Resonant Raman scattering in self-organized InAs/GaAs quantum dots

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The exciton-phonon coupling in self-organized InAs/GaAs quantum dots (QDs) is investigated under resonant excitation of the ground-state transition. First- and second-order phonon sidebands of the TO (30.3 meV) and LO (33.2 meV) modes of the strained InAs QDs as well as an interface (35.9 meV) mode are resolved. Huang–Rhys factors of 0.012, 0.026, and 0.006, respectively, indicate enhanced polar exciton-phonon coupling in such strained low-symmetry QDs. Time-resolved measurements support the local character of the phonon modes. © *2000 American Institute of Physics.* [S0003-6951(00)04448-X]

The exciton–phonon interaction in quantum dots (QDs) has attracted increasing interest in the last decade,[1–3] in particular with the success of self-organized QDs.[4] The interaction with phonons is intricately connected to relaxation processes as well as the homogeneous broadening at elevated temperatures, both being of fundamental importance for device applications. The limitations in achieving energy conservation in carrier relaxation processes have led to the prediction of a phonon bottleneck effect.[5] Confinement-induced local charge neutrality is expected to decrease the polar coupling whereas the gradual breakdown of *k*-selection enhances the coupling to acoustic phonons.[1] Nevertheless, such predictions are seemingly in contradiction to experimental results for self-organized QDs showing fast carrier relaxation[6] and LO-phonon scattering to dominate the temperature dependence of the homogeneous linewidth.[7,8]

The investigation of the exciton–phonon coupling in QDs requires to overcome the generally large inhomogeneous broadening employing size-selective spectroscopy, e.g., using selectively excited photoluminescence (SEL)[9,10] or resonant Raman scattering.[11,12] The observed phonon sidebands give access to the involved phonon modes as well as the coupling strength. In the adiabatic approximation the phonons are described by an unperturbed Hamiltonian and the exciton–phonon interaction Hamiltonian is taken linear in the vibrational amplitude leading to a displaced harmonic oscillator model with the dimensionless displacement Δ.[1] This model predicts a $T=0$ Poisson distribution of the intensity among the phonon sidebands ($I_N \sim S^N/N!$) with N the number of phonons generated and S being the Huang–Rhys factor ($S=\Delta^2$) describing the coupling strength.

Here, we present a time-integrated and time-resolved study of resonant Raman scattering for self-organized InAs/GaAs QDs yielding detailed information on the interaction of excitons with local phonon modes of the InAs QD layer.

The investigated sample was grown by molecular beam epitaxy on semi-insulating GaAs(001) using punctuated island growth as described in Ref. 13. InAs was deposited at 500 °C, an As_4 partial pressure of 6×10^{-6} Torr, and a growth rate of 0.22 ML s^{-1}. The total InAs amount of 3 ML was deposited in a two-step process with a growth punctuation of 60 s after the deposition of the first 1.74 ML. Immediately following the InAs deposition the temperature was reduced to 400 °C for the growth of the GaAs cap by migration enhanced epitaxy minimizing modifications of the island shape and composition during overgrowth.[14] For an equivalent uncapped sample an island density of $\sim$660 μm^{-2} and an average island height of $\sim$8 nm were found.[13] The samples were mounted in a He-immersion cryostat and excited by spectrally narrow ($\sim$1 meV) ps pulses of an optical parametric oscillator pumped by a Ti:sapphire laser. The signal was detected either time integrated through an additive double monochromator with a Ge diode or time-resolved through a subtractive double-grating monochromator with a multi-channel-plate photomultiplier with a *S*1-cathode in photon-counting mode. In the latter case the system response with a full width at half maximum (FWHM) of $\sim$20 ps was taken into account in the analysis of the transients using convolution techniques.

