Phonons in InAs/AlAs single quantum dots observed by optical emission

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Photoluminescence emission spectra of self-assembled single InAs/AlAs quantum dots with variable size reveal typical sharp lines below 1.8 eV, corresponding to transitions between fully confined states. Above this energy the sharp peaks disappear due to the Γ-X scattering of conduction electrons. Excitation spectra of the biexciton line in single quantum dots show marked maxima at the optical-phonon frequency of the quantum dot and at its first overtone. The quantum dot phonon energy observed is 31 meV. Our results indicate an efficient polaron coupling in these quantum dots.

I. INTRODUCTION

Optical emission by single InAs self-assembled quantum dots (QDs) has been extensively studied in the past 20 years[1–11] to get insight into their fundamental properties in the absence of inhomogeneous broadening. Carrier confinement in three dimensions, which gives rise to sharp emission lines, and strong polaronic coupling,[12–17] make QD optical properties very different from higher dimensional systems. They also open the possibility for applications as lasers or single-photon sources.[18] Strong polaronic coupling of the electronic states to optical phonons has been demonstrated to be the origin of carrier energy relaxation in QDs, even for large detuning of the electronic intersublevels from the phonon energy.[16] The otherwise everlasting polaron[13] decays by anharmonic coupling to bulk acoustic phonons.[19,20] Most of the work published so far involves InAs QDs embedded in GaAs barriers. In these systems, phonons of the InAs or InGaAs QDs are close in energy to the GaAs-LO one, because of strain and intermixing. Therefore, interaction effects are expected between QD phonons and barrier ones, which can affect the polaron formation and its decay time. Indeed, phonons associated with InAs QDs, its interface, the wetting layer, and with the GaAs barrier have been reported in photoluminescence excitation[14,15,21] and Raman scattering[22,23] experiments in QDs as well as in calculations.[17] This situation may be changed by using a higher gap barrier as AlGaAs (Ref. 8) or AlAs.[24,25] Higher barriers have several advantages, as an increase of the carrier confinement, bringing the QD emission to higher energies. Also, the use of a harder barrier material as AlAs suppresses coupling of the QD optical phonon to the barrier optical phonons and can result in a slower polaron decay. Extremely long decay times (microseconds) in ensemble-averaged InAs/AlAs QDs have been reported[24,26] for high-energy excitation. They have been attributed to spatial separation of electrons and holes in neighboring QDs caused by scattering of InAs Γ-band electrons into the AlAs X band. In this paper we present photoluminescence (PL) and PL excitation (PLE) intradot measurements on a single InAs/AlAs QD. Our results show a transition at 1.8 eV from sharp PL lines associated with totally confined states in the QD to a broader emission above 1.8 eV as a consequence of the crossover of the Γ band of the QD with the X band of the barrier. The PLE spectra of the exciton and biexciton lines of a single QD display strong resonances at energies multiple of 31 meV. This energy corresponds to the optical phonon confined in the quantum dot. The resonances are particularly strong for the biexciton emission and are interpreted in terms of the polaron formation in the QD.

II. EXPERIMENT

The InAs/AlAs QD samples were grown by molecular-beam epitaxy (MBE) on GaAs (100) substrates as described in Ref. 25. The active region consists of InAs dots embedded in the center of an altogether 40-nm-thick AlAs barrier. Finally, the sample was capped by a 10-nm GaAs layer. Sample rotation was interrupted during QD deposition resulting in an InAs coverage gradient across the wafer. The coverage varied between 1.6 and 2.0 monolayers, corresponding to different dot sizes and densities, as measured by atomic force microscopy.[25] Typical values used in this work are QD densities of 10^{10} cm^{-2}, dot diameters between 25 and 30 nm, and dot heights around 3 nm. The samples were covered by an aluminium mask containing different square apertures fabricated by electron-beam lithography. The aperture sizes range from 10 to 0.2 μm, allowing for detection of single QD luminescence. The PL emission was excited with either an argon-ion or a Ti-sapphire laser with a typical power density on the sample of 10^4 W/cm^{-2}. This rather large excitation power was necessary to compensate the low light intensity collected from the dots. The emitted light was collected with a microscope setup using a long working distance objective ($50\times$) and a double-grating spectrometer with a
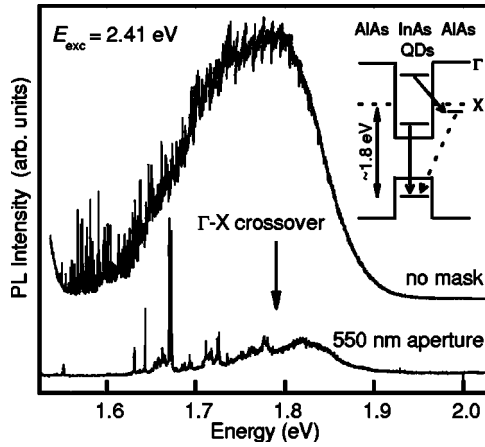
FIG. 1. PL with and without mask. PL from QDs is observed only below 1.8 eV as a consequence of the Γ-X crossover with the AlAs barrier. Inset: Band sketch of the active region illustrating transitions between fully confined QD states and recombination of electrons scattered into the AlAs X band.

charge-coupled device detector. The excitation spot size was 5 μm. The sample was cooled in a continuous flow He cryostat for microscope application. All the measurements presented were taken at 8 K.

