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Temperature dependence of the exciton homogeneous linewidth in $In_{0.60}Ga_{0.40}As/GaAs$ self-assembled quantum dots

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Single dot photoluminescence spectroscopy was used to study the homogeneous linewidth Γ of the ground-state exciton in $In_{0.60}Ga_{0.40}As/GaAs$ quantum dots as function of temperature T. In high resolution experiments at 2 K, we find a linewidth that is limited by the excitonic lifetime corresponding to a dephasing time of almost a ns. The approximately linear increase of Γ with temperature up to ~ 30 μeV at 60 K is considerably weaker than in structures of higher dimensionality. For higher T we observe a strong enhancement of the linewidth reaching eventually a few meV at room temperature that depends on the confined electronic shell structure.

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Currently there is great interest in obtaining solid-state implementations for quantum information processing, including quantum cryptography and quantum computation.[1] The essential building blocks of such implementations are two state quantum bits (qu-bits), which store quantum information in the form of linear combinations of the two states. Qu-bits being considered often are based on the electronic states of quantum dots (QDs). A key requirement of qu-bits is that they remain coherent for the times needed to perform operations on them. An important perturbation of coherence in a solid-state environment is the carrier-phonon interaction. For QDs, this interaction has attracted considerable attention over the past several years which was stimulated by the proposal of a phonon-relaxation bottleneck.[2,3] But still a conclusive picture of the exciton-phonon interaction has not been developed yet.

The phonon interaction determines also the temperature dependence of the exciton homogeneous linewidth Γ which for QDs has been addressed in several reports.[4–8] Using single dot photoluminescence spectroscopy, values of Γ larger than ~ 40 μeV have been reported for $T<10$ K.[4–7] Further, a rather weak dependence of the emission half-width on T was demonstrated up to about 80 K. At room temperature, the homogeneous linewidth was studied by spectral hole burning and four-wave mixing on QD ensembles that were embedded in waveguides.[8] In these experiments surprisingly large half-widths on the order of a few meV were measured. However, no data have been given for the full temperature range from liquid helium up to room temperature.

Here we have addressed the temperature dependence of Γ by photoluminescence spectroscopy on single QDs, for which a detailed understanding of the confined electronic shell structure had been obtained earlier.[9] Above 60 K, the thermal excitation of optical phonons enhances exciton scattering considerably resulting in a strong increase of Γ, that depends on the electronic shell structure of the QDs. By contrast, for lower temperatures the line broadening shows no significant dependence on the level structure. At 2 K, Γ is less than 2 μeV indicating that the dephasing is lifetime-limited.

For performing photoluminescence spectroscopy, single QDs were isolated in lithographically fabricated mesa structures.[10] The structures were placed in the variable temperature insert of an optical cryostat which permitted us to vary the temperature between 2 K and room temperature. Optical excitation was done either nonresonantly by an Ar or a He-Ne-laser or resonantly by a Ti-sapphire laser. Low excitation powers <1 W m^{-2} were used in order to avoid thermal heating by laser irradiation. The emission was dispersed by a double grating monochromator (with a focal length of $f=1$ m per stage) or single grating monochromators with $f=0.6$ m or 0.32 m.

The signal was detected by a Peltier-cooled photomultiplier or a LN_2-cooled charge-coupled-device camera using integration times that were chosen according to the signal strength. At low temperatures <60 K the emission is rather intense so that integration times of 60 s were sufficient to obtain reasonable signal-to-noise ratios. In these experiments the high resolution double monochromator was used. For higher temperatures up to about 150 K, the integration time had to be increased up to 5 min due to a decrease of signal strength arising from thermal emission of carriers. When raising the temperature further, the adjustment of the laser beam on a single QD according to the dot emission intensity becomes problematic because of its weakness within reasonable signal collection times.