A low-temperature photoluminescence (PL) spectrum is shown in Fig. 1(a) for low-density excitation in the GaAs barrier. A single PL peak with a FWHM of 25 meV is observed at 1.062 eV. The PL peak shows weak low-energy shoulders due to phonon-assisted exciton recombination, which evolve in a well-resolved phonon sideband under resonant excitation.[10] Intense laser excitation improves the signal-to-noise ratio by orders of magnitude allowing to investigate the resonant excitation behavior in more detail. Figure 1(b) shows a logarithmic plot of a series of PL spectra for which the excitation energy is stepped across the inhomogeneously broadened ground state transition. The spectra have been vertically shifted for clarity and the dotted vertical lines indicate the excitation energies. Exciting below the QD ground state transition (topmost spectra) a weak doublet structure due to GaAs Raman scattering is resolved. Increasing the excitation energy the strength of the phonon sidebands became more intense and two-phonon peaks become visible. Finally, exciting in the high-energy shoulder the

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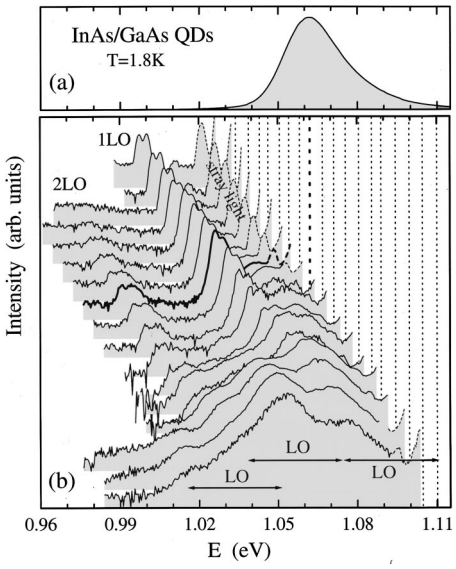

FIG. 1. (a) Low-density PL spectrum of InAs/GaAs QDs for nonresonant GaAs excitation at $T=1.8$ K. (b) Resonantly excited PL spectra on a logarithmic scale (the spectra are shifted vertically for clarity). The dashed vertical lines indicate the excitation energies.

spectra change significantly. The resonances become broad (FWHM$\sim$10 meV) and additional resonances appear at $\sim$1.05 and $\sim$1.01 eV in between the LO-replica. The latter are attributed to SEL generating excitons in excited states.[10,15]

The intensity of the 1LO sideband as a function of the excitation energy shows a double-resonance behavior attributed to phonon-assisted absorption and emission processes in the QDs.[10] Figure 1(b) now demonstrates a qualitative difference between phonon-assisted absorption and emission processes. The satellites observed for resonant excitation of the ground state are spectrally sharp owing to the well-defined phonon energies. The large widths observed for excitation at higher energies suggests the involvement of excited state absorption being subject to the inhomogeneous broadening due to shape and composition fluctuations. For example, the broad resonance appearing $\sim$20 meV below the excitation energy can be attributed to the formation of an exciton with the hole in the second excited state.[15]

In the following we concentrate on spectra excited in the low-energy shoulder of the ground state PL showing clear phonon sidebands. Figure 2 depicts the 1LO resonance for various excitation energies as indicated in the inset. For nonresonant excitation below the QD ground state absorption a double-peak structure is observed with peaks at 33.6 and 36.5 meV corresponding to the GaAs TO(Γ) and LO(Γ) modes, respectively. Tuning the excitation energy into the maximum of the ground state PL leads to a significant increase in signal strength and a low-energy shift of the phonon sidebands indicating resonant Raman scattering with the incoming resonance corresponding to the QD ground state transition. The experiments probe the coupling of the localized exciton to the local phonon spectrum of the QD structure.

Figure 3 depicts the phonon sidebands for resonant excitation in the maximum of the ground state PL. The full lines represent fits of three Gaussians to both the 1LO and

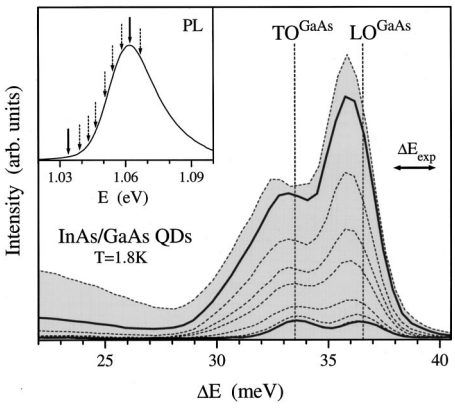

FIG. 2. Resonant Raman scattering of self-organized InAs/GaAs QDs. Arrows in the inset show the excitation energies.

