

## Resonant saturation laser spectroscopy of a single self-assembled quantum dot

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### Abstract

We performed high resolution resonant laser spectroscopy on a single self-assembled quantum dot (QD) at liquid He temperatures. We explore the two-level nature of the QD excitonic transition through the investigation of its behavior as a function of the laser power. The quantum ground state exciton absorption peak size diminishes with increasing power while its width increases. Fitting these dependencies to the predictions of a two-level atom model we extract unambiguously a radiative lifetime of 660 ps, a residual collisional broadening about 0.18  $\mu\text{eV}$  as well as the spectral fluctuation 1.3  $\mu\text{eV}$ . We find that at high power the line width of the exciton absorption is essentially given by the Rabi frequency.

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The atom-like optical behavior of self-assembled quantum dots (QD) makes them interesting candidates for quantum optics and quantum information processing. In particular the two-level nature of their ground state exciton provides a crucial prerequisite to use them as quantum bits [1] and non classical light sources for quantum cryptography [2] or QD lasers [3]. High resolution, resonant laser spectroscopy [7] gives direct access to the two-level properties of the excitonic transitions in a single QD.

Here we present a detailed analysis of the power dependency of the line width and peak intensity, which are obtained by resonant laser spectroscopy in transmission at liquid He temperatures. We study a single self-assembled InGaAs QD grown by molecular beam epitaxy in the Stransky–Krastanow mode. The QD is embedded in a field effect structure [4] allowing discrete charging of the

dot with single electrons through the control of a gate voltage [5].

We investigated the QD charged with a single resident electron, so that the excitonic transition was that of a trion or  $X^{1-}$ . Unlike the neutral exciton transition ( $X^0$ ) which has a fine structure splitting, the  $X^{1-}$  transition manifests itself through a single Lorentzian peak at zero magnetic field [6]. Resonant laser transmission spectroscopy has a typical resolution limited by the line width of our tunable external cavity laser diode which was typically 0.01  $\mu\text{eV}$ . Details of this technique can be found in Ref. [6].

In Fig. 1 we show two typical transmission spectra for two different laser powers. We choose to plot the contrast in the differential transmission ( $\alpha$ ), since it will appear later in the text as the variable used in our analysis. Increasing the laser power from 0.5 nW to 3  $\mu\text{W}$  we find that the line width increases from 3.5 to 40  $\mu\text{eV}$  while at the same time the contrast drastically diminishes by a factor of 100. To describe this behavior, we calculated the optical resonant transmission properties of the exciton using a two-level

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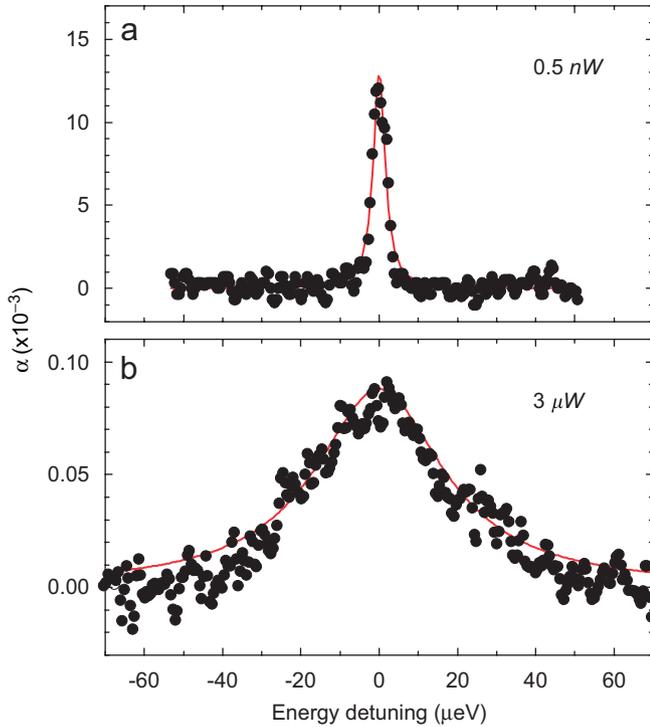


