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Temperature dependence of the zero-phonon linewidth in InAs/GaAs quantum dots

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The temperature dependence of the width Γ of the zero-phonon emission line in self-assembled InAs/GaAs quantum dots has been studied for $T < 50$ K. In single dot experiments on laterally patterned samples we find a linear increase of Γ with T, with a slope that systematically increases with decreasing size of the mesa structure. This result is to be contrasted with the absence of such a dependence in four-wave mixing on unpatterned samples. The features are shown to be consistent with a theory in which the excitons interact with phonons whose linewidths are given by scattering at surfaces due to patterning.

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Recently, the exciton-phonon interaction in quantum dots (QDs) has attracted considerable interest because it is important for processes such as scattering and relaxation. Corresponding studies offer the opportunity to obtain a deeper understanding of this fundamental interaction. They are being performed at a time when there is strong interest in exploiting quantum effects in solid-state systems for quantum information processing.[1] Scattering by phonons represents one of the most severe obstacles for implementations of true quantum devices because it destroys phase coherence. Its detailed understanding may lead to strategies of tailoring and suppressing it.

Major features of phonon-induced dephasing of excitons in self-assembled QDs have been studied in four-wave mixing experiments[2,3] from which information about decay times and resulting spectral line shapes was obtained. The line shape is given by a sharp emission feature, the so-called zero-phonon line (ZPL), that is superimposed on a broad background due to phonon-assisted transitions. This two-component behavior reflects the different time scales on which dephasing occurs. After pulsed electron-hole excitation, there is a loss of phase coherence on a scale of a few ps, which arises from dressing of the exciton by phonons and relaxation into a finite lattice distortion.[4] Further dephasing occurs on a longer time of roughly a nanosecond, which has been shown to be limited by the radiative decay of excitons at low T.[5]

Despite intense research efforts there are still important open questions, one of which we address here: the T dependence of the ZPL width for $T < 50$ K, where carrier scattering by optical phonons can be neglected. For it, seemingly contradictory results have been reported. On the one hand, in single dot spectroscopy a linear increase of the linewidth with T has been found.[6–9] On the other hand, no linear T dependence was found in recent four-wave mixing studies on arrays of QDs.[10] For these arrays an exponentially activated Bose-Einstein-like behavior describes the data.

A linear dependence of the linewidth on T might seem somewhat surprising in QDs due to their discrete energy-level structure, whereas it is well established for systems of higher dimensionality[11] that have a continuum of states into which ground-state excitons can be scattered by acoustic phonons. For extended systems, integrals over the Bose-Einstein phonon population give rise to the linear T dependence. For QDs the electronic continuum becomes discretized, resulting in level splittings of at least a few meV even for the holes due to the strong geometric confinement in self-assembled dots. Simple processes in which an exciton goes to a higher confined level due to phonon absorption would not give a linear T dependence.

Theoretically, the exciton-phonon interaction has often been treated in the framework of an independent boson model.[4,12] This model often employs a linear coupling of excitons and phonons, and it is sufficient to describe the broad phonon background. On the other hand, it gives a ZPL that does not broaden unless damping of the oscillators is introduced by hand. To explain such a broadening, several approaches have been pursued in which quadratic couplings to optical and acoustic phonons have been introduced.[13–15] They have addressed only the long decay time behavior, however. Very recently, Muljarov *et al.* developed a model that includes linear and quadratic coupling for transitions into higher lying virtual states, which considers both the short- and long-time behavior.[16] The calculated broadening of the ZPL at low T is, however, not sufficient to describe the experimental results.

The goal of the present work is to understand the origin of the very different experimental results for the T dependence of the ZPL in self-assembled QDs. In single dot experiments on patterned samples we find a linear T dependence, and we find that the strength of this dependence increases with decreasing mesa size. By detailed calculations we trace this behavior to interaction of excitons with phonons with finite lifetimes due to scattering at surfaces resulting from the patterning.

