Efficient acoustic phonon broadening in single self-assembled InAs/GaAs quantum dots

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(Received 16 July 2001; published 28 December 2001)

We report systematic temperature-dependent measurements of photoluminescence excitation spectra in single self-assembled InAs/GaAs quantum dots. We studied the increase with temperature of the excited-state homogeneous linewidth for different quantum dots. We found a correlation between the acoustic phonon broadening efficiency and the background intensity in the photoluminescence excitation spectra. These results demonstrate the interaction of the discrete quantum dot excited states with a quasicontinuum of states and impose severe limitations on the isolated artificial macroatom scheme for a single quantum dot.

DOI: 10.1103/PhysRevB.65.033313 PACS number(s): 78.66.Fd, 78.55.Cr, 78.67.Hc

The interaction between the lattice vibration modes and the electronic states in a semiconductor quantum dot (QD) is an extremely debated issue in nanostructure research.[1–10] Since three-dimensional (3D) carrier confinement leads to a discrete energy spectrum for a semiconductor QD, the coupling to acoustic and optical phonons is expected to be less efficient in a QD than in a semiconductor quantum well (QW). In this latter heterostructure the translational invariance in the QW plane gives rise to a continuum of states providing a large density of states for phonon absorption or emission. Because of suppression of this continuum in a semiconductor QD, a bottleneck effect has been predicted for phonon-assisted relaxation in QD's. For acoustic phonons it has been shown that the relaxation is totally inefficient because the energy mismatch between electronic states is larger than the mean acoustic phonon energy.[1,2] For optical phonons it has been argued that the relaxation could be efficient only if the energy distance between electronic states matches one (or several) optical phonons. As far as applications in high-performance semiconductor devices are considered, the slowing down of the electronic relaxation is a central issue for the development of semiconductor-QD-based detectors and lasers.[11,12]

In fact all these considerations rely on the assumption that the carrier-phonon interaction is in a weak-coupling regime where the interaction is treated in the framework of Fermi's golden rule. However, in addition to the discretization of the energy spectrum in a semiconductor QD, the coupling to phonon may be influenced by the 3D carrier localization. Indeed, upon the optical transition, the lattice configuration locally changes according to the different electronic states in the QD. The degrees of freedom for the lattice and electron may no longer be treated separately. Evidence of a strong-coupling regime has recently been reported by infrared magnetotransmission experiments where the signature of mixed optical phonon-electron states, the so-called polarons, was found for intraband transitions in self-assembled InAs/GaAs QD's.[6] For interband transitions there is no experimental evidence of polaron states in InAs/GaAs QD's but rather an enhanced coupling to optical phonons.[5,7] As far as acoustic phonons are concerned a nonperturbative interaction may lead to the appearance of low-energy acoustic phonons sidebands in the line profile.[13] A recent observation of this lattice relaxation effect has been reported in QD's formed by interface defects in II-VI CdTe QW's[14] but there is no evidence of a deviation from a Lorentzian profile in InAs/GaAs QD's (Ref. 9, 10, and 15) and to the best of our knowledge no quantitative data on the acoustic phonon coupling efficiency in this system.

In this paper, we report efficient acoustic phonon broadening in a weak-coupling regime for the excited states of single InAs/GaAs QD's. We performed temperature-dependent measurements of the excited-state linewidth by photoluminescence excitation spectroscopy (PLE). We find a correlation between the acoustic phonon broadening efficiency and the intensity of the background measured in the PLE spectra. Such a quasicontinuum of states was observed by different authors.[4,7,10] We recently reported photoluminescence (PL) up-conversion in InAs/GaAs QD's where we showed that this quasicontinuum is a wetting layer (WL) band tail and provides intermediate states for the up-conversion process.[16] We show here that this continuum allows efficient coupling to acoustic phonons with an efficiency greater than for InGaAs/GaAs QW's. As a matter of fact, the WL band tail provides a large reservoir of states for acoustic phonon scattering and this explains the efficient acoustic phonon broadening for the QD's.