Fig. 1. Two typical transmission spectra of the  $X^{1-}$  transition at two different laser powers. The upper spectrum (a) was obtained by illuminating the QD with 0.5 nW, the lower one (b) with 3  $\mu$ W. The lines correspond to Lorentzian line fits with an FWHM of 3.5  $\mu$ eV (a) and 40  $\mu$ eV (b).

model, as known from atom optics [8,9]. In all measured dots we observed that even at lowest laser power ( $\sim 0.1$  nW) the measured line widths were about a factor two broader than expected from lifetime measurements obtained on similar QD samples [10]. This excess broadening is believed to originate from dot-level fluctuations through electron and host-material interactions. We treat these fluctuations in two different limits, dependent on their timescale. Fluctuations that occur on a timescale shorter than the exciton lifetime contribute to a homogeneous collisional-like broadening accounted for in typical two-level atom models [8]. Fluctuations with larger timescales but still small compared to the measurement time (here  $\sim 1$  s) leads to inhomogeneous broadening. Such broadening is generally not expected to be Lorentzian, however, most of our data show invariably Lorentzian lines. In all cases broadened lines have a weaker peak than expected from an ideal two-level system. The differential transmission line shape for a homogeneous broadened excitonic transition is given by [8]

$$\frac{\Delta T}{T} = \alpha_0 \frac{\gamma_0^2}{\delta^2 + \gamma_0^2 + \gamma_0/2\gamma_{sp}\Omega^2}. \quad (1)$$

Here  $\alpha_0$  is the contrast in the limit of vanishing laser power  $P$ .  $2\gamma_0$  is the effective full width at half maximum of the resonance likewise at vanishing  $P$ . In a collisional broadened two-level system  $\gamma_0 = \gamma_{sp} + \gamma_{coll}$  where  $1/\gamma_{sp}$  is

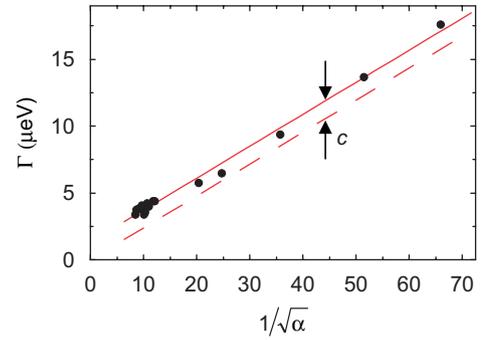


Fig. 2. Plot of the measured line width as a function of the square root of the differential transmission contrast. The line is a linear fit to the data with an offset by a constant  $c$  from the theoretically expected dashed line.

the exciton radiative lifetime through spontaneous emission and  $\gamma_{coll}$  is the contribution of the collision broadening.  $\delta = \hbar\omega_L - \hbar\omega_X$  is the detuning between the photon energy  $\hbar\omega_L$  and the exciton energy  $\hbar\omega_X$ . The Rabi frequency  $\Omega^2 = 2\alpha_0\gamma_0 P/\hbar\omega_L$ . The oscillator strength of the resonance is proportional to  $\alpha_0\gamma_0$  implying that  $\alpha_0\gamma_0 = \alpha_{sp}\gamma_{sp}$  where  $\alpha_{sp}$  would be the resonance contrast in an ideal two-level system. This contrast is  $\alpha_{sp} = 3(\lambda/n)^2/2\pi A$  which only depends on the laser wavelength at resonance  $\lambda$  and the area  $A$  of the focussed laser spot in the plane of the dot.  $n$  is the refractive index of the QD host material.

