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Line narrowing in single semiconductor quantum dots: Toward the control of environment effects

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We report systematic linewidth measurements on the fundamental transition of single InGaAs quantum dots. We demonstrate the quenching of the acoustic-phonon dephasing for quantum dots spectrally well separated from the band tail of the wetting layer. We achieve a line narrowing with linewidth of the order of few μeV by tailoring the influence of the electrostatic environment through a decrease of the excess energy in the nonresonant excitation process.

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Semiconductor quantum dots (QD's) are artificial nanostructures where the three-dimensional quantum confinement leads to a discretization of the energy spectrum. Thus, an inhibition of the inelastic- and elastic-scattering processes is predicted for carriers in QD's because of the suppression of the continuum of final electronic states.[1,2] The expected long decoherence time (T_2) and reciprocally narrow linewidth Γ ($\Gamma = 2\hbar/T_2$) of the discrete QD levels make semiconductor QD's very promising for cavity quantum electrodynamics experiments in the solid state[3–5] and also for some challenging applications in quantum information processing.[6–9]

In these prospects self-assembled InGaAs QD's are one of the main focal points in nanostructure research.[10] Recently, time-resolved measurements on a large *ensemble* of QD's have given experimental evidence of a spin-relaxation quenching[11] and long decoherence[12,13] at low temperature under resonant excitation of the QD fundamental interband transition. With a radiative lifetime T_1 of the order of one nanosecond in InGaAs QD's,[3,4] the regime of a decoherence time limited only by the radiative lifetime ($T_2 = 2T_1$) appears accessible at low temperature for the fundamental transition of these nanostructures. However, quantum information devices demand coherent manipulation of *single* qubits. An experimental access to the fundamental transition decoherence in *isolated* QD's is still missing and appears as a crucial task before implementation of quantum logic within the decoherence time.[7] In fact, one of the limitations of QD-based devices arises from the interaction of a QD with its environment, which may differently influence the decoherence from QD to QD.

In this context, the study of single QD's by means of space-resolved techniques is highly desirable and the development of near-field and confocal microscopy has allowed optical spectroscopy in isolated QD's.[14–17] In this case, the resonant excitation of a single QD produces an optical response completely dominated by laser light scattering from the QD surrounding. Consequently, investigation of the fundamental transition decoherence in a single QD is more easily performed under nonresonant excitation. Although homogeneous linewidth measurements in photoluminescence (PL) spectroscopy under cw excitation appear as a potential alternative to time-resolved techniques on an ensemble of QD's, the spectral resolution achieved so far was not sufficient to characterize the fundamental transition decoherence in single QD's.[14–17]

In this paper, we report linewidth measurements on the fundamental interband transition of single InGaAs QD's. By inserting a Michelson interferometer in the detection path, we apply Fourier-transform (FT) spectroscopy on the PL signal of a single QD to perform systematic measurements of the fundamental transition linewidth on QD's from two different samples. With our compact and flexible technique, we combine PL, photoluminescence excitation (PLE), and FT spectroscopy to study the temperature-induced dephasing and the influence of the electrostatic environment. We show that the band tail of the wetting layer (WL) (Refs. 16 and 18) highly determines the fundamental transition linewidth. For QD's spectrally well separated from the WL band tail, we demonstrate the quenching of the acoustic-phonon dephasing. Furthermore, we achieve a line narrowing with linewidth down to 7.5 μeV by tailoring the influence of the electrostatic environment through a decrease of the excess energy in the nonresonant excitation process.

We have studied two InGaAs QD samples grown in the Stranski-Krastanov mode in separate molecular-beam epitaxy machines. Self-assembled QD's are obtained on top of a quasi-two-dimensional InGaAs WL. The samples present an areal QD density of 10^{10}–10^{11} per cm^2 and they are processed on the surface with sub-μm mesa patterns in order to isolate single QD's.[14] Single QD spectroscopy is performed at low temperature with the confocal setup described in Ref. 18 where the excitation is provided by a tunable cw Ti:sapphire laser.

PL spectra recorded at 10 K in unprocessed regions of the samples show a broad line corresponding to electron-hole recombination in many QD's of various sizes. The inhomogeneous linewidth is 30 meV in sample A and 60 meV in sample B because of larger size fluctuations in the second sample. In *both samples* the PL line is centered at 1.32 eV. However, PLE spectroscopy reveals two distinct regimes in the three-dimensional (3D) quantum confinement, as discussed below.