Therefore first the temperature was fixed and then the optical alignment was done by focusing the laser beam on a large mesa structure containing $\sim 10^6$ QDs. This measurement also gave us information in which energy range the single dot emission was expected at the given temperature. Since the mesa structures were arranged in periodic patterns with defined separations between them, the laser beam afterwards was moved to the position at which the single QD under study was located: Then collection of the signal was started with total integration times up to 4 h. After each of these measurements the temperature was lowered to $T<77$ K and the emission from the single dot was checked. This procedure was repeated several times. In addition, to obtain enough signal, the high resolution monochromator was replaced by a low resolution one. Care was taken, however, that the measured linewidths were not resolution-limited.

The temperature dependence of the homogeneous linewidth $\Gamma(T)$ of an exciton is determined by its interaction with phonons. It is given by the probability for the exciton to scatter from the ground shell to higher lying states. Therefore

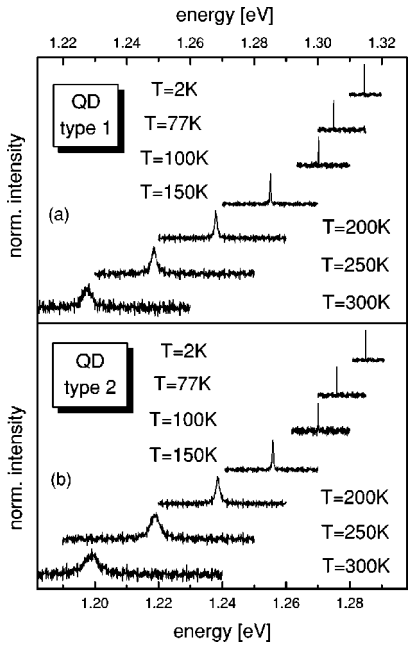

FIG. 1. (a) Photoluminescence spectra of a single $In_{0.60}Ga_{0.40}As/GaAs$ QD of type 1 with two confined electronic shells recorded for varying temperatures. (b) Same as (a) but for a single $In_{0.60}Ga_{0.40}As/GaAs$ QD of type 2 with three confined electronic shells.

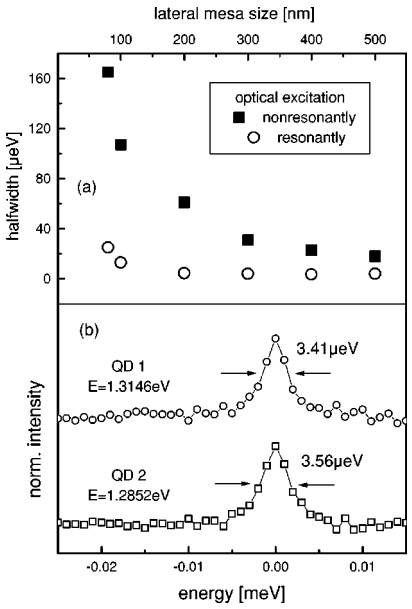

FIG. 2. (a) Minimum measured emission linewidth in photoluminescence studies of a single $In_{0.60}Ga_{0.40}As/GaAs$ QD at $T=2$ K plotted against the nominal lateral size of the mesa structure in which the dot is contained. (b) High resolution photoluminescence spectra of single $In_{0.60}Ga_{0.40}As/GaAs$ QDs of type 1 and 2 recorded at $T=2$ K. To facilitate resolution and comparison, the energy of the center has been subtracted for each emission line. These energies are denoted at each trace.

it is crucial to have a detailed understanding of the electronic shell structure of the QDs. In Ref. 9 we studied the excitonic absorption of self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ single QDs that exhibit high geometric symmetry resulting in simple absorption spectra. Two different types of dot structures were investigated: For the first type two electronic shells, the *s* and the *p* shell, were confined. For the second dot type, the *d* shell was confined in addition. Regarding spin and orbital angular momentum degeneracies, these shell structures lead to a total of 6 and 12 confined single particle states for type 1 and type 2, respectively. Taking the electron-hole interaction into account, the excitonic absorption spectrum consists of only two features for type 1 dots (one each in the *s* and the *p* shell) as compared to five features in the type 2 QDs (one in the *s*, two each in the *p* and the *d* shell).