The resonance contrast  $\alpha$  is obtained at zero detuning ( $\delta = 0$ ):

$$\alpha = \frac{\alpha_0}{1 + pP/\hbar\omega\gamma_{sp}\alpha_0}. \quad (2)$$

Likewise the power dependency of the full width at half maximum ( $\Gamma$ ) is given by

$$\Gamma = 2\gamma_0 \left( 1 + \frac{pP}{\hbar\omega\gamma_{sp}} \alpha_0 \right)^{1/2}. \quad (3)$$

The QD is illuminated through an Al shadow mask with a circular aperture of diameter  $d = 300$  nm. The transmitted light is collected using a 4 mm<sup>2</sup> PIN photodetector placed 1.5 mm behind the sample. In such a geometry and because of diffraction the transmitted photons are collected only in part and we calculated that the power  $P_{det}$  at the detector is related to  $P$ , the power experience by the dot, through  $P = pP_{det}$ <sup>1</sup> with here  $p = 17$ .

We obtain both the line width and the amplitude of the resonance independently and using Eqs. (2) and (3) we can eliminate their explicit power dependency in order to get  $\Gamma = \Gamma_0 \sqrt{\alpha_0/\alpha}$  which we plot in Fig. 2 along with the data. In order to fit the data, we found it necessary to add a constant  $c = 1.34$   $\mu$ eV to the line width. Such an additional constant is not expected in a collisional broadened two-level model but would be the slow contribution to dot level

<sup>1</sup> $p = (1 - J_0(2\pi nr \sin(\theta)/\lambda) + J_0(2\pi nr \sin(\theta)/\lambda))^{-1}$  [11].  $J_n$  is the  $n$ th Bessel function and  $\theta = 0.15$  is the illumination angle of the detector, seen from the aperture with radius  $r$ . The diffraction index of GaAs is  $n$  and the laser wavelength is  $\lambda$ .

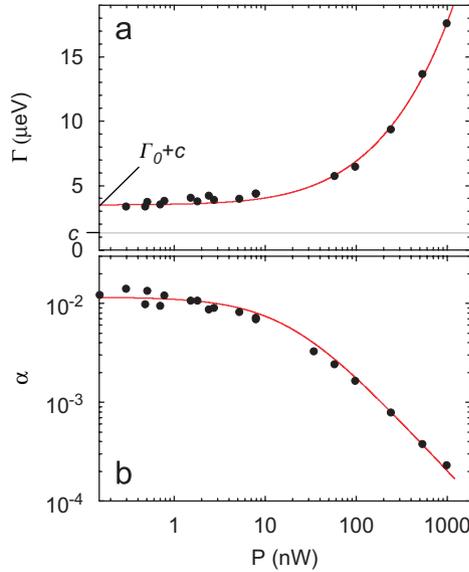


Fig. 3. Plots of the measured line width (a) and contrast (b) of the excitonic transition. The lines are Eqs. (2) and (3) fit to the data. The power axis has been calibrated by the obtained geometry factor  $p$ .

fluctuations. At the lowest power we measure an effective line width of  $3.5 \mu\text{eV}$  so we deduce that  $\Gamma_0 = 2.36 \mu\text{eV}$ . The linear dependency gives a slope  $\Gamma_0 \sqrt{\alpha_0} = 0.24 \mu\text{eV}$  from which we deduce  $\alpha_0 = 0.012$  which also turnout to be the contrast directly measured at very low power.

Now we turn to the explicit power dependency the resonance contrast and the line width. Using  $\Gamma_0$  obtained above, we fit Eq. (2) to the measured contrast and using Eq. (3) we fit  $\Gamma + c$  to the measured line width (see Fig. 3). The only fit parameters are now  $\alpha_0$  and  $\gamma_{\text{sp}}$  which we find to be  $\alpha_0 = 0.02$  and  $\gamma_{\text{sp}} = 1.0 \mu\text{eV}$ . We deduce therefore a  $\gamma_{\text{coll}} = 0.18 \mu\text{eV}$ . We notice a discrepancy between  $\alpha_0 = 0.02$  obtained here and  $\alpha_0 = 0.012$  inferred earlier from Fig. 2. Here as well the effect of the slow components of the level fluctuations are likely to explain this discrepancy and  $\alpha_0 = 0.02$  would be the contrast without time-averaged inhomogeneous broadening.