In Fig. 1 we show typical PL and PLE spectra recorded in single QD's of sample A [Fig. 1(a)] and B [Fig. 1(b)]. Each

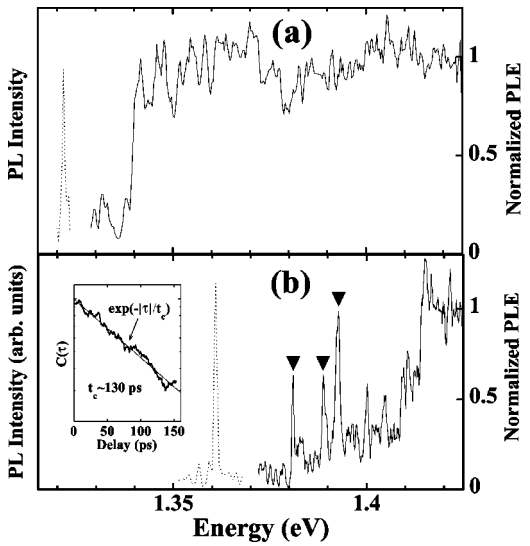

FIG. 1. (a) and (b) Microphotoluminescence (dashed line) and microphotoluminescence (solid line) spectra recorded at 10 K for a single QD of sample A (a) and sample B (b). (b) The arrows indicate QD excited states. Inset (semilog scale): Interferogram contrast $C(\tau)$ decaying exponentially with a time constant of 130 ps for the single QD in (b), excited at 1.38 eV.

microphotoluminescence spectrum (dashed line) shows a sharp line [at 1.32 eV in (a) and 1.36 eV in (b)] corresponding to the PL emission of a single QD. For sample A (a) the PLE signal presents an increase at 1.34 eV corresponding to the WL band edge. At higher energies the constant value is consistent with the expected flat absorption of the quasi-two-dimensional quantum well (QW) formed by the WL. For sample B (b) the WL band edge is at 1.42 eV and the fine structures observable at lower energies (marked by arrows) correspond to QD excited states.[19] The difference between the WL band-edge energy of the two samples arises from various WL thicknesses (3 ML in sample A and 1.7 in sample B) due to the different nature of the WL, namely, $In_{0.6}Ga_{0.4}As$ for sample A and InAs for sample B. With a QD distribution centered at 1.32 eV for both samples, this leads to an average excitonic confinement energy of 100 meV in the QD's of sample B and only 20 meV in sample A.

Both PLE spectra present a background gradually increasing up to the WL absorption edge and corresponding to a WL band tail.[18] The energy difference between the QD and the WL is larger in sample B and the WL band tail is spectrally more extended than in sample A. We will see in the following that the relevant parameter for the fundamental transition broadening is the spectral overlap of the QD fundamental transition with the WL band tail.

To perform high-resolution linewidth measurements on the QD fundamental transition we insert a Michelson interferometer before the grating spectrometer to apply FT spectroscopy[20] on the PL signal of a single QD. With a translation stage we vary the time delay τ for propagation in one arm of the interferometer and we record interferograms of the PL emission $I(\tau)=I_0[1+C(\tau)\cos(E_0\tau/\hbar)]$ where I_0 is the average PL signal intensity, E_0 the central detection energy, and $C(\tau)$ the interference contrast given by the modulus of the FT of the PL intensity spectrum transmitted by the spectrometer. The spectrometer improves the signal-to-noise ratio and we take care of opening the exit slit to avoid any spectral filtering of the QD line. In case of a Lorentzian line of full width at half maximum Γ, $C(\tau)$ is given by $\exp(-\Gamma|\tau|/2\hbar)$. The interferogram envelope of the single QD of Fig. 1(b) excited at 1.38 eV is displayed in the inset. On this semilogarithmic plot we observe a linear decrease of the envelope function $C(\tau)$ with a decay time of 130 ps. In the spectral domain this corresponds to a Lorentzian line of width $\Gamma \sim 10$ μeV.

