Available online at www.sciencedirect.com
SCIENCE DIRECT®
Physica E 21 (2004) 175–179
PHYSICA E
www.elsevier.com/locate/physe

Exciton fine structure splitting of single InGaAs self-assembled quantum dots

A. Högele[a,*], B. Alèn[a,b], F. Bickel[a], R.J. Warburton[c], P.M. Petroff[d], K. Karrai[a]

[a] *Center for NanoScience und Sektion Physik, Ludwig-Maximilians-Universität München, Geschwister-Scholl-Platz 1, Munich D-80539, Germany*
[b] *Instituto de Ciencia dels Materials—Departamento de Física Aplicada, Universidad de Valencia, E46071 Valencia, Spain*
[c] *Department of Physics, Heriot-Watt University, Edinburgh EH14 4AS, UK*
[d] *Materials Department, University of California, Santa Barbara, CA 93106, USA*

Abstract

We show how the resonant absorption of the ground state neutral exciton confined in a single InGaAs self-assembled quantum dot can be directly observed in an optical transmission experiment. A spectrum of the differential transmitted intensity is obtained by sweeping the exciton energy into resonance with laser photons exploiting the voltage induced Stark-shift. We describe the details of this experimental technique and some example results which exploit the $\sim$1 μeV spectral resolution. In addition to the fine structure splitting of the neutral exciton and an upper bound on the homogeneous linewidth at 4.2 K, we also determine the transition electric dipole moment.

PACS: 81.07.Ta; 78.67.Hc; 78.55.Cr; 71.35.Cc; 71.70.Gm

Keywords: Quantum dots; Homogeneous linewidth; Fine structure splitting; Absorption

Self-assembled semiconductor quantum dots (QDs) have been proposed to serve as fundamental components of the solid-state quantum computer [1] or nonclassical light sources [2]. High efficiency single photon devices have been demonstrated from QDs embedded in a micropost cavity [3]. Crucial to such investigations is the knowledge of the excitonic decoherence time as well as the strength of the coupling to electromagnetic fields, the transition dipole moment. These physical properties can be accessed directly by optical absorption spectroscopy. Absorption experiments on single ions [4] and single molecules [5] have been demonstrated. Recently, the absorption of single QDs formed naturally in GaAs/AlGaAs quantum wells has been measured using near-field microscopy or submicron aperture masks [6,7]. A similar approach was made with nonlinear absorption spectroscopy on single self-assembled InGaAs QDs [8] but showed poor signal.

In this paper, we demonstrate a new absorption experiment which exploits the Stark effect in the energy of the QD exciton. This technique allows us to measure directly single dot excitonic resonances with high spectral resolution. We explain in detail

* Corresponding author. Tel.: +49-89-2180-3586; fax: +49-89-2180-3182.
E-mail address: hoegele@lmu.de (A. Högele).

1386-9477/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/j.physe.2003.11.007

how these experiments are performed. Example results are presented on the ground state exciton of a single self-assembled InGaAs QD. These results complement the results we obtained on the first excited exciton state [9].

The self-assembled InGaAs QDs investigated here were grown using solid source molecular beam epitaxy and embedded in a field effect heterostructure [10]. A highly n doped GaAs layer acts as the back contact. Above is 25 nm of undoped GaAs tunnel barrier and the InGaAs QDs. The subsequent overgrowth with 30 nm GaAs and 120 nm short period AlAs/GaAs superlattice defines the total separation of 150 nm between the QDs and the semitransparent NiCr gate electrode deposited on the surface. By applying a gate voltage V_g with respect to the back contact, QDs can be filled sequentially with up to 3 electrons within a voltage range of < 700 mV depending on the individual QD confinement potential. Additionally, QDs can be populated with excitons generated by an external light source. For a single QD, the degree of charge and excitonic population can be monitored by photoluminescence (PL) spectroscopy [11,12].

In our experiment we focus on the neutral exciton state X^0. By resonant absorption of laser light, the QD vacuum state $|0\rangle$ is transformed into the ground state of the neutral exciton $|X^0\rangle$, as indicated in Fig. 1a. The energy splitting Δ_1 between the ground state excitonic energy levels E^x and E^y accounts for the electron–hole exchange interaction in a weakly asymmetric dot. In order to perform interband optical transitions on the ground state of the selected QD, the laser wavelength was set to match the X^0 recombination energy obtained from PL measurements. The fine tuning of the excitonic energy into resonance is achieved using the Stark effect by sweeping the gate voltage (Fig. 1b).

