Quantum dot decoherence measured by ensemble photoluminescence

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We propose and experimentally demonstrate that a very simple, noninvasive, and common technique, i.e., standard nonresonant photoluminescence of large ensemble of quantum dots (QDs), can be used to determine the temperature dependence of homogeneous broadening. The method can be applied to a single quantum dot layer independently of the QD density and it allows to follow the increase of the homogeneous broadening up to high T. The experimental results show that different pictures apply depending on the QD size. © *2005 American Institute of Physics*.
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I. INTRODUCTION

Semiconductor quantum dots (QDs) are potentially basic elements for advanced optoelectronic nanodevices and for quantum information technologies.[1–4] Several promising applications in these fields, such as single-photon source for quantum cryptography,[1] quantum bits,[4] or quantum logical elements,[3,4] require the coherent optical manipulation of the electronic levels in the QDs. Most of the proposals take advantage from both the atomiclike nature of the QD states and the enhancement of light-matter interaction typical of condensed-matter systems. The price to be paid, with respect, for instance, to isolated atoms, is the influence of the solid-state environment which strongly interacts with the QD few-level system in the form of decoherence or dephasing mechanisms.[5–9] It is well known that charges localized inside the QD interact with lattice vibrations via acoustic and optical phonons, producing a strong temperature dependence of the homogenous broadening. Non-Lorentzian line shapes[6,8,9] and nonexponential decay of coherence[6] have been reported and interpreted in semiconductor QDs. In particular, the single QD PL line shape is found to be the sum of a sharp Lorentzian line and pronounced sidebands.[6,9] The sharp component of the single QD emission is attributed to the zero-phonon line (ZPL). The asymmetric sidebands are interpreted as the result of the elastic electron-phonon interaction with acoustic phonons,[9–13] within the picture that the electronic excitations imply a distortion of the equilibrium lattice position. The dependence of the relative weight of these two contributions on the QD size and shape, on temperature, and on structural disorder has not yet been completely clarified. For example, the structural disorder of the QD environment seems to play a relevant role in determining the dephasing processes, but also the electron-phonon interaction and different temperature dependences of the homogeneous broadening have been reported.[9] Recently it has been reported that annealing can produce radiatively limited dephasing in QDs.[14]

Despite the relevant progress achieved in the last few years on the understanding of the complicate dynamics of decoherence in QDs, driven by the tremendous interest for dephasing processes (there are no doubts that decoherence is the fundamental limit to be overcome for achieving QD devices in the quantum computational area), a deep and unified comprehension is still missing, mainly due to the lack of systematic studies on large numbers of samples. The reason for that is mainly related to the intrinsic difficulties associated with the two experimental techniques so far used for analyzing decoherence phenomena in QDs, such as microphotoluminescence (micro-PL) and four wave mixing (FWM). Micro-PL requires samples with very low QD density and/or lithographic techniques (i.e., invasive processing of the samples) for selecting a very small area. Moreover the detected signal is quite small and it is very difficult to follow it when increasing the temperature due to the broadening and the quenching of the emission bands; many studies are then limited to the range below 100 K.[8,9,15] FWM is a very powerful technique for measuring microscopic dephasing even in largely inhomogeneosly broadened QD ensembles. However, it requires femtosecond laser pulses, complicated setup, and, more importantly, great care due to the very low optical density of QDs. In fact, most FWM reports indeed refer to QD multilayers.[5–8,16] In addition, the standard laser pulse duration of 100 fs limits the investigation to homogeneous broadening smaller than few meV.[5] Here we propose, and experi-

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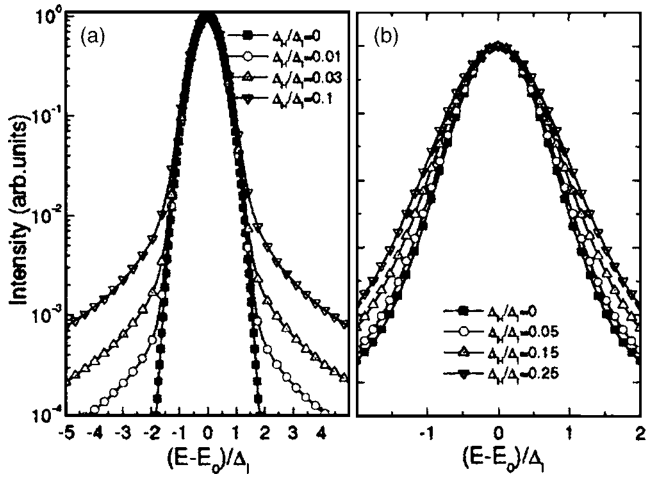

FIG. 1. Voigt profiles for different values of homogeneous broadening Δ_H (here $\Delta_I=1$). (a) Semilogarithmic scale and (b) linear scale. The changes in the line shapes are clearly resolvable.

mentally demonstrate, a simple, noninvasive, and very common technique, i.e., standard PL of large ensembles of QDs with nonresonant excitation, can be used to determine the temperature dependence of the homogeneous broadening in QDs. The method can be applied to a single QD layer independently of the QD density and it allows to follow the increase of the homogeneous broadening up to high temperature *T*.

