Temperature dependent high resolution resonant spectroscopy on a charged quantum dot

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We present temperature dependent high resolution resonant optical spectroscopy on a single, negatively charged InGaAs quantum dot. We performed laser transmission measurements yielding the natural linewidth of the excitonic ground state transition of a quantum dot in a temperature range from 4.2 K up to 25 K. Here, we describe the linewidth evolution and the temperature induced red shift of the resonance energy with simple models based on the exciton–phonon coupling in the quantum dot. The resonant spectroscopy measurements are complemented with results from non-resonant PL measurements on the very same quantum dot. Here we observe a simple linear behavior of the linewidth according to an effect of a fluctuating environment.

1 Introduction Self-assembled semiconductor quantum dots (QDs) exhibit the remarkable spectral feature of optical resonance linewidths $(1-3\,\mu\text{eV})$ far below the thermal energy $(362\,\mu\text{eV}$ at 4.2 K) [1]. The small linewidth corresponds to the long coherence time of the exciton in the quantum dot of up to ≈ 1 ns [2]. The strong confinement of the excitons in the QD and the resulting large intraband level spacing of several meVs [3–5] suppress the interaction of the electronic states in the QD with the solid crystal lattice of the host material. This remarkable feature shows a close resemblance between atom and QD optics. Many of the proposed applications of QDs in the fields of quantum communication and quantum information processing are based on these atom like optical properties. However, despite the importance of dephasing mechanisms for the application of solid state quantum systems in novel quantum-electronic devices, there is still no consistent microscopic picture of their temperature dependence.

The optical spectra of strongly confined electronic systems (QDs or molecules) are expected to show two spectral features. The so called zero-phonon line (ZPL) is temperature broadened due to acoustic phonon scattering, with a certain threshold or activation energy which corresponds to a phonon mediated excitation of the electron or hole into an excited state. These mechanisms correspond to pure dephasing and hence to a lorentzian lineshape of the resonance. Apart from the ZPL, there are phonon sidebands which correspond to a mixing of the excitonic states with phonon modes: the absorption or emission of a photon in the QD leading to creation or recombination of an exciton involves the emission or absorption of acoustic phonons. In molecular spectroscopy, this is known as the Frank–Condon principle.

In QD spectroscopy the ZPL and phonon sidebands have been observed in non-linear spectroscopy (four-wave-mixing) on an ensemble of InGaAs dots [2]. However, only at temperatures above 50 K the phonon sidebands contributed significantly to the spectra [2]. In PL experiments, the observation of phonon sidebands was reported on single CdTe QDs [6] which exhibit a stronger coupling to phonons than III–V semiconductors. In PL spectroscopy on single InGaAs QDs however, only a linear increase of the linewidth of the ZPL was reported without any significant phonon sidebands [7–10]. This can most probably be
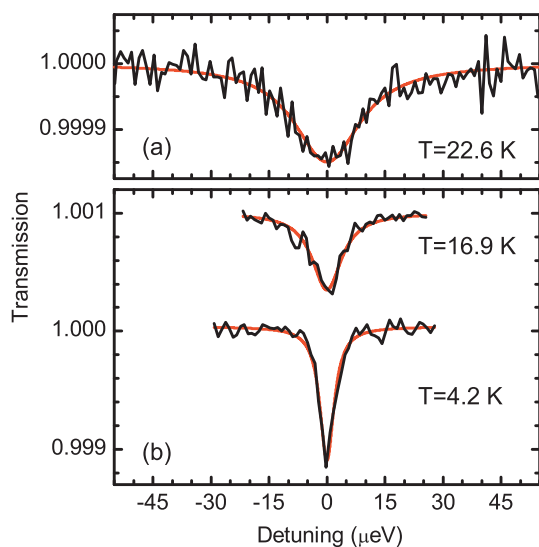
Figure 1 (online colour at: www.pss-b.com) Transmission spectra of a single, negatively charged QD at different temperatures. In (a) the temperature was 22.6 K, the resonance line is broadened and hence the contrast is reduced. In (b) two spectra for a temperature of 4.2 K and 16.9 K are plotted. The offset for the spectrum at 16.9 K was added for clarity. The energy axis is corrected for the red shift due to the heating. The resonance energy is 1.280497 eV at 4.2 K, 1.280372 eV at 16.9 K and 1.280007 eV at 22.6 K. The laser power was ≈90 nW for all measurements presented in this paper.

explained by thermally activated fluctuations of photo created charges in the vicinity of the QD [9, 10].