Because we perform measurements in the time domain, we get an extremely high spectral resolution (0.2 μeV in our case) limited only by the length of the translation stage. Such a resolution is orders-of-magnitude below the best resolutions (20–30 μeV) reported with double or triple monochromators.[13,21] As a consequence we get a straight and unambiguous experimental access to the fundamental excitonic state linewidth in single QD's. Moreover, this flexible setup allows a very powerful investigation of single QD optical properties since we combine PL, PLE, and FT spectroscopy in a simple way. By blocking one arm of the interferometer, one records PL and PLE spectra. By leaving both arms free and opening the exit slit, one performs FT spectroscopy for linewidth measurements. Finally, the first remarkable observation is that the linewidth strongly varies from QD to QD. We get this essential information by investigating single QD's which is not the case in previous works on T_2 measurements.[12,13]

An increase of the temperature hastens the dephasing. In semiconductor bulks and 2D QW's,[22] the excitonic linewidth increase as a function of temperature is usually described by $\Gamma(T)=\Gamma_0+aT+b\exp(-\Delta E/kT)$, where Γ_0 is the zero-temperature linewidth, and aT and $b\exp(-\Delta E/kT)$ account for acoustic-phonon and optical-phonon broadening, respectively. We studied the phonon induced dephasing by systematic temperature-dependent measurements of the fundamental transition linewidth on several QD's from the two samples, excited above the WL band edge. Our experiments are performed below saturation where the QD is occupied on average by less than one electron-hole pair. In the temperature range investigated here (5–90 K), we never observed any deviation from an exponential decay for the interferogram envelope. This means that the line profile remains Lorentzian in our InGaAs QD's in contrast to the recent observation of low-energy sidebands in II–VI CdTe single QD's,[23] and in InGaAs QD arrays by photon-echo experiments.[12] Therefore, our temperature-dependent data are fully characterized by the linewidth increase with temperature as shown in Fig. 2(a) for one single QD of each sample. The phonon-induced broadening exhibits a linear variation for $T \leqslant 40$ K followed by an exponential variation at higher temperature.

Let us first focus our attention on the low-temperature part where the linear contribution aT of acoustic-phonon dominates. For four QD's investigated in sample A, we measured an acoustic-phonon broadening efficiency a ranging from 0.12 ± 0.03 to 2.1 ± 0.2 μeV/K. For five QD's in sample B, we found a very small dispersion and the five a

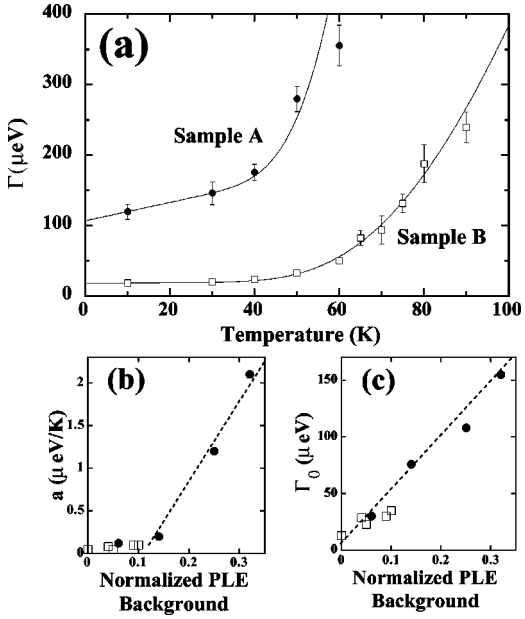

FIG. 2. (a) Fundamental transition linewidth Γ versus temperature for a single QD of sample A (filled circles) and B (squares) both excited above the WL. (b) and (c) Acoustic-phonon broadening efficiency a and zero-temperature linewidth Γ_0 versus normalized PLE background.

values range between 0.05 and 0.1 μeV/K (± 0.03 μeV/K). These latter values are the smallest ever measured in semiconductor heterostructures. They are more than one order-of-magnitude lower than in 2D InGaAs/GaAs QW's,[24–26] and more than two orders-of-magnitude lower than for the QD excited states of the *same sample*.[19] As a matter of fact, our data show the predicted quenching of the acoustic-phonon dephasing in QD's due to the lack of final electronic states for emission and absorption of acoustic-phonons.[1] On the contrary, acoustic-phonon scattering is not necessarily inhibited in sample A and one may invoke a residual quasicontinuum of final electronic states to explain this effect.

As a matter of fact, the WL band tail observed as a broad background in PLE may provide the density of states for quasielastic acoustic-phonon scattering. To check this we need to estimate the corresponding density of states in resonance with the QD fundamental transition. Assuming that no resonance occurs during population relaxation, the PLE background is a direct measurement of the optical density, proportional to the residual density of states. Since it is not possible to perform PLE very close to the QD fundamental transition because of laser light scattering, we tentatively take the PLE intensity 10 meV above the QD fundamental transition as an estimate of the residual density of states due to the WL band tail. Furthermore, to get QD independent values we normalize the PLE background by the PLE intensity at the WL band edge (1.34 eV for sample A and 1.42 eV for sample B, as shown in Fig. 1).