II. GENERAL CONSIDERATIONS

The idea is extremely simple and it is based on the line-shape analysis, over several orders of magnitude in intensity, of the PL emission band. It is well known that, in the presence of both homogeneous and inhomogeneous broadenings, the optical density is given by the convolution of the two line shapes. In most of the accessible *T* range, the phonon-induced dephasing produces a homogeneous broadening which is usually more than one order of magnitude smaller than the inhomogeneous broadening, thus making its effect on PL emission linewidth hardly detectable. However, while the homogeneous broadening increases with the temperature, the inhomogeneous broadening, that is due to static structural disorder, is independent on the temperature. Therefore, the effects of homogeneous broadening should be searched in the temperature-dependent modifications of the PL line shape. The tails of the PL band are the most sensible part of the PL line shape where modifications, due to the increase with *T* of the homogeneous broadening, should be looked for. Let us justify this statement. For simplicity we assume that the homogeneous line shape is Lorentzian, although deviation from this form has been observed in single QD photoluminescence.[6] In Fig. 1 a simulation of the effects of a small Lorentzian broadening within a large Gaussian inhomogeneously broadened band, often denominated Voigt profile, is reported. Here Δ_H and Δ_I represent the half width at half maximum (HWHM) of the homogeneous and inhomogeneous line shape, respectively; in Fig. 1 the energies are scaled by Δ_I (i.e., $\Delta_I=1$). It is clear from Fig. 1(a) that the effect of the increase of Δ_H is a change of the band tail profiles. At least three to four orders of magnitude in the signal-to-noise ratio (S/N) are needed to extract a homogeneous broadening of the order of $\Delta_H/\Delta_I=0.01$. However, with three decades of S/N the method is sensitive to $\Delta_H/\Delta_I=0.03$. To quantify this estimate for the case of QDs, where Δ_I is in the range of 10–30 meV, *a priori* one can measure Δ_H as small as 0.1–0.3 meV, which is of the same order of magnitude as the linewidth of the single QD PL at low *T*. At the same time all the studies so far reported show that the QD homogeneous broadening at high *T* is of the order of several meV and therefore it can be easily measured by the proposed analysis. Figure 1(b) reports, in a linear scale, the simulation in the cases of larger homogeneous broadenings. We clearly see that Δ_H is resolved by measuring the HWHM without any fitting procedure. It is worth stressing that the Voigt profile broadens linearly (slope 0.6) when increasing the homogeneous Lorentzian broadening Δ_H. In fact, fits of the HWHM of the PL emission of quantum well structures have been used in the past for measuring the *T* dependence of the homogeneous broadening of excitons.[17]

Before going on and discussing the experimental data, let us analyze the method for homogeneous broadening different from Lorentzian. This is because it has been recently pointed out that the phonon sideband in strongly confined QD is likely better approximated by a Gaussian, even if generally it does not follow a simple analytical expression. In principle, deconvolution methods, by means of Fourier transform analysis, may be used to resolve the homogeneous contribution without any assumption on its line shape. However, in the case of a Gaussian homogeneous broadening, the convolution with the inhomogeneously broadened Gaussian band will be still a Gaussian with total width Δ_T given by $\Delta_T=\sqrt{(\Delta_I^2+\Delta_H^2)}$. This means that the sensitivity of the method becomes much worst for Gaussian homogeneous broadening: in this case the PL tails would not bring information on Δ_H. In order to fix the order of magnitude we can state that Δ_H can be resolved when $\Delta_T/\Delta_I=1.02$, that is, $\Delta_H/\Delta_I=0.2$. Only a very large Gaussian homogeneous broadening would be detectable in ensemble PL measurement.