We present resonant, high resolution transmission spectroscopy on a single, negatively charged QD in the temperature range of 4.2–25 K [1, 11]. We study the linewidth and resonance energy evolution with temperature of two different QDs. Due to the resonant excitation, we can reduce the contribution of fluctuations of optically excited charge carriers in the hetero structure to the resonance and hence, we can directly study the homogeneous broadening of the ZPL.

2 The sample The QDs under investigation were embedded in a field effect device [3], which allowed for a controlled charging of the QD with single electrons [12]. The device consists of a highly n-doped back contact, separated from the QD-layer by a tunnel barrier of 25 nm i-GaAs. The InGaAs QDs were grown including an annealing step shifting their interband optical transition to wavelengths around 950 nm. The QDs were capped with a 10 nm thick layer of i-GaAs separating them from an AlAs/GaAs superlattice which prevents a current flow in the device. On the top of the sample a semitransparent metallic top gate was evaporated to complete the field effect device. By applying a bias between the back contact and the top gate, the conduction and valence band edges are tilted following a simple leverarm argument [12, 13]. The fermi energy is pinned to the conduction band due to the

high doping of the back contact. By shifting the electronic states of the QD below the fermi energy, electrons will tunnel one by one into the QD, in compliance with the Pauli exclusion principle and the Coulomb interaction [13]. Furthermore, the electric field in the device induces an energy shift of the excitonic resonances of the QD, the Stark effect. To overcome the fine structure splitting due to electron−hole spin interaction, we choose to charge the QD with one electron and perform all temperature dependent experiments on the unpolarized, single resonance line of the negatively charged QD (X^{1-}) [1, 13, 14].

3 Single QD spectroscopy The sample is mounted in a diffraction limited, fiber based, confocal microscope which is immersed in a liquid He bath cryostat [18]. Thermal equilibration of the sample with the liquid He is provided by a small amount of He exchange gas. On the metallic sample holder two Allen−Bradly resistors were glued such that they have the same distance between them and the sample. One of the resistors was used for heating while the other was measured to determine the temperature of the sample. The local heating allowed to keep the photodetector at a constant temperature providing a constant sensitivity.

The sample is positioned in the focal plane of the confocal objective (NA = 0.65) and the transmission of a narrow band laser (Sacher, TEC-500-0960-30) through the sample is measured by a photodiode mounted directly behind the sample. In Fig. 1(b) the lower spectrum represents a typical transmission spectrum, measured at 4.2 K [1, 11, 13, 15, 16].

Care was taken to use low enough laser power (≈ 90 nW) to remain in the linear regime of QD absorption [17].

For PL measurements the emitted light from the QD is guided out of the microscope through the same single mode fiber as used for illumination and dispersed in a 50 cm grating spectrometer with a resolution of ≈45 μeV [18, 19].

4 Temperature evolution of the X^{1-} resonance energy The temperature dependent band gap of semiconductor materials leads to a frequency shift of optical interband transitions. For bulk semiconductors this red shift of the transition energy with increasing temperature can be described by the empirical Varshni model [20]:

$$E_g(T) = E_g(T = 0) - \frac{\alpha T^2}{\beta - T}, \tag{1}$$

where E_g is the temperature dependent band gap and α and β are obtained by fitting the equation to the data. For GaAs these parameters are $E_g(T = 0) = 1.5216$ eV, $\alpha = 8.871 \times 10^{-4}$ eV/K^2 and $\beta = 572$ K while for InAs: $E_g(T = 0) = 0.426$ eV, $\alpha = 3.189 \times 10^{-4}$ eV/K^2 and $\beta = 93$ K [20]. The evolution of the band gap of GaAs and InAs are plotted in Fig. 2(a) by the dotted and dashed lines.
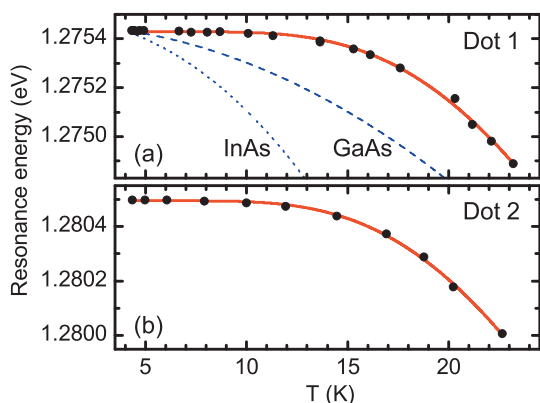
Figure 2 (online colour at: www.pss-b.com) Resonance energy of the X^{1-} transition for two different QDs as a function of the temperature of the crystal. The dots are the measured resonance energies, the solid lines are Eq. (2) fit to the data points. The dashed and dotted lines represent the temperature band gap evolution of GaAs and InAs shifted to the X^{1-} resonance energy. The two dots were chosen randomly from the ensemble of dots on the sample.