Figure 1(a) shows photoluminescence spectra of a QD characteristic for highly symmetric type 1 structures that were recorded at varying temperatures using He-Ne laser excitation. For each temperature a single emission line is observed. The band-gap shrinkage with increasing T results in a considerable shift to lower energies that is comparable with that of the GaAs band gap. Up to temperatures of about 100 K, the dot emission is a sharp line with a width below 100 μeV (FWHM), then its width starts to increase strongly. At room temperature it is ~ 3 meV. Figure 1(b) shows the corresponding measurements for a QD of type 2. Its behavior is qualitatively similar to that of QD 1: The emission is spectrally narrow up to 100 K, for higher values it becomes a broad band with a width of more than 5 meV at 300 K.

In general, the linewidths measured in such nonresonant photoluminescence studies of single QDs do not correspond to the homogeneous ones intrinsic of the dots because they might be influenced by fluctuating charge distributions at the lateral sidewalls of the mesa structures or at defects in the dot environment. As is shown by the squares in Fig. 2(a), we find a correlation between the minimum single dot emission linewidth and the lateral size of the mesa in which the dot is located ($T=2$ K). The excitation was done by a He-Ne laser. For mesa structures with sizes below ~ 100 nm the minimum linewidths are always larger than 100 μeV. With increasing size it decreases to about 15 μeV for the largest studied structure. Even for large sizes, however, no saturation of the emission linewidth is observed demonstrating that the surfaces still influence the dot confinement potential.

To avoid creation of fluctuating charges by carrier diffusion, experiments with resonant excitation below the wetting layer into the *p*-shell states of the QDs were performed. Figure 2(b) shows the corresponding spectra of QDs 1 and 2 recorded at $T=2$ K. Extremely sharp Lorentzian emission lines with linewidths of 3.4 ± 0.4 μeV are observed for both dot types. Deconvolution of the spectral resolution of the setup which is 2.0 μeV gives a Γ of 1.8 ± 0.4 μeV ($T_2 \sim 730$ ps). Under these experimental conditions the dependence of the minimum emission linewidth on mesa structure size is reduced as well [the circles in Fig. 2(a)]. For sizes larger than 200 nm it saturates below 5 μeV. For smaller sizes, however, there is still an increase of Γ which might originate from tunneling of carriers to sidewall traps. We note that the QDs under study are located in mesa structures with lateral sizes of ~ 400 nm.

The temperature dependences of Γ for the QDs 1 and 2 are summarized in Fig. 3, where Γ is plotted on a logarithmic scale versus temperature on a linear scale.[11] The right axis shows the corresponding dephasing times which have been

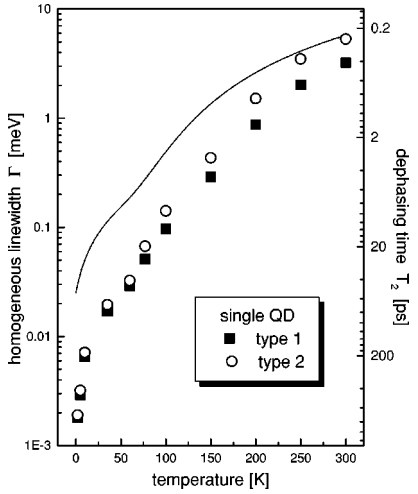

FIG. 3. Homogeneous linewidth (left axis), respectively, dephasing time (right axis) of the ground-state exciton in $In_{0.60}Ga_{0.40}As/GaAs$ single QDs of type 1 and 2 plotted on a logarithmic scale versus temperature on a linear scale. The solid line gives the T-dependence of Γ for an $In_{0.18}Ga_{0.82}As/GaAs$ QW calculated after Ref. 14.

calculated according to $\Gamma = 2\hbar/T_2$. Over the whole studied T range, the spectral linewidth increases by three orders of magnitude. For $T < 60$ K, the increase of Γ with T is very similar for the two dot types. For higher temperatures, in contrast, the emission broadens more strongly for the QD of type 2 than for that of type 1. At room temperature, for example, the half-widths differ roughly by a factor of 2: For dot 2, Γ is 5.3 meV as compared to the 3.2 meV observed for dot 1.[12] We note that the observed line shapes can be well described by Lorentzians for $T < 40$ K only.[13]