III. RESULTS AND DISCUSSION

Let us now discuss the experimental data. We have investigated two different samples. QD1 is a molecular-beam epitaxy (MBE)-grown, Stranski-Krastanow, InAs/GaAs QD sample with emission band peaked at 1.08 eV and PL-HWHM of 14 meV at $T=10$ K. The wetting layer (WL) electronic states lie 380 meV above the QD ground-state levels. QD2 is an InGaAs/GaAs QD structure grown without a WL by taking advantage of a modified MBE method, the heterogeneous droplet epitaxy (HDE).[18,19] The HDE growth method permits the assembling of nanometer size InGaAs inclusions in a GaAs matrix. The typical dimensions of the HDE-QDs are 30 nm base width and 12 nm height and the average In content of the QD is around 20%. The QD2 PL peaks at 1.285 eV and the HWHM is 11 meV at $T=10$ K. More details on the HDE-QD growth may be found in Ref. 19.

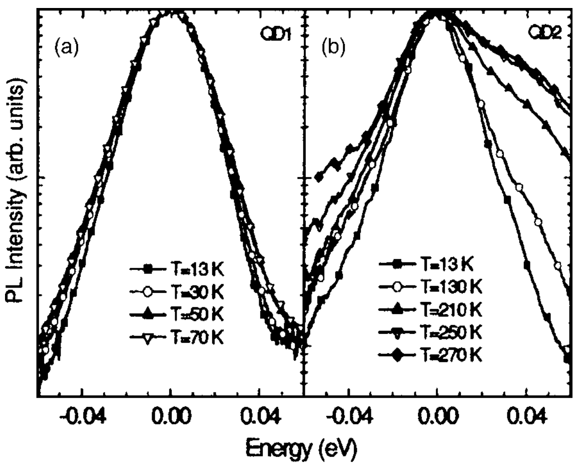

FIG. 2. a) QD1 PL spectra for different temperatures. (b) QD2 PL spectra for different temperatures.

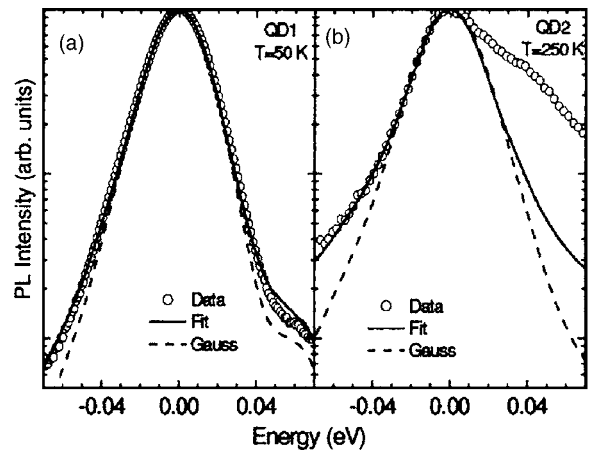

FIG. 3. (a) Data (open circles), Lorentzian fit (solid black line), and Gaussian simulation (dashed line) for QD1 at $T=50$ K. (b) Data (open circles), Lorentzian fit (solid black line), and Gaussian simulation (dashed line) for QD2 at $T=250$ K.

The experimental data are reported in Figs. 2(a) and 2(b), for samples QD1 and QD2, respectively, where the PL spectra in a semilogarithmic plot at different temperatures are reported. At high T, the recombination from the excited states becomes evident on the high-energy side for all the samples. To avoid artifacts due to this contribution we consider only the low-energy tails of the PL bands. Figures 2(a) and 2(b) clearly show that the PL bands broaden when the temperature was increased. The fit of the low-energy tail can be therefore used to extract information on the T dependence of the homogeneous broadening, once an analytical expression is assumed to model it. The fact that the low-energy tail of the QD-PL spectra is more and more relevant when T is increased suggests a major role of Lorentzian-like broadening with respect to Gaussian-like broadening. We therefore assume a Lorentzian line shape for the homogeneously broadened band. Within this assumption the fitting procedure becomes very simple.

It is reasonable to assume that the low-T PL band represents the QD optical density due to the stochastic carrier injection into the QDs and the lack of coupling between adjacent QDs. In other words we can use the low-T PL line shape $I_0(\omega)$ as a reference to measure the T dependence of the homogeneous broadening. $I_0(\omega)$ is given by the convolution of the inhomogeneously broadenend band $i(\omega)$ and the low-T homogeneously broadened Lorentzian band $L_0(\omega)$, with HWHM Δ_0,

$$I_0(\omega) = \int L_0(\omega')i(\omega-\omega')d\omega'. \tag{1}$$

With increasing T, due to the phonon-assisted dephasing, the homogeneous broadening $\Delta(T)$ increases,

