Local Phonon Modes in InAs/GaAs Quantum Dots

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Abstract. Local optical phonons in self-organized InAs/GaAs quantum dots (QDs) are investigated by Raman scattering under resonant excitation of the QD ground state transition. All QD sample structures, including single and stacked QD layers, pure InAs and $In_xGa_{1-x}As$ QDs as well as InAs QDs overgrown by GaAs and $In_xGa_{1-x}As$ quantum wells, show an interface mode at ~36 meV, a LO-like confined InAs QD mode at ~33 meV and a TO-like confined InAs QD mode at ~31 meV. For all samples the interface mode shows the strongest exciton-phonon coupling, followed by the QD LO mode.

INTRODUCTION

The observed rapid carrier relaxation in self-organized semiconductor quantum dots (QDs) [1] can only be explained by strong coupling of the carriers to optical phonons [2]. Electron-phonon coupling constants in III-V semiconductors, however, are small. Thus there must exist mechanisms to enhance the coupling. Charge localization of the carriers [3], enhanced polarity of the excitons [4] or localization of the participating phonon modes might enhance the electron-phonon coupling.

Here we present resonant Raman measurements revealing detailed information about local phonon modes in a variety of different InAs/GaAs QD structures and their strong coupling to the QD ground state exciton.

EXPERIMENT

We performed resonant Raman measurements on five different QD structures. All samples were grown in the Stranski-Krastanov growth mode by molecular beam epitaxy except sample E which was grown by metal-organic chemical vapor phase deposition.

Sample A consists of a single QD layer of 3 monolayers (ML) InAs, sample B of a seed QD layer of 1.74 ML InAs and an active layer of 3 ML InAs separated by a 36 ML GaAs spacer and sample C of a seed QD layer and five layers of 2.5 ML InAs and 45 ML GaAs spacer each. Sample D consists of 3 layers of 2.5 ML InAs each overgrown by an $In_{0.13}Ga_{0.87}As$ quantum well (17 ML) and a GaAs spacer (110 ML). Sample E has a single layer of $In_{0.8}Ga_{0.2}As$ QDs. Low-temperature photoluminescence (PL) spectra of all samples are shown in Fig. 1 for low-density excitation above the GaAs bandgap.

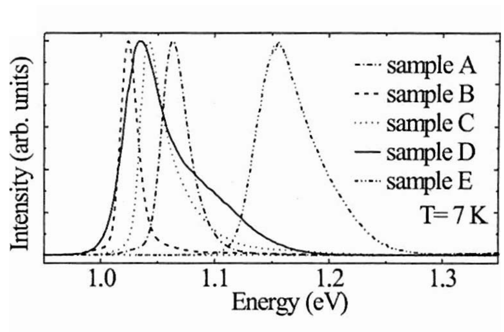

FIGURE 1. Normalized low-density PL spectra for all QD samples for non-resonant GaAs excitation at T=7 K.

The samples were mounted in a He-immersion cryostat and excited by spectrally narrow (~2 meV) ps-pulses of an optical parametric oscillator pumped by a Ti:sapphire laser. The signal was detected time-integrated through an additive double monochromator and a Ge diode.

RESULTS

The PL peaks in Fig. 1 show weak low energy shoulders due to phonon-assisted exciton recombination which is well-resolved by resonant excitation of the ground state transition. A series of such resonantly excited Raman spectra is depicted in Fig. 2 (a) for sample A and (b) for sample E. The excitation energy, which is marked by arrows in the insets of Fig. 2, is stepped across the inhomogeneously broadened ground state transition. When exciting below the ground state transition a weak doublet attributed to GaAs bulk LO and TO phonon modes is visible. With increasing excitation energy the

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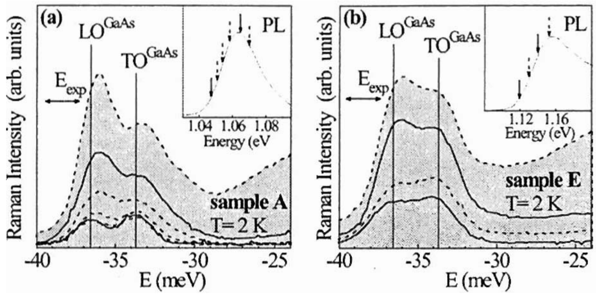

FIGURE 2. Resonant Raman spectra for (a) sample A and (b) sample E. Arrows in the insets show the excitation energies. The experimental resolution is indicated by ΔE_{exp}.

peaks become more intense and show a clear low-energy shift. Finally, exciting on the high energy side of the PL peak leads to additional broad peaks which are attributed to electronic transitions due to excitons generated in excited states putting an upper limit to the excitation energies giving interpretable Raman spectra [5].

The intensities of the Raman peaks as a function of the excitation energy show a resonance behavior following the inhomogeneously broadened absorption profile of the ground state transition proving the local character of the interaction. The low-energy shifts of the Raman peaks in resonance show that the phonons coupling to the QD excitons also have a local character. The apparent continuous peak shifts arise from the superposition of non-resonant (GaAs bulk) and resonant (QDs) signal resulting in a well-defined energy and coupling of the local phonon modes. The intensity ratios of resonant and non-resonant Raman signal are determined by the coupling constants of the local modes on the one hand

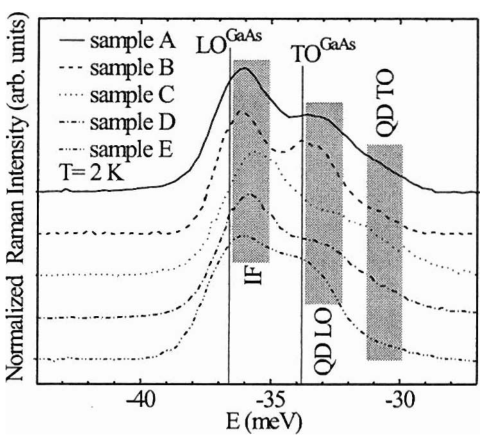

FIGURE 3. Full resonance Raman spectra for all investigated samples. Spectra were vertically shifted and normalized for clarity. The signal underground from electronic transitions (see text) was substracted.

and the number of resonantly excited QDs on the other hand. The number of excited quantum dots depends on the spectral width of the excitation light source and the spectral and spatial QD density.

Full resonance spectra for all investigated samples are shown in Fig. 3. All spectra show 3 local modes attributed to the interface mode (IF, ~36 meV), the confined LO phonon of the strained InAs QDs (~33 meV) and the QD TO phonon (~31 meV). The interface phonon shows the strongest coupling for all samples followed by the QD LO phonon. The coupling of QD TO phonon is comparatively weak.

Identification of the confined QD modes is based on our strain modelling presented in [6]. Strain-induced phonon frequency shifts are described by phonon deformation potentials. The compressive strain present in the InAs QDs increases the Γ-Point LO phonon energy from 30.3 meV [7] to 33.8 meV, whereas the TO phonon energy is increased from 27.9 meV [7] to 31.5 meV. The energy and stronger coupling of the 36 meV mode suggests this mode to be an interface mode, which is supported by Raman studies presented in [8].

CONCLUSION

We have reported resonant Raman measurements on various self-organized InAs/GaAs QD structures including single and stacked QD layers, $In_xGa_{1-x}As$ QDs and InAs QDs overgrown by $In_xGa_{1-x}As$ quantum wells. Resonant excitation of the QD ground state exciton reveals three local phonon modes at ~36 meV (interface phonon), ~33 meV (QD LO phonon) and ~31 meV (QD TO phonon) for all samples. The interface phonon shows the strongest coupling for all samples followed by the QD LO phonon.

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