In structures of higher dimensionality the temperature dependence of the homogeneous linewidth is described by the following form:

$$\Gamma(T) - \Gamma(T=0) = \gamma_{ac} T + \gamma_{op} \frac{1}{\exp\left(\hbar\omega_{LO}/kT\right) - 1}, \quad (1)$$

where the first term gives the acoustic phonon scattering, while the second one gives the scattering with optical phonons. At low T ($kT \ll \hbar\omega_{LO}$), the second scattering channel can be omitted because of the negligible LO-phonon population. At high T, on the other hand, the first channel can be neglected because $\gamma_{op} \gg \gamma_{ac}$. The solid line in Fig. 3 shows a typical temperature dependence of the homogeneous linewidth in an $In_{0.18}Ga_{0.82}As/GaAs$ quantum well calculated after Ref. 14.

Let us turn now to its temperature dependence in the QDs: First the low-temperature range shall be discussed. For $T < 60$ K the linewidth increase is to a good approximation proportional to the temperature, $\Gamma = \Gamma_0 + \gamma_{ac} \times T$. From a corresponding fit we determine the coefficient of the increase γ_{ac} to be ~ 0.5 μeV K^{-1}, independent of the structure type and thus also independent of the confined electronic level structure. This value is considerably smaller than for higher dimensional structures: For $In_{0.18}Ga_{0.82}As/GaAs$ QWs, for example, Borri *et al.*[14] have reported a γ_{ac} around 2.5 μeV K^{-1} that does not vary strongly with well width.[15] Therefore, due to the three-dimensional confinement, the γ_{ac} in the $In_{0.60}Ga_{0.40}As/GaAs$ QDs is decreased by a factor of 5 compared to the QW case. That there still is a T dependence of Γ at these low temperatures is not fully understood yet. In the excitation spectra we find no indication for a continuum of wetting layer states[16] that is extended in energy all the way down to the ground-state exciton. The linewidth increase might be traced to scattering of holes which are much closer spaced energetically than the electrons.[17,18]

Even more interesting is the homogeneous linewidth Γ_0 obtained by a zero-temperature extrapolation. A conservative estimate gives us an upper limit for Γ_0 of clearly less than 1.5 μeV. This Γ corresponds to a dephasing time T_2 of at least 900 ps. From time-resolved studies of QD ensembles, the decay time of the photoluminescence is determined to be about 1 ns. Therefore the data clearly indicate that coherence in the QDs is maintained within the exciton lifetime, which is independent of T up to 80 K. This might prove to be important for applications of QDs in quantum information processing, in which the QD is optically addressed by ultrashort laser pulses.

Now we turn to the regime above 60 K, in which thermal excitation of optical phonons influences considerably the exciton scattering. Here a strong increase of the homogeneous linewidth is observed, the temperature dependence of which can neither quantitatively nor qualitatively be described by Eq. (1): For $T < 200$ K, in QDs the increase of Γ with temperature is weaker compared to QWs. For larger T this relation is reversed, before Γ converges to values of a few meV at room temperature which are comparable to those reported in structures of higher dimensionality and correspond to dephasing times of less than 500 fs. Recently, there have been reports of a considerable dipole moment in self-assembled QDs due to a vertical displacement of the electron and hole wave functions[19] which would enhance the exciton-phonon interaction. We note that the excitation spectra of the QDs under study show only weak phonon replica indicating a rather small dipole moment.

In general, the scattering strength is given by (a) the interaction matrix element and (b) by the phase space of the final scattering states. We conclude that for temperatures below 200 K the number of states in the QD confinement potential that can be reached from the exciton ground state by phonon scattering is still restricted, resulting in the weaker dependence of Γ on T as compared to QWs. This reduced phase space in highly symmetric QDs apparently dominates over any potential modification of the scattering matrix element due to, for example, piezoelectric effects.[20]

Above 200 K the available phase space for scattering evidently is strongly extended: More and more scattering channels open up resulting in a somewhat faster rise of Γ up to room temperature than in QWs. Enlargement of the phase space by increasing T in this range suggests that not only single phonon absorption is important but also multiphonon processes[21–23] contribute to exciton scattering so that also states located more than one LO-phonon energy above the ground state exciton become accessible. These processes can have a significant probability only at temperatures at which the phonon population is considerably enhanced.