$$\Delta(T) = \Delta_0 + \delta(T), \tag{2}$$

and the profile $L(\omega)$ broadens. We now take advantage of two simple mathematical results. The first is that a Lorentzian band $L(\omega)$ can be written as the convolution integral of two Lorentzian profiles $L_0(\omega)$ and $\delta L(\omega)$,

$$L(\omega) = \int L_0(\omega')\delta L(\omega-\omega')d\omega', \tag{3}$$

with the constraint that the sum of the broadenings of $L_0(\omega)$ and $\delta L(\omega)$ equals the broadening of $L(\omega)$. The second is that the convolution integral satisfies the associative property. Then it follows that the high-T PL line shape $I(\omega)$ can be written as

$$I(\omega) = \int \delta L(\omega')I_0(\omega-\omega')d\omega', \tag{4}$$

where $\delta L(\omega)$ is a Lorentzian line shape having a width corresponding to the increase of the QD homogeneous broadening $\delta(T)$.

In summary we fit the PL band at different temperatures by using the low-T PL of the QDs as the reference line shape to be convolved with a Lorentzian profile. The broadening of the Lorentzian then directly provides the increase of the homogeneous broadening.

Typical Lorentzian fits are reported as solid black lines in Figs. 3. The experimental data are reproduced with very high accuracy by using $\delta=1.6$ meV and $\delta=4.5$ meV for QD1 and QD2, respectively. This also denotes the validity of the Lorentzian line-shape approximation. For comparison, simulations with Gaussian line shape for the homogeneous broadening are also reported, as dashed lines in Figs. 3 by using a Gaussian HWHM of 5 and 10 meV for QD1 and QD2, respectively. Note that the assumption of a Gaussian line shape for the homogeneous broadened band does not fit the PL spectra within a similar accuracy. Moreover the values of the homogeneous broadening do turn out to be much larger than the ones obtained within the Lorentzian assumption.

Figure 4 shows the T dependence of the homogeneous broadening, as calculated via the described methodology and Lorentzian line-shape fits, for the two samples. The homogeneous broadening of sample QD1 sharply increases in the low-T regime. The QD1 PL broadening stops around $T=50$ K where it reaches a value of 1.6 meV. Due to the pres-

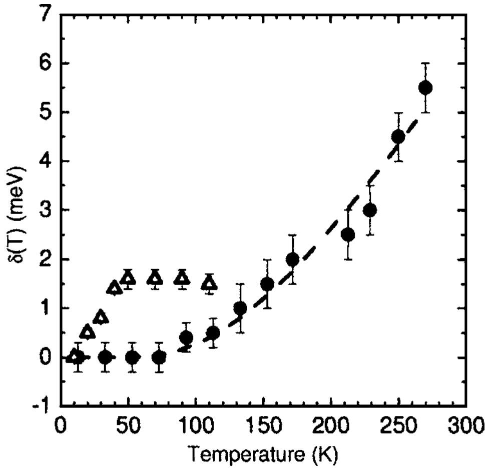

FIG. 4. Summary of the homogeneous broadening $\delta(T)$ for both QD1 and QD2.

ence of concurrent, temperature-activated, anomalous behavior,[20] the maximum temperature that we can evaluate for the determination of the $\delta(T)$ behavior is limited to 110 K. The data for QD2 reveal a different trend. The QD2 homogeneous broadening is almost constant in the low-T region, up to a temperature of ≈ 70 K. Then, it increases rapidly and at room temperature the measured value of $\delta(T)$ is 5.5 ± 0.5 meV.

Let us discuss the phenomenology presented on the basis of the existing reports. As stated in the Introduction, the single QD PL line shape constitutes by a sharp ZPL with pronounced sidebands. In Ref. 6 a strong increase of the sidebands, with respect to the ZPL, is found with increasing temperature becoming dominant above 70–100 K.[6,9] The acoustic-polaron sideband depends on the temperature in a quite complicated way. After an initial nonmonotonouos dependence on T (Refs. 16 and 21), at high T ($T > 100$ K) a weak linear increase, with a slope of few μeV/K, is eventually observed, possibly due to an intrinsic cut-off energy of the acoustic-phonon modes, determined by the inverse of the carriers localization length.[6,11] The ZPL broadening, usually interpreted as due to inelastic scattering, is described by the phenomenological dependence,[6,9] derived for the quantum well case,[5]

