Enhanced Polar Exciton-LO-Phonon Interaction in Quantum Dots

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Phonon-assisted exciton transitions in self-organized InAs/GaAs quantum dots (QDs) are investigated by photoluminescence (PL) and PL excitation spectroscopy. An unexpectedly enhanced polar exciton-LO-phonon interaction for such strained low-symmetry QDs is found. Calculations in the adiabatic approximation indicate that the particular quantum confinement and piezoelectric effect together account for the enhanced coupling. The results provide new insights into the long-standing problem of the exciton-phonon interaction in zero-dimensional systems.

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In recent years considerable effort has been devoted to the understanding of the exciton-LO-phonon interaction in low-dimensional semiconductor heterostructures [1,2], especially semiconductor quantum dots (QDs) [3–6]. For QDs in the strong confinement regime with confinement energies exceeding the exciton binding energy and the optical phonon energy the mixing of different electronic states by the phonon coupling can be neglected. In this adiabatic approximation the phonons are described by the unperturbed Hamiltonian, and the interaction Hamiltonian, taken to be linear in the vibrational amplitude, leads to a displaced harmonic oscillator model with the dimensionless displacement Δ [7,8]. This model predicts that the electronic oscillator strength distribution over phonon sidebands follows, at $T = 0$, a Poisson distribution $I \sim S^n e^{-S}/n!$ in which S is the Huang-Rhys parameter ($S = \Delta^2$) and n the number of phonons generated in the transition. The experimental observation of such phonon sidebands in resonant Raman scattering experiments [9–11] and in studies of phonon-assisted exciton transitions [12–14] yielded Huang-Rhys parameters S between 0.1 and 1 for nearly spherical II-VI QDs.

Theoretical descriptions of the polar exciton-LO-phonon interaction based on the adiabatic description have proven successful in higher dimensional systems [1,2] but seem to fail in the case of QDs. Calculations for unstrained spherical QDs [3,5] predict 1 to 2 orders of magnitude smaller Huang-Rhys parameters S than experimentally observed. For an exciton the polar coupling strength (described by S) is proportional to the squared absolute value of the Fourier transformed *difference* of the probability densities of the electron and hole. Therefore, the confinement-induced local charge neutrality is predicted to reduce the coupling [3]. To reconcile this striking discrepancy, charge separation by defect or surface states has been proposed as a possible extrinsic origin of increased LO-phonon coupling [5]. Alternatively, Fomin *et al.* [6] proposed a breakdown of the *adiabatic* approximation and showed that a *nonadiabatic* treatment of the electron-phonon interaction might offer an intrinsic means to explain the experimental data.

In this Letter, we present the first experimental and theoretical study of the polar exciton-LO-phonon coupling in self-organized InAs/GaAs QDs. The study reinforces the *adiabatic* description. For such defect-free QDs [15] the low symmetry and the macroscopic piezoelectric fields induced in the binary materials by the shear strains [16] lead intrinsically to distinctly different electron and hole wave functions and, thus, present a model system for the study of "distorted" QDs. Recently, the observation [17–19] of pronounced multi-LO-phonon resonances in photoluminescence excitation (PLE) spectra of such QDs has, despite predictions of generally low transition probabilities [20], unambiguously established inelastic LO-phonon scattering as the dominant relaxation process. However, the coupling strength relevant to the relaxation processes could not be established. Here, we report on phonon-assisted exciton transitions in self-organized InAs/GaAs QDs that allow extracting the Huang-Rhys parameter S. The experimental results are discussed based on calculations in the adiabatic approximation using *realistic* wave functions obtained in the eight band $\mathbf{k} \cdot \mathbf{p}$ approximation for pyramidal InAs/GaAs QDs [16].

The investigated samples were grown by molecular beam epitaxy on semi-insulating GaAs(001) as described in detail in Refs. [21,22]. For the active QD layer 3 ML InAs were deposited in the conventional continuous fashion, by using punctuated island growth (PIG) [21], or by using the variable deposition amount (VDA) approach [22]. In the latter approach vertically stacked pairs of unequal-sized QDs are formed in which only the larger QD in the second layer is optically active [23]. The samples are dubbed 3ML, PIG, and VDA, respectively. Atomic force microscopy (AFM) studies of equivalent uncapped samples show the island density to decrease ($\sim$900, $\sim$660, and $\sim$480 μm^{-2}, respectively) and the island height and width to increase in this series [21,22]. Cross-sectional transmission electron microscopy data and AFM images [22] yield an average island base length of $\sim$19 nm and height of 9.6 $\pm$ 0.7 nm, respectively, for an uncapped 3ML VDA sample. The increased island densities in the

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PIG and 3ML samples suggest smaller average island base lengths of ~16.2 and ~14.5 nm, respectively. The PL and PLE spectra were taken at 7.0 K in a continuous-flow He cryostat with a tungsten lamp dispersed by a 0.27 m double-grating monochromator as a tunable light source and a 0.3 m double-grating monochromator and a cooled Ge^- diode for detection.