2LO sidebands, yielding phonon energies of 30.3, 33.2, and 35.9 meV. The FWHM of the highest energy mode (35.9 meV) is limited by the experimental resolution, suggesting a well-defined phonon energy in the QD ensemble. Raman data for InAs/GaAs QD layers exciting resonantly the $E_0+\Delta_0$ band gap[16] suggest this mode to be an interface mode (IF). The 33.2 meV mode is likely to be a bulk-like mode of the compressively strained InAs inside the QDs. Based on the strain distribution in coherent InAs/GaAs pyramids an energy of $\sim$32.6 meV was estimated for the InAs LO(Γ) mode.[6,10] The energy of this mode depends on the strain distribution and, therefore, the actual shape and composition of the islands. The large FWHM of 4.9 meV of the 2LO resonance (not limited by the experimental resolution) is attributed to the inhomogeneity of the QD ensemble. Following the same line of arguments the 30.3 meV mode is attributed to the InAs QD TO phonon. Recently, coupling to the InAs wetting layer and GaAs phonons was reported for smaller ($\sim$12 nm base length) InAs/GaAs QDs,[6] which is not resolved in the present resonant Raman scattering experiments. The results suggest that the smaller size favors coupling to the barrier modes compared to the QD modes due to the increased penetration of the exciton wave function into the wetting layer and GaAs barrier.

The polar coupling of excitons to LO phonons is a function of the local charge density.[2] Thus, for QDs with identical electron and hole wave functions the polar coupling should vanish.[1] The separation of the electron and hole

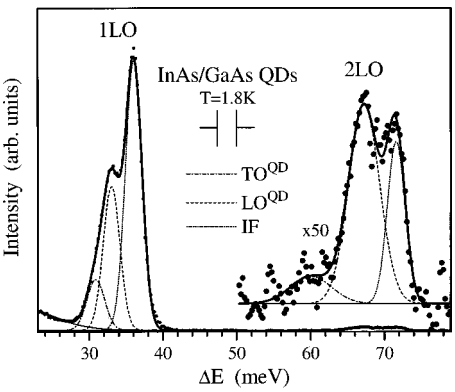

FIG. 3. First and second order resonant Raman scattering of self-organized InAs/GaAs QDs (dotted spectra). Full lines represent fits assuming, respectively, three Gaussians.

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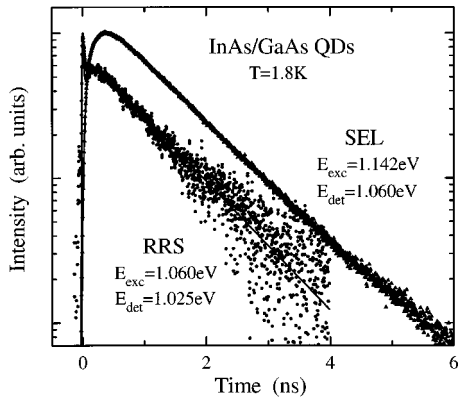

FIG. 4. Dynamical response of InAs/GaAs QDs to resonant excitation. The upper transient (triangles) shows selectively excited luminescence (SEL) of the ground state exciton when exciting at 1.142 eV an excited state transition. The solid line represents a two-exponential fit. The lower transient (dots) shows the first order resonant Raman scattering (RRS) upon resonant excitation of the ground state exciton at 1.060 eV.

charges in the complex confinement potentials of strained self-organized QDs potentially enhances the coupling.[10] The intensity ratios of the 1LO and 2LO sidebands (Fig. 3) correspond to Huang–Rhys factors S of 0.012, 0.026, and 0.006 for the QD TO and LO modes as well as the interface mode, respectively, in good agreement with Ref. 10.

Finally, the data presented for resonant excitation raise the question of the nature of the scattering process. Previously, we have attributed the phonon sidebands observed in SEL spectra to phonon-assisted exciton recombination based on the excitation behavior.[10] PL excitation spectra revealed the same excitation channels for the zero-phonon and the one-phonon transition indicating an incoherent three-step process. An exciton generated optically in an excited state, relaxes to the ground state loosing its coherence, and finally generates a LO-phonon in the recombination process. This process is shown time resolved in Fig. 4. The transient marked SEL (triangles) shows the dynamic response of the ground state exciton recombination when exciting at 1.142 eV an excited state transition.[15] A fit assuming exponential rise and decay processes (solid line) yields time constants of 150 and 975 ps, respectively. The long rise time is attributed to slowed-down carrier relaxation in the QDs. The decay time of 975 ps is typical for the spontaneous radiative decay of the ground state exciton.[6] The transient marked RRS (dots) shows the time evolution of the 1LO sideband for resonant excitation at 1.060 eV. The initial fast component is attributed to bulk-GaAs Raman scattering. Subsequently, the signal decays exponentially with the spontaneous lifetime of the ground state exciton demonstrating the resonant origin of the observed phonon sidebands.