In Fig. 2(b) we display the acoustic-phonon broadening efficiency a for the nine single QD's investigated in sample A (filled circles) and B (squares). We plot these data as a function of the normalized PLE background. Below a 0.1 PLE background value, all the QD's exhibit the quenching of the acoustic-phonon dephasing. Above this value, we observe a linear variation of the acoustic-phonon broadening efficiency. This increase reveals a density of states effect for the activation of acoustic-phonon scattering. Note that *despite the fact that the QD's have different Ga concentrations and come from two laboratories*, the normalized PLE background appears as a very reliable parameter for characterizing the influence of the environment on the fundamental transition dephasing. Since the normalized PLE background is measured 10 meV above the QD fundamental transition, such an estimation gives only an upper limit for the residual density of states available for acoustic-phonon scattering and explains the artificial threshold in Fig. 2(b).

As far as optical-phonon broadening is concerned, we extract an activation energy of 25 ± 5 meV from the fit displayed in Fig. 2(a) for the QD of sample B. Our data give good indication that the activation energy is significantly lower than in the InGaAs/GaAs 2D QW's case where ΔE corresponds to the optical-phonon energy (36 meV) in GaAs.[25] More accurate measurements require the investigation on a larger temperature range. Experiments on QD's emitting at lower energy are in progress and will be reported elsewhere.

We now address the zero-temperature limit. In the regime of a decoherence limited only by the radiative coupling, one should measure Γ_0 of the order of 0.6 μeV in InGaAs QD's. In Fig. 2(c) we plot the nine Γ_0 values which are all above 13 μeV. Furthermore, they present a linear variation with the normalized PLE background, showing again an influence of the WL band tail. Unlike the acoustic-phonon broadening, there is no thresholdlike variation in Fig. 2(c) and indeed we will see now that Γ_0 is more precisely determined by a large portion of the WL band tail.

To further elucidate the broadening mechanisms in the zero-temperature limit, we studied the fundamental transition linewidth at low temperature (10 K) as a function of the laser energy E_{ex} in sample B. For these QD's we take advantage of the presence of excited states [marked by arrows in Fig. 1(b)] to perform linewidth measurements with an excitation below the WL band edge. When the laser is tuned to lower energies, we systematically observe a striking *increase of the interferogram decay time* up to 50% (giving decay time as long as 170 ps, i.e., $\Gamma_0 \sim 7.5$ μeV). To evaluate the corresponding line narrowing, we normalize the zero-temperature linewidth Γ_0 by its value measured for an excitation above the WL band edge. We plot this ratio versus the excess energy $E_{ex} - E_{QD}$ in Fig. 3 for the five QD's of sample B. Although the energy distance to the WL varies from 100 to 130 meV for these QD's, we observe an amazing systematic line narrowing as a function of the excess energy $E_{ex} - E_{QD}$.

We interpret this effect by a reduction of the influence of the electrostatic environment on the QD.[27] Recently, spectral diffusion has been reported in CdSe nanocrystals[28] and in self-assembled InAlAs QD's with blinking periods of the order of seconds together with meV spectral shifts of the QD transition energies.[29] In our case we do not observe any

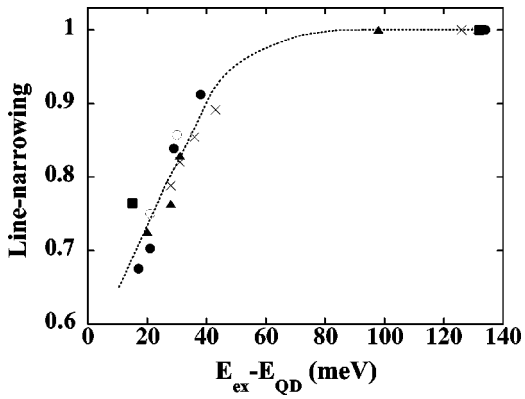

FIG. 3. Line narrowing ($\Gamma_0/\Gamma_0^{E_{ex}=E_{WL}}$) versus excess energy $E_{ex}-E_{QD}$ for the five QD's of sample B. The dashed line is a guide for the eyes.

blinking nor large spectral shifts similar to Ref. 29, indicating a weaker electrostatic interaction in our QD's. Moreover, any line multiplet in the PL spectrum coming from smaller spectral shifts within the detector integration time would result in beatings in the interferogram contrast decay. On the contrary, we systematically observe an exponential decay of the interferogram contrast showing that the environment induces pure dephasing as recently calculated in Ref. 30.