Our sample was grown on a GaAs substrate by molecular beam epitaxy. Rotation of the sample was stopped during the InAs deposition step in order to obtain a variable concentration of InAs islands. The 20-sec deposition of the 1.7-monolayer-thick InAs layer at 530°C was followed by a 7-sec growth interruption under As vapor in order to increase the size of the QD's and get excited-state transitions. The cover layer is made of a 50-nm GaAs layer. In order to isolate single QD's, a mesa pattern was finally designed by *e*-beam lithography and reactive ion etching.

Microphotoluminescence measurements were performed in the far field using a microscope objective (numerical aperture 0.5) in a confocal geometry. The excitation beam, provided by a tunable cw Ti:sapphire laser, was focused on the sample with a spot size ~ 1 μm, accurately positioned using $X-Y$ piezoelectric stages moving the microscope objective with a precision ± 0.05 μm. The QD sample was mounted

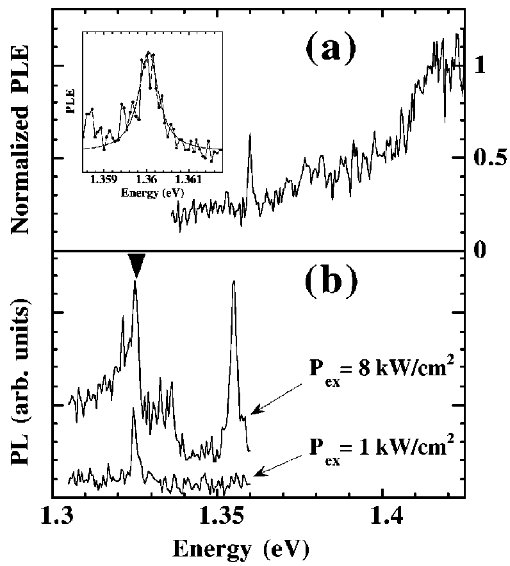

FIG. 1. (a) Normalized microPLE spectrum recorded at 10 K for a detection energy of 1.325 eV and a power density P_{ex} of 6 kW/cm^2. The inset is a high-resolution scan around 1.36 eV showing the excited-state line together with a Lorentzian fit ($\Gamma = 0.6$ meV). (b) Corresponding microPL spectra for an excitation energy of 1.44 eV and two excitation power densities P_{ex}. The arrow in the PL spectra indicates the detection energy for the PLE.

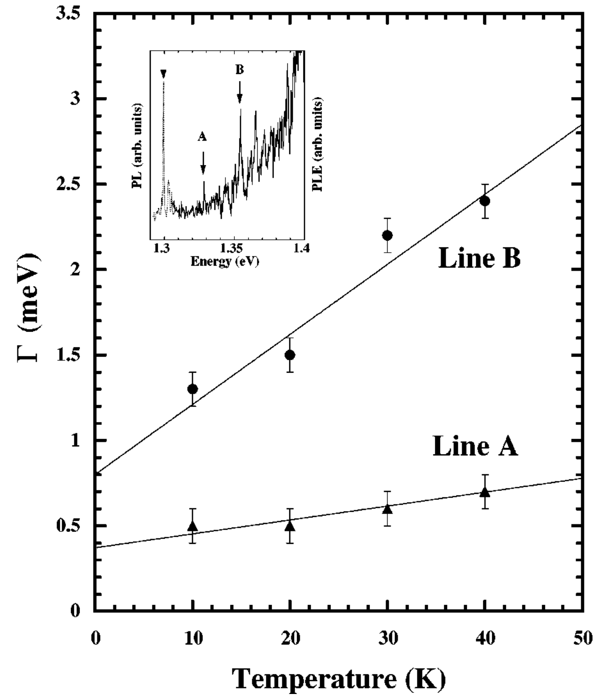

FIG. 2. Full width at half maximum Γ of the lines A and B (indicated by arrows in the inset) vs temperature. The slopes of the linear fits are $10 \pm 2\,\mu$eV/K for line A and $41 \pm 2\,\mu$eV/K for line B. The inset shows the microPL (dashed line) and microPLE (solid line) of the investigated QD.