The energies were shifted by an offset to match the energy window given by the QD resonance. The empirical Varshni relation as given above can not describe the observed behavior for the red shift of the X^{1-} resonance [21]. There is no common theory on the temperature shift of the QD resonance, although it is an important tool to tune the transition energy of a QD in cases where the QD can not be embedded in a field effect device (e.g. for coupling a QD transition to a cavity mode [22]) [21].

We observe almost no shift of the resonance energy for temperatures below 10 K. Above 10 K the resonance shows a red shift with a linear asymptotic behavior with temperature. The simple Bose−Einstein type model successfully describes the temperature evolution of interband transitions of bulk semiconductors as well as QDs [21, 23, 24]:

$$E_{\mathrm{res}}(T) = E_{\mathrm{res}}(T=0) - S\hbar\omega\left(\coth\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right) - 1\right). \qquad (2)$$

Here S is a dimensionless coupling constant and the coth term describes the electron–phonon coupling to phonons of energy $\hbar\omega$, according to the Bose−Einstein occupation number of the phonon mode. We find a perfect agreement upon fitting this relation to our data sets (Fig. 2). The parameters are: $S_{\mathrm{Dot1}} = 0.9$ and $\hbar\omega_{\mathrm{Dot1}} = 7.99$ meV for Dot 1 (Fig. 2(a)) and $S_{\mathrm{Dot2}} = 0.8$, $\hbar\omega_{\mathrm{Dot2}} = 7.61$ meV for Dot 2 (Fig. 2(b)). The found energies are close to the TA phonon energies at the L-point for bulk GaAs, $\hbar\omega_{\mathrm{TA}}(\mathrm{L}) = 7.7$ meV [25].

5 Temperature broadening of the X^{1-} resonance Additional to the red shift of the transition frequency we observe a broadening of the resonance line. In Fig. 1 spectra of the X^{1-} of Dot 1 (see Fig. 2) for three dif-

ferent temperatures are plotted. It is obvious that the linewidth increases with temperature while the strength of the resonance is reduced. The area, however, remains more or less constant over the observed temperature range. The spectra remain lorentzian shaped within the signal to noise, and no phonon sidebands are detected. This gives rise to the interpretation that the mixing of excitonic states with phonon modes is very weak in III−IV semiconductor nano-structures at low temperatures.

In Fig. 3 the linewidths obtained from lorentzian fits to the spectra are plotted against the temperature for the two different quantum dots. We find a weak temperature dependence of the linewidth for low temperatures. At temperatures above ≈15 K the slope changes indicating some thermal activation of a dephasing mechanism for the exciton. Similar behavior has been observed on ensembles of InGaAs QDs and quantum wells [2, 26]. We use a simple model for describing our observation based on a linear term and a term representing the occupation of a phonon mode similar to the model used for the resonance shift:

$$\Gamma = \Gamma_0 + aT + \frac{b}{\exp\left(\dfrac{\hbar\omega}{k_{\mathrm{B}}T}\right) - 1}. \qquad (3)$$