When the excess energy E_{ex}-E_{QD} is reduced below 40 meV, fewer charge carriers contribute to the electrostatic broadening of the QD fundamental transition, leading to an efficient line narrowing (Fig. 3). In fact, nonresonant excitation in the WL band tail produces electron-hole pairs localized in the QD's surrounding which relax into the QD. We end up with a picture of a fluctuating environment intrinsically bound to the nonresonant capture process. However, from the observation of the line narrowing effect in Fig. 3, we deduce that a control of the excess energy $E_{ex}-E_{QD}$ allows to tailor the influence of the electrostatic environment on the QD dephasing.

In conclusion, we report high-resolution linewidth measurements on the fundamental transition in single InGaAs QD's. A compact and flexible setup based on a Michelson interferometer allows a powerful investigation of single QD optical properties by combining PL, PLE, and FT spectroscopy in a simple way. We clarify the limits of the isolated macroatom scheme and we show how to achieve the quenching of acoustic-phonon dephasing and a control of the electrostatic broadening.

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[1] U. Bockelmann *et al.*, Phys. Rev. B **42**, 8947 (1990).
[2] T. Takagahara, Phys. Rev. B **62**, 16 840 (2000).
[3] L.C. Andreani *et al.*, Phys. Rev. B **60**, 13 276 (1999).
[4] J.M. Gérard *et al.*, Phys. Rev. Lett. **81**, 1110 (1998).
[5] A. Kiraz *et al.*, Appl. Phys. Lett. **78**, 3932 (2001).
[6] A. Imamoglu *et al.*, Phys. Rev. Lett. **83**, 4204 (1999).
[7] P. Chen *et al.*, Phys. Rev. Lett. **87**, 067401 (2001).
[8] P. Michler *et al.*, Science **290**, 2282 (2000).
[9] E. Moreau *et al.*, Phys. Rev. Lett. **87**, 183601 (2001).
[10] For a review, see, e.g., *Quantum Dot Heterostructures*, edited by D. Bimberg, M. Grundmann, and N.N. Ledentsov (Wiley, Chichester, 1998).
[11] M. Paillard *et al.*, Phys. Rev. Lett. **86**, 1634 (2001).
[12] P. Borri *et al.*, Phys. Rev. Lett. **87**, 157401 (2001).
[13] D. Birkedal *et al.*, Phys. Rev. Lett. **87**, 227401 (2001).
[14] J.Y. Marzin *et al.*, Phys. Rev. Lett. **73**, 716 (1994).
[15] M. Grundmann *et al.*, Phys. Rev. Lett. **74**, 4043 (1995).
[16] Y. Toda *et al.*, Phys. Rev. Lett. **82**, 4114 (1999).
[17] K. Matsuda *et al.*, Phys. Rev. B **63**, 121304 (2001).
[18] C. Kammerer *et al.*, Phys. Rev. Lett. **87**, 207401 (2001).
[19] C. Kammerer *et al.*, Phys. Rev. B **65**, 033313 (2001).
[20] See, for instance, R. J. Bell, *Introductory Fourier Transform Spectroscopy* (Academic Press, New York, 1972).
[21] D. Gammon *et al.*, Science **273**, 87 (1996).
[22] S. Rudin *et al.*, Phys. Rev. B **42**, 11 218 (1990).
[23] L. Besombes *et al.*, Phys. Rev. B **63**, 155307 (2001).
[24] W. Braun *et al.*, Phys. Rev. B **56**, 12 096 (1997).
[25] P. Borri *et al.*, Phys. Rev. B **59**, 2215 (1999).
[26] G. Cassabois *et al.*, Phys. Rev. B **61**, 1696 (2000).
[27] A heating due to phonon emission during population relaxation could also broaden the QD transition. In that case the linewidth would increase with the incident power. On the contrary the linewidth does not vary in the incident power range we investigated.
[28] R.G. Neuhauser *et al.*, Phys. Rev. Lett. **85**, 3301 (2000).
[29] H.B. Robinson *et al.*, Phys. Rev. B **61**, R5086 (2000).
[30] A.V. Uskov *et al.*, Appl. Phys. Lett. **79**, 1679 (2001).