on the cold finger of a continuous-flow helium cryostat. The photoluminescence signal (collected in our setup by the same microscope objective as for the excitation) was detected by a low-noise Si-based photon counting module after spectral filtering by a 32-cm monochromator. With this setup we obtain a resolution of 1 meV in PL spectra (determined by the response function of the monochromator) and a resolution of 0.07 meV in PLE spectra (determined by the laser linewidth).

A typical low-temperature PLE spectrum is shown in Fig. 1(a). The corresponding PL spectrum for an excitation at 1.44 eV is displayed in Fig. 1(b) (bottom curve). We observe a sharp line at 1.325 eV corresponding to the fundamental transition of a single QD. The PLE spectrum has been performed by recording the PL emission of this line (indicated by the arrow). In the PLE spectrum [Fig. 1(a)] we first observe a background gradually increasing up to the WL absorption edge (1.42 eV). On top of this broad signal finer structures are observable and in particular a sharp line at 1.36 eV. This peak could correspond to an excited-state of the single QD under investigation. Assuming a truncated pyramid for the QD shape theoretical calculations predict 18 nm and 1.1 nm for the radius and height of this QD.

In order to check that this resonance in the PLE signal is the signature of a QD excited-state we performed high-excitation PL. By increasing the excitation power density we may saturate the fundamental transition and observe recombination from excited-state transitions. In Fig. 1(b) we show PL spectra on a linear scale for two values of the excitation power density P_{ex}. By increasing P_{ex} by a factor 8 (top curve), one clearly sees that the QD fundamental transition at 1.325 eV saturates (on top of a broad background) while electron-hole recombination also occurs at higher energy (1.355 eV). From the comparison between the PLE spectrum of Fig. 1(a) and the high-excitation PL spectrum of Fig. 1(b) (top curve), we identify the peak at 1.36 eV as an excited-state of the single QD. The 5-meV redshift of this line between high-excitation PL and PLE is attributed to Coulomb effects that take place when the fundamental and excited-states are populated under high-excitation of the QD.[17]

In the inset of Fig. 1, we show a high-resolution scan of the PLE spectrum around the excited-state transition. Note that the excited-state line can be fitted by a Lorentzian profile on top of a background. In fact, because we perform micro-PL measurements to isolate single QD's, we resolve homogeneous lines with Lorentzian profiles and we thus attribute their full width at half maximum (FWHM) to the homogeneous linewidth of the excited state under investigation.

We studied the phonon-assisted broadening by temperature-dependent measurements of the PLE spectra up to 50–60 K, for a power density of 6 kW/cm^2, in QD's of various sizes. We observed an increase with temperature of the FWHM of the excited-state lines which remain Lorentzian in the temperature range we investigated. The absence of low-energy sidebands in the spectral profile means that there is no signature of a nonperturbative regime for the interaction between the acoustic phonon and electron-hole pair in our InAs/GaAs QD's.

In Fig. 2 we display typical temperature variations of the homogeneous linewidth Γ of QD excited-states. These measurements refer to a QD different from the one shown in Fig.

1. The corresponding PL (dashed line) and PLE (solid line) spectra are displayed in the inset of Fig. 2. The fundamental transition of this QD is at 1.3 eV and the peaks at 1.328 eV and 1.355 eV (labeled A and B and indicated by arrows) correspond to two excited-states. The important point to be noticed here is that line B is lying at higher energy where the PLE background is larger (roughly 2 times) than for line A. For lines A and B we observe a linear increase of their homogeneous linewidth with temperature. However, the slope is greater for line B (41 ± 2 μeV/K) than for line A (10 ± 2 μeV/K).