In spite of qualitatively similar *T*-dependences of Γ for QDs 1 and 2, the linewidth above 60 K increases more strongly for QD 2 than for QD 1 because the first dot offers more scattering channels in the excited shells eventually leading to the difference in Γ by a factor of 2 at room temperature. This observation is supported by studies of QDs with reduced geometric symmetry which exhibit more complicated absorption spectra than those of QDs 1 and 2.[24] Besides a larger number of sharp absorption features, in such QDs the intensity of the phonon replica is significantly increased and a considerable state continuum appears above the *p* shell[16] due to extended states from the wetting layer. For these structures the increase of Γ in the intermediate temperature regime (which is moderate for symmetric QDs) is much stronger. The open symbols in Fig. 4 give the data for two asymmetric dots of type 1 as compared to the symmetric one of Fig. 3. While at low T the dependence is hardly changed, above 50 K Γ rises much faster than in the symmetric case.

In summary, we have studied the homogeneous linewidth of self-assembled QDs by single dot spectroscopy. At room temperature the dephasing time is considerably shorter than 1 ps, which precludes application in quantum information processing. At very low temperatures, however, T_2 is about a ns which is limited by the exciton lifetime. Since the QDs can be designed to have a confined level splitting of up to 100 meV, the structures may be addressed by sub-100 fs pulses without exciting energetically higher lying states. Hence it might be possible to perform up to $\sim 10^4$ coherent optical manipulations on the QD ground-state exciton before decoherence destroys information. After having performed this work, we learned that similarly long dephasing times have been observed recently for InAs/GaAs self-assembled QDs studied by four-wave mixing.[25]

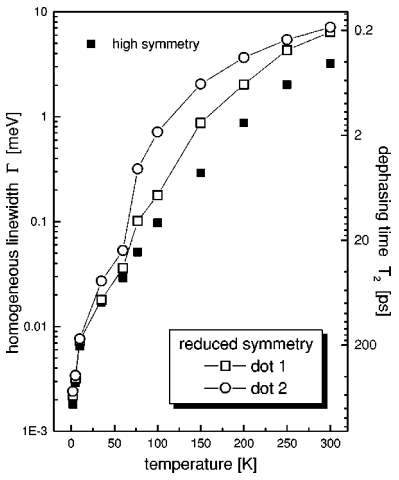

FIG. 4. Homogeneous ground-state exciton linewidth of three different $In_{0.60}Ga_{0.40}As/GaAs$ QDs of type 1 plotted versus temperature. The open symbols give the data for QDs with a reduced geometric symmetry, while the solid squares give those for a symmetric QD.

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[11] We note that for $T > 60$ K the linewidths measured for resonant and nonresonant excitation become comparable in large mesa structures indicating that for higher temperatures the influence of the phonons dominates over that of the sidewall charges on the QD confinement potential.

[12] This behavior is characteristic for the two QD classes: For different type 1 single QDs of high symmetry, FWHMs of 2.7, 3.1, 3.2, and 3.4 meV have been measured at room temperature, for corresponding type 2 QDs 4.9, 5.8, and 6.3 meV.

[13] In a *T*-range from $\sim$40 to 80 K, however, deviations from a Lorentzian line shape occur: The Lorentzians are superimposed on a spectrally rather broad background, which originates from low energy acoustic phonon sidebands (Ref. 7) due to exciton recombination with a simultaneous change of the phonon occupation number. As compared to the CdTe/ZnTe QDs in Ref. 7 and to the InAs/GaAs in Ref. 25, the zero-phonon recombination line remains dominant and the background is rather weak, which is attributed to a smaller phonon coupling constant *g*, in particular, because of a larger dot size.

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