Multi-phonon Raman scattering in semiconductor quantum dots: the polaron effect

R. P. Miranda and M. I. Vasilevskiy

Centro de Física, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal

The electron-phonon interaction is important in semiconductors and, especially, in semiconductor nanostructures since it determines hot carrier relaxation, influences light absorption and emission processes and is responsible for Raman scattering. Owing to the electron and phonon confinement in quantum dots (QD's), the intensity of this interaction is enhanced, leading to multi-phonon processes and formation of a polaron [1]. The correct description of optical transitions requires a non-adiabatic and non-perturbative treatment of the many-body interactions taking place in these systems.

In this work, we propose a general non-perturbative approach to the calculation of the multi-phonon Raman scattering cross section for semiconductor QD's. The basic idea is to consider the most relevant exciton states and several confined optical phonon modes with the appropriate symmetry and truncate the Hamiltonian matrix by including a certain number of phonons allowed for each mode, large enough to guarantee that the result can be considered exact in the physically important region of energies. In this way, the system's Hamiltonian can be easily diagonalized numerically, yielding a very accurate solution for the polaron spectrum and stationary states of the quantum dot. Given the eigenvalues and eigenstates of the Hamiltonian, any physical quantity of interest, such as the Raman scattering cross section, can be easily obtained. In the polaronic picture, the Raman scattering process consists in two virtual transitions, one with absorption and the other with emission of a photon, in the exciton-polaron spectrum, which change the QD state (exciton vacuum with a different number of phonons in the beginning and in the end).

By applying the formalism to a set of CdSe model quantum dots, it is found that the overall scattering spectra can differ considerably from the perturbation theory predictions, especially in what concerns to the relative intensities between different order scattering processes. In particular, the Raman spectra calculated in the polaron picture for samples with typical size distributions show quite good agreement with previously published experimental results [2]. On the contrary, the perturbation theory approach fails in reproducing the features of these spectra. These results suggest that even for materials which have relatively small exciton-phonon coupling constants, such as CdSe, the polaron effects play an important role in the Raman scattering and can not be properly understood within the standard perturbation theory approach.

[1] S. Hameau *et al*, Phys. Rev. Lett. **83**, 4152 (1999)
[2] M.C.Klein *et al*, Phys. Rev. B **42**, 11123 (1990)