$$\delta(T) = \gamma T + b/[\exp(E_A/k_B T) - 1]. \tag{5}$$

The same phenomenological law is also used whenever the single QD spectrum is dominated by a sharp, Lorentzian-like emission.[22–24] The linear term is due to acoustic-phonon interaction and the value of γ is rather spread in the literature from 0.2 μeV/K to a few μeV/K.[6,23,25] Values of E_A are also spread in the literature, possibly due to the lack of studies up to room temperature. In Refs. 9, 22, and 23 is found $E_A \approx \hbar\omega_{LO}$ ($\hbar\omega_{LO} = 36.6$ meV in GaAs); lower values of E_A are also reported in the literature, ranging from 15 to 20 meV and usually attributed to the lowest separation energy of the electronic transitions in the QD,[6,24] suggesting a signature of a phonon bottleneck. A different interpretation of the ZPL broadening is given in Ref. 16, where the temperature-activated ZPL broadening show two activation energies (6 and 28 meV) and it is regarded as stemming from pure dephasing due to the elastic interaction with phonons.

Coming back to our results, the $\delta(T)$ dependence of sample QD1 can be reasonably fitted, at low T, by a linear dependence with a slope of 43 ± 4 μeV/K. This value is an order of magnitude larger than the so far reported γ for the linear broadening of the ZPL, washing out the possibility to associate it to the homogeneous broadening of the ZPL. We believed that such large increase of the homogeneous linewidth reflects the transition from a ZPL to a spectrum dominated by large phonon sidebands due to electron-acoustic phonon elastic interactions expected for strongly confined QD. Assuming $\delta(T)$ to be the broadening of the phonon sidebands, our data nicely agree with those obtained by FWM by Borri *et al.*[6] for a similar Stranski-Krastanow InAs/GaAs QD sample (PL peak at 1.16 eV). Our PL findings thus confirm the results, extracted from FWM experiments,[6] that the high-T QD emission linewidth is dominated by large acoustic-phonon sidebands. Our data also point out that the line shape of acoustic-phonon sidebands should have a Lorentzian-like line shape. This is in contrast with a recent determination of a Gaussian-like initial decay of the QD coherence.[16] A possible explanation is that the line shape of the acoustic-phonon sidebands does not follow a simple analytical expression and, similar to Voigt profile, may present Gaussian shape around the maximum and Lorentzian-like profile far in the tails. At the same time, since it has been suggested that the QD environment plays a relevant role in the electron-phonon interaction,[9,16] we may infer that extrinsic effects due to the QD surrounding could also influence the line shape of the acoustic-phonon sidebands.

The temperature behavior of sample QD2 shows a different picture. The increase of the homogeneous linewidth is very small, if any, in the low-T region, while an exponential increase is found at high T. One can qualitatively comment that in the case of QD2, the QD dimensions are large enough to inhibit the polaron picture. In other words, the acoustic-phonon cut-off energy is small. The data can indeed be nicely fitted by Eq. (5) in the whole temperature range, which extends up to RT, as shown by the dashed line in Fig. 4, with $E_A = 36$ meV. We conclude that in the case of sample QD2, the ZPL is broadened mainly by optical-phonon interactions, similar to bulk and quantum wells.[5,26] We like to stress that in both samples we observe the emission, at high temperatures, from the QD excited states with a probability determined by the thermal equilibrium in the QDs. This supports an efficient coupling of the QD-confined excitons with the LO phonons, thus implying the lack of a phonon bottleneck for carrier thermalization in the QDs.

IV. CONCLUSIONS

In summary we have demonstrated that a straightforward and noninvasive method, such as the ensemble PL line-shape analysis, can be used to extract information on the decoherence in semiconductor QDs. We have studied two different QD systems. The results show that different pictures apply depending on the QD size. For small QDs, the four wave mixing results on the importance of nonperturbative elastic interaction of confined excitons with the acoustic phonon in determining the QD emission linewidth are confirmed. For large QDs the zero-phonon line broadens via LO-phonon interaction and a very small, if any, contribution from acoustic sidebands is inferred. We obtain a very accurate determination of the activation energy of the inelastic-scattering processes determining the ZPL linewidth, owing to the extended temperature range.

As concluding remarks we would like to stress that care has to be taken into account in checking that the assumption of a constant inhomogeneous broadening applies for the samples under investigation. For instance, the use of this procedure should be avoided when the selective PL quenching due to the WL becomes relevant, since it is well known that this produces a reduction of the PL linewidth.[20] Nevertheless, the method is very general and it is not limited to low-density or processed samples needed for micro-PL nor to QD multilayers required for FWM measurements, which are far more complicated techniques. We believe that our proposal offers the opportunity to investigate this relevant topic in an extremely simple way and hopefully this will produce the possibility to achieve a much better understanding of QD dephasing mechanisms, eventually leading to the control and even to the tuning of decoherence in QDs.

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