Figure 1 compares the PL spectra of the three samples excited in the GaAs barrier on a linear [1(a)] and a semilogarithmic [1(b)] scale. The 3ML sample shows a PL peak at 1.084 eV with a full width at half maximum (FWHM) of 38 meV which is shifted to lower energies for the PIG and VDA samples (1.062 and 1.027 eV, respectively), consistent with the increasing QD size. An improved island uniformity contributes to the narrower FWHM (25 meV) in the latter two samples and a finite, growth-mode dependent, abundance of not fully developed islands accounts for the comparatively flat high-energy PL slopes [1(b)] [21,22].

The features of interest in the present paper are the weak low-energy shoulders of the QD PL peaks, marked by dashes in Fig. 1(b). These structures are clearly resolved for the PIG and VDA samples, which exhibit a PL FWHM smaller than typical LO-phonon energies in the InAs/GaAs system. The second derivative of the PL intensity [see inset of Fig. 1(a) for the VDA sample] reveals two clear dips forming a progression with a period of ~34 meV that is close to the QD LO-phonon energy [18]. These sidebands, at energies lower than the ground state transition energy, suggest phonon-assisted exciton recombination. The main PL peak is the zero-phonon line (ZPL) and the low energy shoulders are the 1 and 2 LO-phonon sidebands.

The phonon-assisted exciton transitions become well resolved in size-selective PL and PLE spectra, circumventing the inhomogeneous broadening of the QD ensemble. Figure 2(a) depicts such PL and PLE spectra for the VDA sample together with the inhomogeneously broadened PL spectrum (unscaled, uppermost spectrum). Equivalent spectra are observed for the 3ML and PIG samples (not shown). The PLE spectrum, recorded at the maximum of the QD PL peak (dashed line), reveals a series of resonances, which correspond to excited state transitions of the QDs [24,25] with a ground state transition energy matching the detection window at (1027.2 ± 1.7) meV. The only exception is the resonance at 1.062 eV which, independent of the detection energy, appears ~35 meV above the ground state transition, suggesting a phonon resonance (dubbed +LO). However, on the high-energy side of the ground state transition superposition with excited state absorption [18] might contribute to the comparatively strong signal. Upon size-selective excitation of the PLE resonance at 1.103 eV, the QD PL peak at 1.027 eV becomes narrower (FWHM of 11 meV) [26] and a peak (dubbed −LO) is clearly resolved in the region of the first low-energy shoulder, having ~1.5% of the intensity of the ZPL. The −LO replica becomes a separate peak when exciting directly the ZPL. The integrated intensity of the

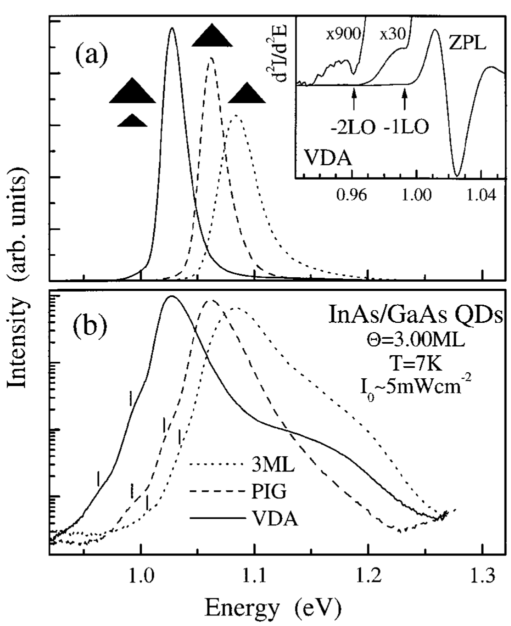

FIG. 1. Low-temperature PL spectra of the 3ML, PIG, and VDA samples excited nonresonantly at 2.41 eV on linear (a) and semilogarithmic (b) scales. The inset of (a) shows the second derivative of the PL spectrum for the VDA sample.