Resonant Raman scattering and luminescence under resonant excitation can be described as a unified second order process.[17,18] The processes become distinct only in the presence of pure dephasing. The resonant Raman signal occurs during the coherence time of the resonant state, whereas luminescence occurs during the recombination time. Thus, exciting the ground state exciton of the QDs at He temperatures the distinction between resonant Raman scattering and luminescence might become meaningless providing that the localization suppresses pure dephasing processes as observed, e.g., for bound excitons in CdS.[19,20] An experimental distinction between resonant Raman scattering and luminescence would require detailed polarization-dependent measurements or the independent determination of the dephasing time, which is beyond the scope of this letter.

In conclusion, we have reported on resonant Raman scattering for self-organized InAs/GaAs QDs. Resonant excitation of the QD ground state exciton reveals coupling to the QD TO and LO phonons as well as an interface mode. The coupling to the local InAs QD modes is enhanced compared to the bulk case. The phonon sidebands are shown to decay with the radiative lifetime of the exciton state demonstrating the local character of the observed phonon sidebands.

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[1] S. Schmitt-Rink, D. A. B. Miller, and D. S. Chemla, Phys. Rev. B **35**, 8113 (1987).
[2] S. Nomura and T. Kobayashi, Phys. Rev. B **45**, 1305 (1992).
[3] V. M. Fomin, V. N. Gladilin, J. T. Devreese, E. P. Pokatilov, S. N. Balaban, and S. N. Klimin, Phys. Rev. B **57**, 2415 (1998).
[4] For a recent review, see, D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, London, 1999).
[5] H. Benisty, C. M. Sotomayor-Torres, and C. Weisbuch, Phys. Rev. B **44**, 10945 (1991).
[6] R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov, Phys. Rev. B **56**, 10435 (1997).
[7] F. Gindele, K. Hild, W. Langbein, and U. Woggon, Phys. Rev. B **60**, R2157 (1999).
[8] V. Türck, S. Rodt, O. Stier, R. Heitz, U. W. Pohl, R. Engelhardt, and D. Bimberg, J. Lumin. **87–89**, 337 (2000).
[9] D. J. Norris, Al. K. Efros, M. Rosen, and M. G. Bawendi, Phys. Rev. B **53**, 16347 (1996).
[10] R. Heitz, I. Mukhametzhanov, O. Stier, A. Madhukar, and D. Bimberg, Phys. Rev. Lett. **83**, 4654 (1999).
[11] A. P. Alvisatos, T. D. Harris, P. J. Carrol, M. L. Steigerwald, and L. E. Brus, J. Chem. Phys. **90**, 3463 (1989).
[12] G. Scamarcio, V. Spagnolo, G. Ventruti, M. Lugará, and G. C. Righini, Phys. Rev. B **53**, R10489 (1996).
[13] I. Mukhametzhanov, J. Wei, R. Heitz, and A. Madhukar, Appl. Phys. Lett. **75**, 85 (1999).
[14] Q. Xie, P. Chen, A. Kalburge, T. R. Ramachandran, A. Nayfonov, A. Konkar, and A. Madhukar, J. Cryst. Growth **150**, 357 (1995).
[15] R. Heitz, O. Stier, I. Mukhametzanov, A. Madhukar, and D. Bimberg, Phys. Rev. B **62**, 11017 (2000).
[16] Yu. A. Pusep, G. Zanelatto, S. W. da Silva, J. C. Galzerani, P. P. Gonzalez-Borrero, A. I. Toropov, and P. Basmaji, Phys. Rev. B **58**, R1770 (1998).
[17] Y. Toyozawa, J. Phys. Soc. Jpn. **41**, 400 (1976).
[18] Y. Toyozawa, A. Kotani, and A. Sumi, J. Phys. Soc. Jpn. **42**, 1495 (1977).
[19] H. Stolz, V. Langer, E. Schreiber, S. Permogorov, and W. van der Osten, Phys. Rev. Lett. **67**, 679 (1991).
[20] B. Lummer, R. Heitz, V. Kutzer, J.-M. Wagner, A. Hoffmann, and I. Broser, Phys. Status Solidi B **188**, 493 (1995).

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