The phonon-assisted broadening has been extensively studied for semiconductor bulk and QW's. The homogeneous linewidth Γ(T) of excitons is usually described as the sum of a static broadening Γ_0 including radiative broadening and a temperature-dependent one accounting for acoustic and optical phonon broadening.[18] For low temperature ($T \leqslant 60-70$ K) where the interaction with acoustic phonons is dominant, $\Gamma(T)$ increases linearly with temperature and the homogeneous linewidth can be written

$$\Gamma(T)=\Gamma_0+aT. \tag{1}$$

For $In_xGa_{1-x}As/GaAs$ QW's typical values for the acoustic phonon broadening efficiency *a* of the excitonic ground state range between 1.5 and 11 μeV/K.[19–21]

In QW's the acoustic phonon broadening efficiency *a* characterizes the scattering of excitons into a continuum of final excitonic states with large in-plane wave vectors. In semiconductor microcavities measurements of *a* have shown that the strong exciton-photon coupling regime modifies the density of final states accessible for acoustic phonon scattering.[21]

In our single InAs/GaAs QD's, despite the fact that a phonon bottleneck had been early predicted for acoustic phonon scattering in semiconductor QD's, we do observe a linear increase with temperature of the excited-state linewidth. We therefore extract the acoustic phonon broadening efficiency *a* in our self-assembled InAs/GaAs QD's and in the following we show that we can correlate the obtained values with a density of final electronic states.

The existence of a quasicontinuum of states is of course suggested by the background signal observed in PLE and recently reported by different authors.[4,7,10] In order to compare our different values of the acoustic phonon broadening efficiency *a* measured for a set of 13 QD excited states, we would like to get QD-independent values of the PLE background signal. In fact, for a given excitation energy fluctuations of the PLE background from dot to dot may arise from different spectral profiles of the background and/or from different radiative efficiencies. In order to correlate our *a* values with the PLE background signal we normalize the PLE spectra with the PLE signal at 1.42 eV, i.e., at the WL absorption edge where the PLE spectrum reaches its maximum for each QD investigated. For instance, in Fig. 1(a), the normalized PLE signal is 1 at 1.42 eV and 0.23 ± 0.02 at the QD excited-state energy (1.36 eV).

In Fig. 3 we show the acoustic phonon broadening efficiency *a* for 13 excited-states coming from 9 different QD's.

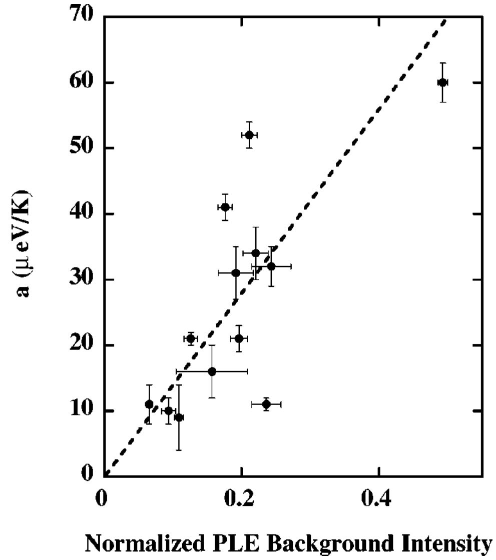

FIG. 3. Scatter plot of the acoustic phonon broadening efficiency *a* for 13 excited-states (9 different QD's) vs normalized PLE background intensity at the energy of each excited-state. The dashed line is a guide for the eyes showing the increase of the acoustic phonon broadening efficiency with the PLE background.