The sample used here is InAs/GaAs QDs grown by Stransky-Krastanow growth.[17] We note that this sample is from the same wafer as the one investigated in Ref. 18 as the single dot layer reference in studies aimed at understanding exciton dephasing in quantum dot molecules. The dots are capped by a 100-nm-thick GaAs layer. To obtain single dot resolution the as-grown samples were patterned by state-of-the-art electron-beam lithography and wet chemical etching. The etch depth was ~120 nm, and the lateral mesa sizes were varied down to about 200 nm.

For optical studies the QDs were placed in a helium-flow microscope cryostat. Optical excitation was done by a frequency doubled Nd-YAG laser. Since we aimed at studying single excitons only, excitation powers as low as possible were used while maintaining good signal-to-noise ratios. In this way effects of charges in the dot environment (either trapped at defects or floating around the dot) were also minimized. In addition, sample heating by laser irradiation was minimized. The laser was focused down to a spot size of ~1 μm. For dispersing the emission, we use a spectrometer that gave a large enough spectral resolution to detect the effects under study without losing too much signal (single grating monochromator with $f=0.5$ m). Detection was done by a charge-coupled device (CCD) camera.

Figure 1 shows photoluminescence spectra of three single QDs located in mesa structures of different lateral sizes L of 400, 304, and 224 nm, as indicated in each panel. The sizes are the designed ones, which after processing typically become slightly smaller but do not differ too much from these values. The sample temperature was varied from $T=5$ K up to 45 K in steps of 5 K. In each case a single sharp emission line is observed, which we attribute to neutral exciton recombination, as tested through its excitation power and wavelength dependence as well as its fine structure in magnetic field. Further, only the ZPL component in the emission is observed. Four-wave mixing experiments show that the broad background is very weak below 50 K.[2] In cw experiments, in particular those with high spectral resolution, it can hardly be resolved.[19] With increasing T the line shifts toward lower energies, and further, simultaneously its width increases. Two features are seen in these panels: (a) for a given T the linewidth increases with decreasing mesa size, and (b) the lines broaden more strongly with increasing T for smaller mesas.

These results can be seen more quantitatively in the left panel of Fig. 2, which shows the ZPL widths Γ as functions of the sample temperature. If a weak background appeared at higher T, it was subtracted for determining the linewidth. The data for the three QDs of Fig. 1 are shown together with data for a QD located in a large mesa with a lateral size of 5 μm.[20] For all T the linewidth becomes larger as the size of the mesa structure becomes smaller. The size dependence of the $T=10$ K linewidths are shown in the inset of Fig. 2, where results for many QDs are included. A systematic linewidth increase with decreasing L is observed, a result which is well-known and arises from fluctuating charge occupation at the lateral mesa sidewalls on the QDs.[7,21] These charges lead to varying Stark shifts of the confined dot levels during the time the spectra are recorded.

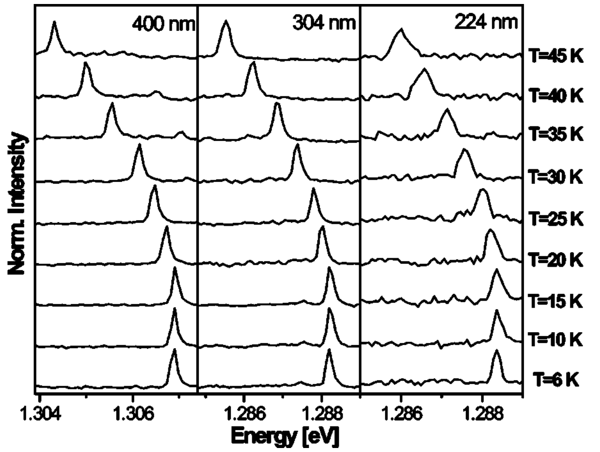

FIG. 1. Photoluminescence spectra of three InAs/GaAs single QDs at different T. The dots were located in mesa structures of different lateral sizes. Signal integration time was 120 s.

