes from the phonon bath. ASPL has been observed in a variety of systems including ensembles of NCs of several semiconductor materials, such as CdSe, CdTe and CdSe/ZnS core/shell nanoparticles [1]. In order to observe the PL up-conversion, the sample must be excited at the lower limit of its absorption spectrum [1]. The anti-Stokes shift can reach 0.2-0.3 eV and the ASPL intensity increases linearly with excitation power and strongly rises with temperature.
We will present recent experimental results and the theory of this effect, based on the QDs polaron effect [2] and explaining, all the principal features of the ASPL in colloidal solutions of QDs. The proposed ASPL mechanism includes (i) polaron-mediated up-conversion by one optical phonon energy in single QDs, and (ii) cascade re-absorption and re-emission processes involving several QDs of successively smaller sizes within the sample, leading to the experimentally observed large anti-Stokes spectral shifts. The results obtained by our Monte-Carlo simulations based on the model outlined above, reproduce all the experimentally observed ASPL trends in ensembles of QDs and are being extended to layers of closely packed QDs [2]. The agreement with the experimental data confirms the validity of the proposed mechanism and provides predictive power for the ASPL applications.
We shall discuss in detail feasibility of one of such applications - the use of semiconductor QDs for optical cooling [1], [3].

References

[1] Y.P. Rakovich, J.F. Donegan, M.I. Vasilevskiy, A.L. Rogach, Phys. Stat. Sol. (a) 206 (2009) 2497
[2] J. R. Santos, M. I. Vasilevskiy, and S. A. Filonovich, Phys. Rev. B **78**, 245422 (2008).
[3] Y. Ninez-Fernandes, M. Vasilevskiy, in: Cadmium Telluride Quantum Dots: Advances and Applications, ed. By J.F. Donagan, Y.P. Rakovich (Pan Stanford, 2014), p.1-22

Figures



Temperature dependence of the ASPL in CdSe QDs (left) and Schematics of anti-Stokes optical cooling (right)