III. RESULTS AND DISCUSSION

The PL spectra of the dots excited at 514.5 nm are presented in Fig. 1 for a large ensemble of QDs taken outside the metal mask (top curve) and for a small number of QDs taken at one of the 550-nm apertures in the mask (bottom curve). One observes sharp peaks corresponding to totally confined states in individual QDs only below 1.8 eV, even in the spectrum taken outside the mask. Spectrum (a) contains the emission of around one thousand QDs. The fact that sharp structures are observed in its low-energy tail reveals that PL from individual QDs can be detected even without the metal mask. The emission from the wetting layer (not shown) is at 2 eV. The inset in Fig. 1 shows a sketch of the band alignment and the PL transitions for large dots (solid arrow), where the confined state is below the conduction-band minimum of the AlAs barrier (at the X point of the Brillouin zone). In smaller dots, the higher confinement energy brings the state above the X minimum and consequently, the electron is no longer confined inside the dot. Recombination in this case involves a dispersive electron state and no sharp peaks are visible (dashed arrow). The crossover energy of 1.8 eV is determined by the strain in the InAs dot, the valence-band offset between AlAs and strained InAs, the confinement energy of the holes, and possibly some diffusion of Al into the dot.

The emission spectra of a single QD are shown in Fig. 2 for different excitation power P. The enlargement factors are indicated. For excitation power below 1 mW the intensities of the peaks labeled X and XX grow as P^α with $\alpha=0.5$ and 1.5, respectively. The expected exponents for the exciton (X) and biexciton (XX) lines at low excitation are $\alpha=1$ and 2, respectively. These values decrease at high excitation

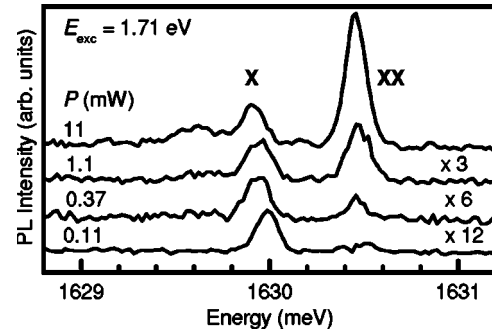

FIG. 2. Power dependence of two peaks in the PL of a single quantum dot, normalized to the X peak intensity. For excitation power below 1 mW the X and XX peaks grow as P^α with $\alpha=0.5$ and 1.5, and are attributed to the exciton and biexciton, respectively (see text).

levels[6,11] and consequently we assign the peaks in Fig. 2 to the exciton and biexciton. The fact that the biexciton appears at higher energy than the exciton might be related with the actual shape of the dots and with the details of the confining potential.[11] The possible formation of charged exciton complexes cannot be completely ruled out. But the use of intra-dot excitation at energies E_{exc} below the Γ-X crossing (at 1.8 eV) strongly reduces their probability.

The intensities of the X and XX emissions change drastically upon changing the excitation energy. This is shown in Fig. 3, where the spectrum of Fig. 2 at 8 mW is presented for excitation energies varying from (a) 1.655 to 1.669 and from (b) 1.685 to 1.699 eV as indicated next to the top and bottom curves, respectively. In the experiment the excitation energy is varied in approximate steps of 1 meV. Besides the small energy fluctuations due to electrostatic changes in the QD neighborhood, one observes strong intensity increases, particularly in the XX peak, for certain excitation energies. This is also clearly seen in Fig. 4, where the PL spectrum of the dots for an intradot excitation energy of 1.71 eV (upper curve) is presented with the PLE spectrum of the XX line. The detection energy is indicated by an arrow. There are two strong and rather broad PLE peaks at 31 and 62 meV, the
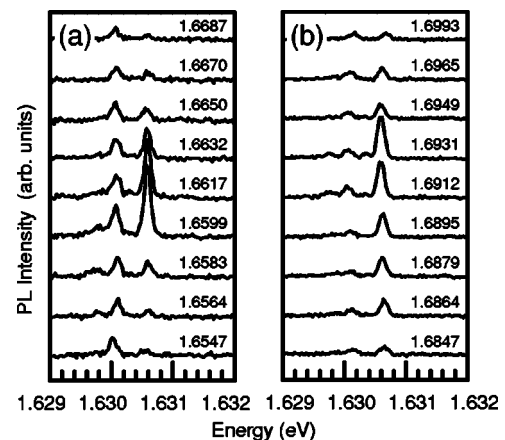