For both the PL and the absorption experiment, the sample was mounted above an unbiased Ge p–i–n photodiode (Thorlabs, FDG05) in a home built fiber based low temperature confocal microscope and cooled in a cryostat to liquid helium bath temperature (Fig. 2). Lateral translation as well as positioning of the sample into the focal plane were performed with a low temperature suitable XYZ-positioner (Atto Cube Systems, ANP-XYZ-100). The miniature microscope has an aspheric objective with numerical aperture of N.A. = 0.55 and spatial resolution of 1.3 μm (full-width at half-maximum at 1.06 μm probing wavelength).

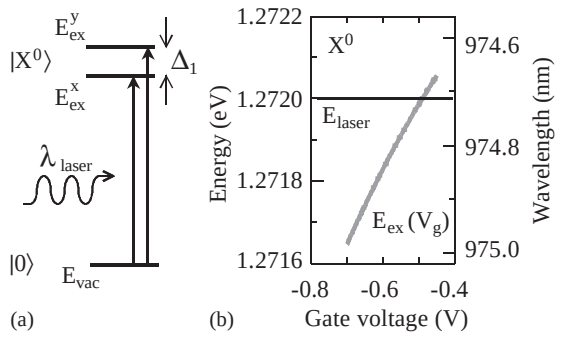

Fig. 1. (a) Schematics of the quantum dot transition from the vacuum state to the neutral excitonic ground state by laser photon absorption. The exitonic energy level is split by fine structure energy Δ_1. (b) Measured gate voltage dependence of the PL energy for the neutral exciton X^0 and the resonance crosspoint with the laser are shown.

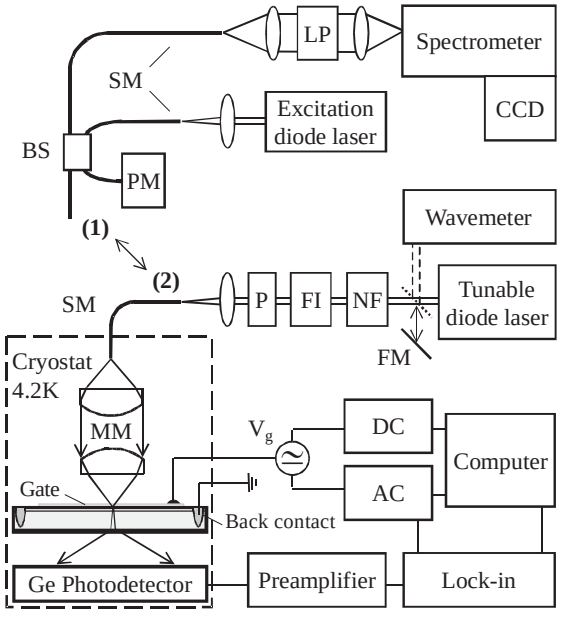

Fig. 2. Schematic of the setup: long wave pass filter (LP), single-mode fiber (SM), fiber based beam splitter (BS), power meter (PM), miniature microscope objective (MM), polarizing components (P), Faraday isolator (FI), neutral density filters (NF), flip-mounted mirror (FM). The arrow between positions (1) and (2) indicates the microscope fiber switched for the luminescence measurements and the absorption experiment, respectively.

The gate voltage V_g is defined with respect to the grounded back contact by superimposed signals from the DC-calibrator Knick S252 and the AC synthesized

function generator and attenuator (Stanford Research Systems, DS335). A $\frac{1}{100}$ attenuator is connected to the output channel of the function generator in order to reduce the modulation amplitude. The Ge photodetector signal is amplified by a low-noise current preamplifier. Two types of preamplifiers were used: Stanford Research Systems, SR570 and Ithaco, Model 1211, the latter having an order of magnitude higher dynamic range for amplifications above 10^8 V/A. The amplified DC part of the photo-signal was recorded with a computer. Demodulation and further amplification of the AC part is performed by an EG& G 7260 lock-in amplifier supplied with the function generator reference signal. Again a computer was used for data acquisition and device control.

