DIPOLE-ACTIVE OPTICAL VIBRATIONS CONFINED IN SEMICONDUCTOR QUANTUM DOTS

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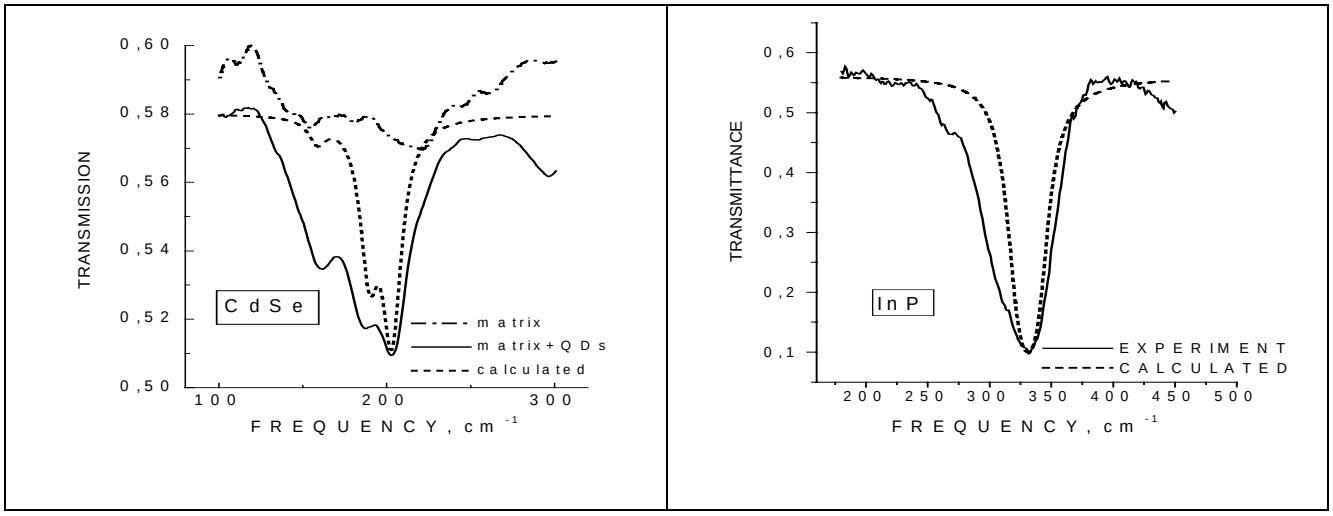
Dipole-active optical phonons confined in nearly spherical quantum dots (QDs) made of a polar semiconductor material were studied, both theoretically and experimentally. Spatial quantization of the phonon modes in a QD was considered theoretically (i) by solving the coupled continuous equation of motion and Poisson equation as proposed in [1], and (ii) numerically, within a model described in [2]. In both approaches, two kinds of boundary conditions (b.c.) have been applied, requiring the vanishing of either displacement or elastic force on the QD surface. It is shown that the FIR polarizability of a spherical QD in the phonon response region has a multimodal structure because of the contribution of different confined optical vibrations with angular momentum l=1. When the QD size increases this structure disappears and only a single peak in the FIR absorption spectra is expected, located at the Fröhlich frequency (ω_F) for both kinds of b.c. We confirmed this result by numerical lattice dynamics calculations. It should be pointed out that the Fröhlich mode is not an eigenmode of the considered macroscopic equations for any b.c. The absorption peak is formed by many those quantized modes which have frequencies close to ω_F. The total density of states calculated for a large spherical particle has only a small peak at ω_F in the case of rigid b.c. and no feature for free b.c.

Some of these theoretical conclusions have been confirmed by our experimental studies. QDs of CdS, CdSe and InP were prepared using colloidal synthesis methods. The mean QD radius ranged from 1 to 3 nm for CdS and CdSe and was 3nm and 5nm for InP. Far-infrared (FIR) reflection and transmission spectroscopy was used to assess the confined optical phonon modes. The quantized dipole-active modes were unambiguously observed in the FIR transmission spectra of very small CdSe nanocrystals (embedded in a matrix) and modelled (see left figure). For larger InP QDs, a single dip at the Fröhlich frequency was found (right figure). In the conference communication, we would like to discuss also the effect of QD concentration on the absorption and the reasons why FIR spectra taken from samples containing nanoparticles often show features near the bulk TO phonon frequency and not at ω_F (such a contribution of some mode near 310cm^{-1} is also seen in the InP spectrum below).



[1] E.Roca, C.Trallero-Giner and M.Cardona, Phys. Rev. B, **49**, 13704 (1994).
[2] M.I.Vasilevskiy et al, J. Phys.: Condensed Matter **13**, 3491 (2001).