To a good approximation, a linear ZPL width increase is obtained for all four QDs. Even for the smallest T, no saturation of linewidth occurs, but it steadily decreases down to the smallest T. These results are to be contrasted with those from four-wave-mixing experiments on arrays of these QDs.[18] There it was found that the T dependence of the homogeneous linewidth at low T cannot be described by a

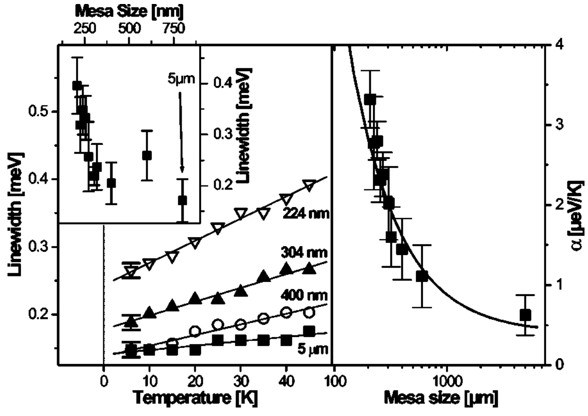

FIG. 2. Left panel: ZPL width of the emission from four different InAs/GaAs QDs as function of T. The dots were located in mesa structures of different lateral sizes as noted in the figure. The lines are linear fits to the data. The inset gives the $T=10$ K linewidth as function of lateral mesa size. Right panel: Slope of the linear increase of the emission linewidth vs the lateral size of the mesa in which the QDs were located. The symbols give experimental data, the line gives results of calculations.

linear increase. Instead, an activated behavior,

$$\Gamma(T) - \Gamma(T=0) = \frac{a}{\exp\left(\frac{E_A}{k_B T}\right) - 1}, \tag{1}$$

had to be used to model the data. Such an activated behavior seems natural for the discrete energy-level structure in QDs, at least in the energy range of the exciton ground state, which is clearly separated from the continuum of wetting layer states.

Before we turn to the discussion of the origin of these different T behaviors, the experimental picture is completed by looking at the mesa size dependence of the slope α of the linear increase in the single dot experiments: $\Gamma(T)-\Gamma(T=0)=\alpha T$. The corresponding data are shown in the right panel of Fig. 2. As expected from the spectra, α increases with decreasing mesa size. For the largest mesa with 5-μm size, α is only 0.5 μeV/K, and it increases up to more than 3 μeV/K for the 0.2-μm mesa. We note that the bars do not give the error of measurement, but rather, give the statistical variation of α that has been determined from results for several QDs for each mesa size.

Let us discuss the results. We have already considered the impact of sidewall charge occupation on the mesa size dependence of the linewidth at 10 K. The position of the QDs in the mesas, and therefore their distance from the sidewalls, is unknown and will vary from dot to dot. However, the variation of Γ (T=10 K) for a given mesa size is small, indicating that the dot position is not too important.[22] One could also imagine that the T dependence of the linewidth is determined by the charge distribution at the sidewalls, which would mean that this distribution is modified by T variations in the range studied here. Since the carriers are trapped at the sidewalls, one would expect an exponentially activated behavior and not a linear T dependence. In addition, the charge occupation would occur at deep traps, for which activation energy is so large that the occupation be changed little for the limited T range studied here. Thus, the possibility that the mesa size dependence of α could originate in variations of sidewall charge occupation can be excluded. This is supported by studies using resonant excitation below the wetting layer, which reduces the surface charging. In them, slopes are observed for the T dependence of Γ that are similar to those for nonresonant excitation.

In order to understand the T dependences observed here, we have evaluated theoretically a range of contributions to the exciton linewidths in QDs by acoustic-phonon scattering.[23] We find that the only process that gives the linear T increase of proper magnitude and has a mesa width dependence of the slope arises from exciton scattering by acoustic phonons with the phonon linewidths given by scattering at the mesa boundaries.[24–27] The interaction between excitons and phonons can be written as

$$H_{int} = \sum_{n,m} \hat{B}_n^+ \hat{B}_m \sum_{\vec{q},\sigma} M_{nm}(\vec{q},\sigma)(\hat{a}_{\vec{q},\sigma} + \hat{a}^+_{-\vec{q},\sigma}), \tag{2}$$