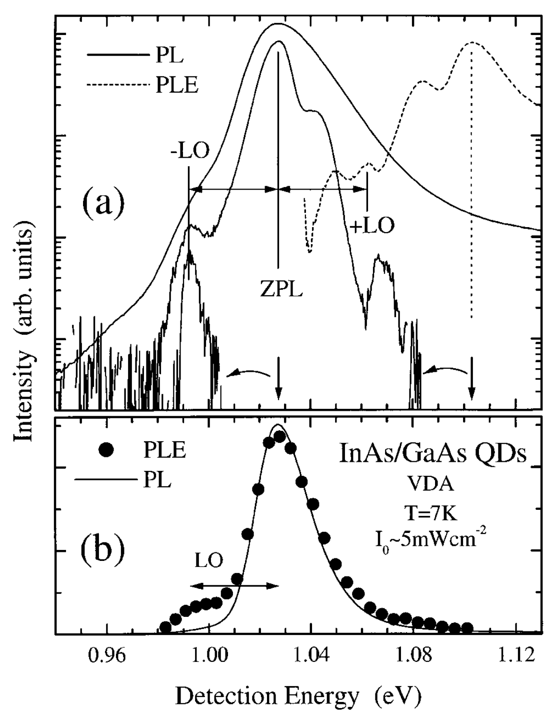

FIG. 2. (a) Low-temperature PL (solid lines) and PLE (dashed line) spectra for the VDA sample on a semilogarithmic scale. The PLE spectrum was detected on the QD PL maximum at 1.027 eV (marked ZPL) and the PL spectra were excited in the GaAs barrier (uppermost spectrum), at 1.103 eV and at 1.027 eV via the ZPL. (b) The intensity of the +LO resonance in PLE spectra as a function of the detection energy.

+LO-phonon replica observed in PLE spectra is depicted in Fig. 2(b) as a function of the detection energy. It reveals a resonance at 1.027 eV matching the inhomogeneously broadened ground state PL peak and another at 0.993 eV shifted by one LO-phonon energy. The LO phonon is excited in the generation and recombination of the ground state exciton, respectively. Furthermore, the PLE spectra of the ZPL (scaled down by a factor of 70) and of the −LO resonance reveal identical excitation resonances (Fig. 3) and, therefore, probe the same QD subensemble [27]. The PLE spectrum of the phonon sideband reveals size-selective ground state absorption in perfect resonance with the ZPL emission, which unambiguously demonstrates intrinsic QD recombination.

Finally, the inset of Fig. 3 depicts on an enlarged energy scale a PL spectrum of the PIG sample excited in the ZPL. The full line represents a fit with two Gaussians, which reveals phonon modes at 32.3 and 35.7 meV. Similar spectra are observed for the 3ML and VDA samples (not shown). The FWHMs correspond to the combined experimental resolution of 3.5 meV, supporting the notion of phonon-assisted recombination. The inhomogeneous broadening of the excited state transitions for a given ground state transition energy is much larger due to shape variations of the self-organized QDs [18]. The 32.3 and 35.7 meV modes have been previously identified with the QD LO phonon and an interface mode [17,18,28].

The observation of phonon-assisted optical transitions allows deriving the Huang-Rhys parameter S from the intensity ratio of the first LO replica and the ZPL [3,7]. We find from phonon-assisted emission $S = 0.012$, 0.020, and 0.014, respectively, for the 3ML, PIG, and VDA samples. These values are ∼5 times larger than in bulk InAs ($S_{\mathrm{bulk}} \sim 0.0033$) [29]. Thus, exciton localization in self-organized InAs/GaAs QDs *enhances* the polar exciton-LO-phonon interaction. The coupling strength depends on the different polaron clouds induced by the electron and hole and, therefore, is very sensitive to the actual wave functions. In the following, we estimate the polar exciton-LO-phonon coupling based on wave functions calculated in the 8 band $\mathbf{k} \cdot \mathbf{p}$ model including Coulomb interaction in the Hartree approximation for pyramidal InAs/GaAs QDs with {101} side facets as described in detail in Ref. [16]. The insets of Fig. 4(a) depict probability density isosurfaces of the electron and hole ground states for base lengths of 10.2 and 20.4 nm, respectively. The electron wave function is predominantly s-like, whereas the hole wave function is partly p-like and, with increasing pyramid size, concentrates in the $[1\,\bar{1}\,0]$ corners as a result of the piezoelectric potential induced by the shear strains. Figure 4(a) demonstrates reasonable agreement between calculated (open diamonds) and experimental (solid symbols) exciton transition energies.