From the measured spectra we can furthermore analyze the signal-to-noise ratio. The noise in our experiment is dominated by the laser shot noise as well as a weak detection dark noise which we systematically subtract. The shot noise is proportional to the square root of the number of photons per unit time ( $\dot{N} = P_{\text{det}}/\hbar\omega_L$ ), while the signal is given by  $\text{signal} = P_{\text{det}}\alpha$ . Hence the signal-to-noise ratio is according to Eq. (2):

$$\frac{\text{Signal}}{\text{Noise}} = \frac{\sqrt{P_{\text{det}}/\hbar\omega}}{(1/\alpha_0) + pP_{\text{det}}/\hbar\omega\gamma_{\text{sp}}}. \quad (4)$$

The measured signal-to-noise ratio is plotted in Fig. 4 as a function of the laser power  $P$  at the detector. In Eq. (4) we used  $\alpha_0 = 0.012$ ,  $p = 17$  and  $\gamma_{\text{sp}} = 1.0 \mu\text{eV}$ .

The presented data strengthens the picture of the QD as an artificial atom, that can be modeled by a simple two level model. We found that very conveniently the radiation

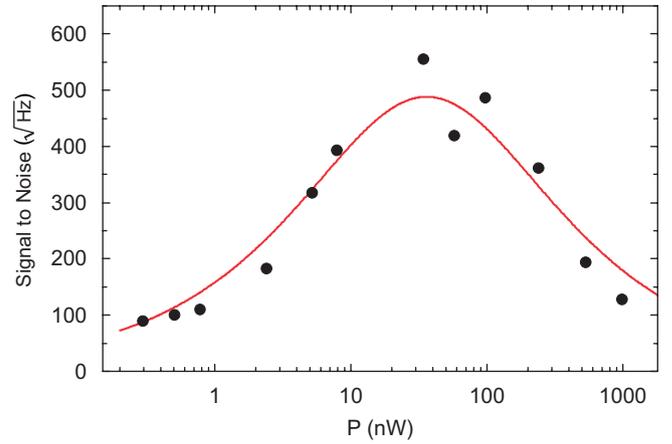


Fig. 4. Signal-to-noise ratio is plotted with a fit of Eq. (4) to the data points, represented by a line.

limited lifetime of the QD ground state exciton is obtained through the analysis of the exciton resonance saturation and power broadening even when collisional and inhomogeneous broadening are taken into account. Furthermore high resolution resonant laser spectroscopy highlights the limitations of the two-level model to describe the QD exciton ground state. Here we revealed that collisional broadening is only a tenth of the spontaneous emission broadening which, given the solid state nature of the device, is surprisingly small. The slow spectral fluctuation accounts for half the line width broadening and its origin still needs to be elucidated. Should one reach the limit of single exciton absorption, spectral fluctuations are not expected to play a role in the line width and contrast of a scattering signal. Recently, more efficient optical measurement schemes have been presented [12,13]. There, the contrast of differential transmission spectroscopy experiments has been dramatically increased opening a path towards single exciton absorption.

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## References

- [1] A. Zrenner, et al., Nature 418 (2002) 612.
- [2] N. Akopian, et al., Phys. Rev. Lett. 96 (2006) 130501.
- [3] H. Ishikawa, et al., J. Vac. Sci. Technol. A 16 (1998) 794.
- [4] H. Drexler, et al., Phys. Rev. Lett. 73 (16) (1994) 2252.
- [5] R.J. Warburton, et al., Nature 405 (2000) 926.
- [6] A. Högele, et al., Physica E 21 (2004) 175.
- [7] A. Högele, et al., Phys. Rev. Lett. 93 (2004) 217401.
- [8] R. Loudon, The Quantum Theory of Light, Oxford University Press, Cambridge, 1995.
- [9] K. Karrai, et al., Superlattices Microstructures 33 (2003) 311.
- [10] Smith, et al., Phys. Rev. Lett. 94 (2005) 197402.
- [11] M. Born, E. Wolf, Principles of Optics, Cambridge University Press, Cambridge, 2005.
- [12] S. Seidl, et al., Phys. Rev. B 72 (2005) 195339.
- [13] A.N. Vamivakas, et al., Nano Lett. 7 (2007) 2892.