where $\hat{B}_m^+, \hat{a}^+_{\vec{q},\sigma}$ are creation operators for excitons in state m and acoustic phonons.[28] The full exciton spectrum is obtained from the Green function $G(t)=\langle \hat{T}\hat{B}_0(t)\hat{B}_0^+(0)\rangle$, where $\hat{T}$ is the time-ordering operator and $\hat{B}_0^+$ refers to the exciton ground state. The spectrum can be obtained by generalizing the method in Ref. 29 to nonzero-phonon widths $\gamma_{\vec{q}}$,[25] which gives

$$G(\omega) = -i\int_0^\infty dt \exp[i(\omega - \omega_0 + i\varepsilon)t]\exp[f(t)](\varepsilon \to +0). \tag{3}$$

Here $f(t)=(i\Delta-\Gamma/2)t/\hbar-\tilde{f}(t)$, and

$$\frac{\Gamma}{2} = \sum_{\vec{q},\sigma} |M_{00}(\vec{q},\sigma)|^2 \gamma_{\vec{q}}(2n_{\vec{q},\sigma}+1)(\hbar^2\omega^2_{\vec{q},\sigma} + \gamma^2_{\vec{q},\sigma})^{-1}. \tag{4}$$

Δ is the real exciton self-energy shift and $\tilde{f}(t)$ is similar to the form given in Ref. 25. The exciton properties were calculated variationally[30] for cylindrical QDs of $In_{0.6}Ga_{0.4}As$ in GaAs with a height of 6 nm and a radius of 10 nm.[31] We find that the dominant coupling between excitons and the acoustic phonons is from the deformation potential.[28] Coulomb correlation effects on the matrix elements M_{nm} were found to be small.

We have examined a number of contributions to the full wave-vector-dependent phonon linewidth $\gamma_{\vec{q}}$, including phonon anharmonicity, and alloy and impurity scattering, and we find that boundary scattering is the dominant contribution in the present structures at low T.[23] The Ziman theory of boundary scattering[32] was used for $\gamma_{\vec{q}}$ giving $\gamma_{\vec{q}}=\gamma^0_{\vec{q}}[1-p(\vec{q})]/[1+p(\vec{q})]$, where the specularity of the surface $p(q)=\exp(-4\pi\delta^2q^2)$ is given in terms of a surface corrugation (or "asperity") parameter δ, which is a measure for the roughness of the interfaces surrounding the quantum dot. The phonon lifetime $\gamma^0_{\vec{q}}$ for purely diffusive scattering was obtained from a Boltzmann equation in the relaxation time approximation. For the present systems, we assume $\delta\sim 1$ nm for the top molecular-beam epitaxy (MBE) surface and $\delta\sim 12$ nm for the lateral etched sidewalls. This lateral asperity is in reasonable accord with roughness fluctuations seen in scanning electron microscopy. With these parameters the calculated results shown in the right panel of Fig. 2 are in good agreement with the measured dependence of the exciton linewidths.

The scattering of phonons from the lateral sidewalls becomes negligible for mesa sizes on the order of a few microns. For these sizes scattering from the top surface, which is 100 nm from the dot, dominates and gives a value of $\alpha\sim 0.5$ μeV/K. This is an MBE grown surface and is smoother than the lateral etched surfaces, which is reflected in the choice of a small surface asperity parameter. Phonon scattering from the lateral etched surfaces gives the increase of α for decreasing mesa size.

In summary, we have studied the origin of the T dependence of the ZPL width in InAs/GaAs QDs. From comparison of experiment and theory, the linear T dependence can be traced to the interaction of confined excitons with phonons whose lifetimes are given by scattering from nearby surfaces arising from lateral fabrication. We suggest that the absence of a contribution to the exciton linewidth linear in T, as seen in experiments in arrays of quantum dots, results from the absence of mesalike structures used in single dot experiments. Conversely, such fabrication is often envisioned for quantum dot implementations in quantum information processing, and it should be kept in mind that it can lead to decoherence in optical processes. An improvement of nanofabrication to obtain smoother lateral surfaces will reduce the phonon scattering.

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