FIG. 3. Waterfall plots of the X and XX emission (see Fig. 2) for varying excitation energy E_{exc} which is indicated in eV. In (a) the 1LO polaron state is excited, in (b) the 2LO polaron state.
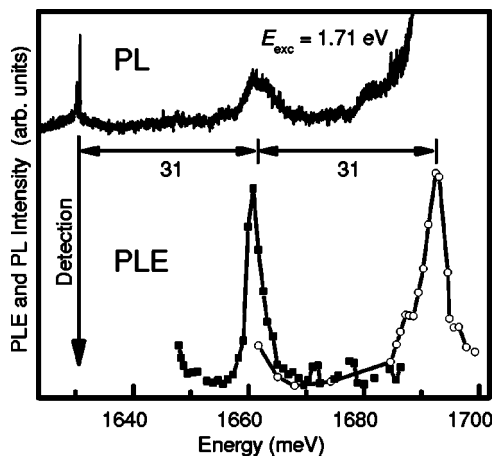
FIG. 4. PL (E_{exc}=1.71 eV) and PLE spectra of the *XX* peak (see Fig. 2). PLE reveals polaron states, separated by the phonon energy of the dot (31 meV), which is close to the bulk InAs LO phonon frequency. Squares and circles correspond to two different experimental runs. At 1LO the polaron state is also observed in PL.

former also observed in the PL spectrum. Narrow PLE lines, which are frequently observed in PLE spectra of InAs/GaAs single QDs,[6,21,27] are missing in our case. Relatively broad (around 1 meV) PLE peaks in the range of 29–31 meV have been reported in InGaAs QDs (Ref. 21) and attributed to exciton-LO phonon coupling in the dots. Also ensemble-averaged PL (Refs. 14 and 15) and Raman[22,23] measurements indicate phonon energies in the same energy range. We therefore ascribe the PLE peaks in Fig. 4 to transitions involving polaronic states due to the coupling between the QD electronic states and the phonon. The QD phonon energy in this case is 31 meV, i.e., 1 meV above the bulk InAs LO energy. The difference can be roughly accounted for assuming full hydrostatic deformation of the QD to match the AlAs lattice parameter. Confinement is expected to have a moderate effect on the phonon energy in our QD due to the slow dispersion of the InAs LO phonon at low wave vector. A rough estimate considering the dot height of 3 nm gives an energy decrease of the order of 1 meV. Consequently, the effect of strain is probably stronger than our previous simple estimate considering only hydrostatic deformation. Aluminium diffusion into the QD would also result in an increase of the phonon frequency. In the present case this effect is negligible due to the low growth temperature. In fact, 5% of Al in an unstrained QD would produce an energy increase of the phonon energy in this range. The stronger resonance of the *XX* peak probably indicates that the energy of the electronic biexciton state is closer to the phonon one. It is worth noting that there is no Stokes shift between PL and PLE lines at

1.662 eV, as expected from the absence of inhomogeneous broadening in the spectrum of a single dot. Consequently, the width of the PLE peaks seems to be intrinsic. Part of it can be due to different phonon contributions as the phonon of the wetting layer, which has been reported at 29.6 meV in InAs/GaAs QDs,[14] or interface phonons.[23] Also, inhomogeneity in the strain of the QD can contribute to the broadening. It is difficult to quantify the different contributions of the broadening, so that we cannot estimate the true polaron decay time from the width of the PLE peaks. PLE spectra of smaller dots emitting at higher energies are difficult to interpret due to the overlap with excited-state transitions in larger dots in the same spectral region. The energy loss in the QD occurs through the coupling of the phonon part of the polaron to optical or acoustical barrier phonons.[13,19,20] This coupling is favored in InAs/GaAs QDs by the fact that the QD phonon energy is very close to the GaAs LO bulk value. Energy can be then easily driven out of the dot via phonon-phonon interaction. In our case, the energies of the optical phonons of the barrier (45–50 meV) are too high to couple to the QD polaron. This can result in an enhancement of the polaron lifetime in InAs/AlAs QDs. An alternative possible decay path of the polaron is the emission of two transversal acoustic AlAs phonons (X, K), whose energy (15 meV) (Ref. 28) is close to one half of the QD one. A marked peak in the density of states for acoustic phonons in AlAs at this energy[28] gives support to this mechanism. However, the coupling to acoustical phonons is comparatively weak, and it might not result in an efficient polaron decay. Time-resolved PL measurements on single QDs with intradot excitation would be needed to decide on the polaron decay time of our system.

In summary, we present emission and excitation spectra on single InAs/AlAs quantum dots, which reveal an energy threshold at 1.8 eV for sharp emission lines between fully confined states. Above this energy, Γ-X scattering of the electron results in a continuous emission spectrum. The PLE spectra of a single QD under intradot excitation presents strong peaks at multiples of 31 meV, which are interpreted as the result of the polaronic coupling of the electronic states in the dot to the QD phonon. This coupling appears to be stronger for the biexciton emission line.

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