We plot these values as a function of the normalized PLE background intensity at the energy of the corresponding QD excited-state. We clearly see the increase of the acoustic phonon broadening efficiency with the normalized background intensity. Before discussing further this correlation and explaining the underlying physics, we want to point out that our *a* values are always greater than the values found in the literature for $In_xGa_{1-x}As/GaAs$ QW's ($1.5\leqslant a \leqslant 11$ μeV/K).[19–21] Of course these parameters refer to the fundamental 1*S* exciton state in QW's and not to an excitonic excited-state but it is very striking to see that an extremely efficient coupling to acoustic phonons exists for QD excited states. Note that this behavior is in contrast to the measurements reported in Ref. 14 for the acoustic phonon broadening efficiency of the ground-state transition of CdTe QD's where a smaller value is measured than for CdTe/CdZnTe QW's.

Because we do not see any experimental evidence of a nonperturbative regime in the spectral profile of the excited states, one shall describe the interaction with acoustic phonons in the framework of Fermi's golden rule. Therefore, the probability per unit time of emitting or absorbing acoustic phonons increases linearly with the density of states describing the quasicontinuum to which the electron-hole pair is coupled. On the other hand, if no resonance occurs during population relaxation, the background observed in the PLE spectrum is a direct measurement of the optical density which is proportional to the quasicontinuum density of states. In consequence, we easily understand the average increase of the acoustic phonon broadening efficiency *a* with the normalized PLE background intensity. In this model it reflects the efficient coupling of the QD's excited states to the quasicontinuum observed in single QD PLE spectra. The acoustic-phonon-assisted interaction with the localized states of the WL quasicontinuum appears to be, at least, as efficient as the broadening of the excitonic ground state in a QW. Obviously the physics is completely different in the case of localized states than in the case of delocalized states. In particular the lack of wave vector conservation in the scattering processes involving the 0D QD states may play a key role. This effect is a direct proof of the interaction of a semiconductor QD with its environment and highlights a severe limitation to the isolated macroatom scheme.

As far as the fluctuations of the acoustic phonon broadening efficiency *a* are concerned, we see two reasons for this. The coupling efficiency first depends on the size of the QD's. Similarly to the QW case, the carrier–acoustic-phonon interaction presents a resonance when the wavelength of the acoustic phonon is comparable to the confinement potential size.[19] Therefore size fluctuations may induce fluctuations of the acoustic phonon broadening efficiency. Moreover, the coupling efficiency depends on the exact density of states at the excited-state energy. Fluctuations of the PLE background signal around its mean value appear as a signature of the detailed density of states in the WL band tail. In fact the WL is far from being an ideal 2D QW and carrier confinement is induced by the strong spatial variations of the WL thickness.[22] From one QD to another, the WL roughness in the vicinity of the QD is for sure different. Therefore the density of localized states giving rise to the WL band tail observed as a background in the PLE may change from dot to dot. Consequently, for the same normalized background intensity the acoustic phonon broadening efficiency may fluctuate.

In conclusion, we bring clear evidence of efficient acoustic phonon broadening for the excited states of single InAs/GaAs QD's. We find a correlation between the coupling efficiency and the background intensity observed in the PLE spectrum. We explain this effect by emission and absorption of acoustic phonons from the excited state to a quasicontinuum coming from the WL band tail. These measurements point out a limitation of the isolated macroatom scheme often used to describe a semiconductor QD. Indeed, we show that the WL band tail provides a continuum of final electronic states that plays a role similar to the reservoir of large wave vector states for the excitonic ground-state broadening in 2D QW's. Our results demonstrate that the presence of a quasicontinuum in the PLE completely breaks down the picture of an inhibited acoustic phonon interaction for carriers confined in semiconductor QD's. For future applications in semiconductor-QD-based devices, one shall try to suppress (at least partially) the existence of the WL band tail in order to slow down the coherence relaxation by acoustic phonon scattering.

We gratefully acknowledge G. Bastard and R. Ferreira for helpful discussion and O. Verzelen for theoretical calculations. LPMC de l'ENS is "Unité Mixte de Recherche Associée au CNRS (UMR 8551) et aux Universités Paris 6 et 7." This work was partly supported by a New Energy and industrial technology Development Organization (NEDO) grant.

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