Resonant Impulsive Generation of Coherent Optical Phonons in Quantum Dots

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We report on the ultrafast impulsive generation of coherent optical phonons in $CdTe_{1-x}Se_x$ quantum dots for laser energies in the vicinity of the absorption edge. Calculations based on a two-level model show very good agreement with the experimental data.

Pump-probe transmission experiments were performed at room temperature on a commercial sample (RG780, Schott Glass Technologies Inc.) consisting of ~ 4 nm – radius $CdTe_{0.32}Se_{0.68}$ nanocrystals [1] embedded in a borosilicate matrix. We used a low power Ti:sapphire oscillator operating at a repetition rate of 80 MHz that produced pulses of width 45-50 fs in the 740-820 nm (1.51-1.67 eV) range and balanced lock-in detection.

The accompanying figure shows in panel (a) the spectrally integrated differential transmission, ΔT, as a function of the probe delay, at various central energies of the laser pulse, E_C. The oscillations are due to the excitation of coherent optical phonons of the nanocrystals. As shown by the Fourier transform spectrum in panel (b), there are two modes at 4.8 and 6.0 THz. These frequencies agree well with those of the CdTe-like and CdSe-like longitudinal optical (LO) phonons determined by spontaneous Raman scattering. Panel (c) shows the measured optical transmission spectrum of the sample; the arrow indicates the energy of the first exciton state. The dependence of the CdSe-like

LO Fourier amplitude on E_C is shown in (d). Here, we followed [2] for calculating the frequency dependence of the dielectric constant of the composite, taking into account the size dispersion of the particles. Using the parameters obtained after fitting the transmission profile in (c), we modeled the system using the formalism of resonant impulsive stimulated Raman scattering in molecules [3], which is analogous to that discussed in [4] for solids. As shown in (d), the calculations show very good agreement with the measurements.
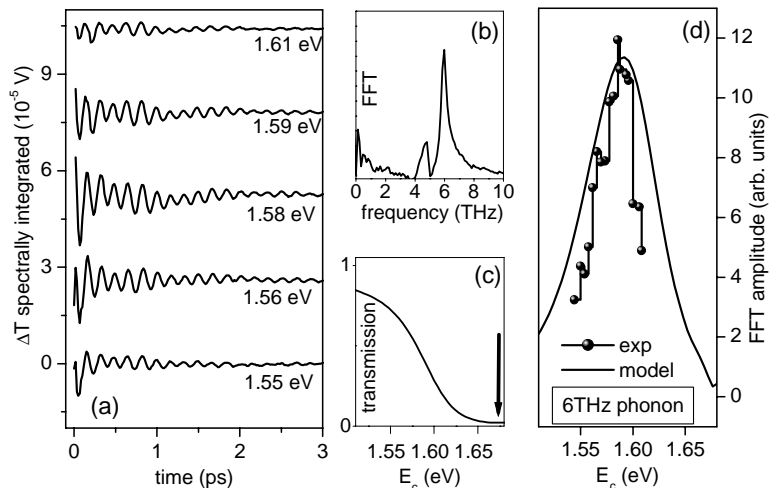
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