The Huang-Rhys parameter S is estimated for the interaction of the ground state exciton with bulk phonon modes, following the calculations of Nomura and Kobayashi [Eq. (8) in Ref. [5]]. The results are depicted as open diamonds in Fig. 4(b). S increases from 0.0020 to 0.027 as the pyramid base length is doubled from 10.2 to 20.4 nm. The enhancement of the exciton-LO-phonon coupling

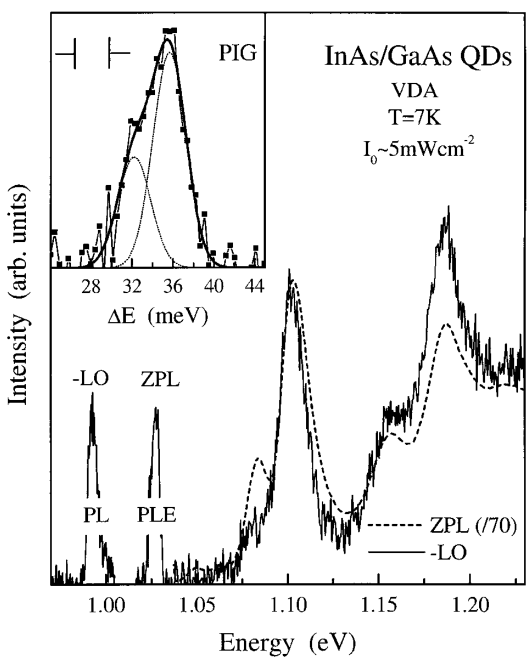

FIG. 3. PL and PLE spectra for the VDA sample. PL was excited resonantly in the ZPL, and PLE was measured for the ZPL (1.0271 eV, dashed line) and the −LO phonon replica (0.9929 eV, solid line). The inset depicts a PL spectrum for the PIG sample excited resonantly in the ZPL at 1.0622 eV together with Gaussian fits.

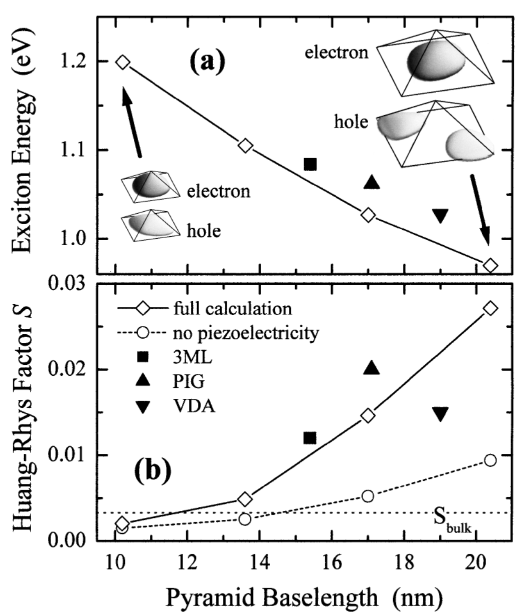

FIG. 4. Experimental (solid symbols) and calculated (open symbols) transition energies (a) and Huang-Rhys parameters S (b) for the ground state exciton in pyramidal InAs/GaAs QDs. The insets of (a) depict probability density isosurfaces ($p = 65\%$) of the electron and hole ground states for $b = 10.2$ nm and $b = 20.4$ nm.

with increasing QD size is attributed to the particular quantum confinement and the piezoelectric potential in the pyramidal InAs/GaAs QDs. Neglecting the piezoelectric potential results in an ~70% reduction of the Huang-Rhys parameter S [open dots in Fig. 4(b)]. The reasonable agreement between experimental (solid symbols) and calculated S values indicates that the adiabatic approximation is able to explain the polar exciton-LO-phonon coupling in coherent InAs/GaAs QDs. The apparent lack of a clear size dependence in the experimental data (solid symbols) results from the fact that the VDA sample contains asymmetric QD pairs instead of single QDs. Calculations for such asymmetric QD pairs show that the strain interaction between the small seed island and the optically active larger QD reduces the piezoelectric potential, causing a negligible blueshift (some meV) of the ground state transition energy but a significant lowering (~40%) of the Huang-Rhys parameter S.

In conclusion, we have identified phonon-assisted exciton recombination in self-organized InAs/GaAs QDs and extracted Huang-Rhys parameters S of ~0.015 for the coupling to the QD LO phonon and an interface mode. Numerical calculations based on realistic electron and hole wave functions demonstrate that Fröhlich-type exciton-LO-phonon interaction, treated in the adiabatic approximation, is consistent with the experimental results. The particular confinement and piezoelectricity in such strained low-symmetry QDs causes the unexpected enhancement of the coupling strength as compared to the InAs bulk case. The presented results support the validity of the adiabatic approximation for the zero-dimensional III-V QDs. The stronger Fröhlich coupling in the II-VI QDs might, however, increase nonadiabatic effects.

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