Interestingly we can describe our data reasonably well with this model by using the same phonon energy $\hbar\omega$ as obtained from the resonance energy shift (Fig. 3). The other parameters for the two dots are: $a_{\mathrm{Dot1}} = 0.05$ µeV/K and $b_{\mathrm{Dot1}} = 94$ µeV for Dot 1 and $a_{\mathrm{Dot2}} = 0.09$ µeV/K and $b_{\mathrm{Dot2}} = 327$ µeV for Dot 2. The parameter b describes the coupling of the exciton to the phonon mode which we find to differ by a factor of three from one dot to the other. The coupling of electronic or excitonic states to phonons is mediated mostly by the piezo electric properties of the system, an effect which is strongly dependent on the strain in the material. In self-assembled QDs the strain is expected to vary from dot to dot indicated already by the differences in magnitude and orientation of the fine structure splitting [27].
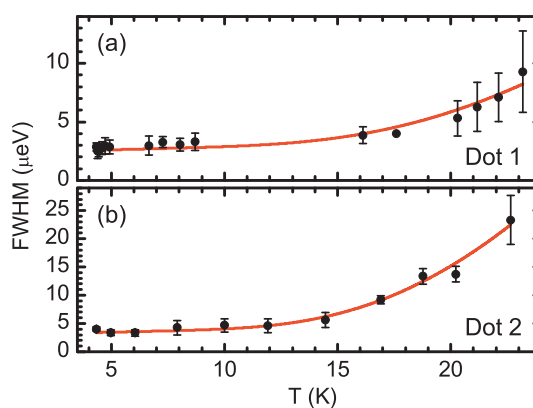
Figure 3 (online colour at: www.pss-b.com) Full width at half maximum of the lorentzian resonance lines of the two X^{1-} from Fig. 2 as a function of temperature. The lines are Eq. (3) fit to the data points.

The linear contribution to the linewidth given by *a* we interpret as an effect of the optically activated fluctuating environment as discussed by Favero et al. [9]. Originally this effect was reported on non-gated samples in PL measurements under non-resonant excitation. In such experiments the non-resonant excitation creates free excitons which can be trapped in local potential minima [10]. The charges are then activated by the thermal energy leading to an inhomogeneous broadening of the ZPL. In the presented experiment such effects are expected to be weak. First, due to the screening of potential fluctuations in the vicinity of the QD due to the gate and back contact. Second, the laser excites no charges in the wetting layer or the bulk GaAs since it is only resonant to the QD transitions. However, spectral fluctuations have been observed in resonant laser spectroscopy experiments [1, 16, 17]. Very recently we reported a background absorption for laser energies far below the GaAs band edge or wetting layer transitions even when there is no QD in the laser focus [5]. These observations give rise to the interpretation that the linear temperature dependence of the linewidth is indeed due to spectral fluctuations in the solid state matrix. Whether these fluctuations are optically activated or intrinsic to the semiconductor hetero structure can not be determined in this experiment.

6 PL measurements of the temperature broadening In order to verify our interpretation of the linear temperature broadening we performed PL measurements of the linewidth temperature dependence on Dot 2. The observed linewidths are plotted in Fig. 4. The spectral resolution is limited by the spectrometer to $\approx 45\,\mu eV$. We observe a more or less linear increase of the linewidth with temperature with a slope of $1.35\,\mu eV/K$. This corresponds to the slope observed on the X^{1-} of similar QDs in PL [8] $(1.2-4.4\,\mu eV/K)$ and [9] $(1.3-3.5\,\mu eV/K)$. These values are at least one order of magnitude larger than those observed in resonant spectroscopy. The experimental details can be found in [13].
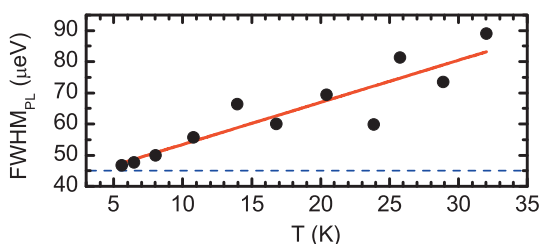
Figure 4 (online colour at: www.pss-b.com) Full width at half maximum of the lorentzian resonance lines of the X^{1-} of Dot 1 measured in PL. The excitation was done with a 822 nm laser diode exciting the wetting layer. The spectral resolution of the 50 cm grating spectrometer used for dispersion was $\approx 45\,\mu eV$ as indicted by the dashed line. The line is a linear fit to the data points.

7 Conclusion We presented resonant and non-resonant optical spectroscopy measurements of the linewidth and the resonance energy of single negatively charged QDs. We find a saturation of the linewidth and resonance energy at low temperatures (<10 K) for resonant excitation, while for non-resonant PL measurements the linewidth follows a linear increase with temperature. We interpret the activation of the red shift of the resonance as well as the temperature broadening by interaction with phonons, most likely TA(L). The small linear contribution to the linewidth evolution we interpret as an inhomogeneous broadening due to temperature activated spectral fluctuations, a contribution which we find to be at least a factor 10 smaller than for non-resonant excitation measurements on the very same QD.

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