The single-mode (SM) fiber (3M, FC-SN-4224) was switched to position 1 in Fig. 2 for luminescence experiments. It guides both the excitation laser light and the luminescence signal. To couple the pump light into the microscope fiber and to direct the PL signal to the spectrometer a 2×2 single-mode fiber coupler (Gould Fiber Optics, 2340541) is used. It acts as a beam splitter with a transmission efficiency of $T = 0.4$ for the PL signal. The output power of the pump laser diode (Roithner, RLT8305MG) emitting at 830 nm wavelength was monitored with a power meter and kept below 30 μW in order to guarantee single exciton regime (intensity within the Gaussian focused illumination spot < 15 W/cm^2). A set of two long pass filters (NDC Infrared Engineering, LWP-880-25) placed in front of the spectrometer (Acton Research Corporation SpectraPro-300i) suppressed the excitation light. The luminescence was dispersed with a 0.25-m grating spectrometer and detected with a nitrogen cooled CCD camera (Princeton Instruments, LN/CCD-1100PB), a setup with a resolution of 150 μeV.

For the absorption experiment the SM fiber was switched to position 2 in Fig. 2. The tuning range of the external cavity semiconductor diode laser (Sacher Lasertechnik, TEC500) is 930–980 nm and covers the relevant spectral range of the ensemble PL. Directing the beam to a wavemeter (Burleigh, Model 20VIS) with a flip-mount mirror allows for laser wavelength monitoring. A narrow and stable laser line ($\Delta\nu(20\text{ s}) < 5$ MHz) is essential in order to perform spectroscopy on the narrow X^0 line (lifetime limited linewidth $\Delta\nu \sim 160$ MHz). The output power is 25–40 mW depending on the wavelength and was attenuated with neutral density filters. To avoid mode hops induced by back reflections we placed a Faraday isolator (Linos, DLI1) into the optical path. The degree of polarization of the incident beam was defined by polarizing components.

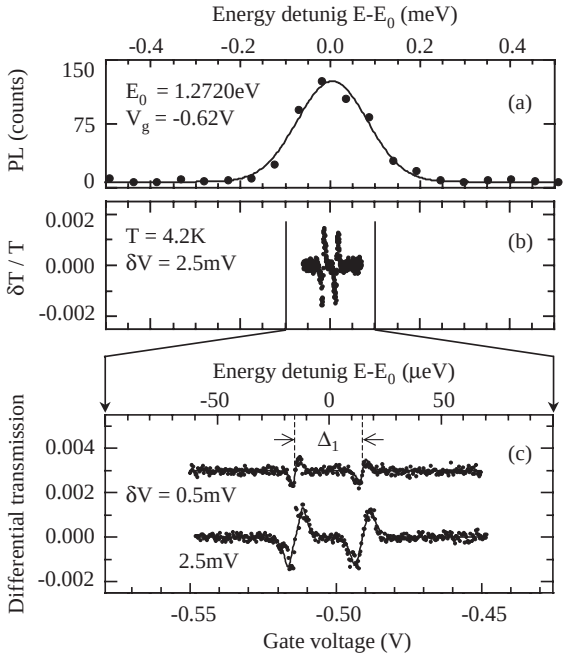

Fig. 3. (a) PL spectrum of a single quantum dot. At $V_g = -0.62$ V, the emission peak centered at $E_0 = 1.2720$ eV is that of a neutral exciton X^0 (dots: Counts in 120 s, line: Gaussian fit to the data). The spectral resolution is limited by the spectrometer to 150 μeV. (b) Differential transmission spectrum of the neutral exciton on the same quantum dot showing two resonances obtained with modulation amplitude $\delta V = 2.5$ mV. (c) Zoom-in of (b) by a factor of 10 in energy. A spectrum obtained with $\delta V = 0.5$ mV is also shown and offset by 0.003 for clarity. The fine structure splitting between the resonances is 30 μeV.

First, we performed luminescence spectroscopy on a spectrally isolated single QD. The PL was recorded as a function of gate voltage V_g. At high negative gate voltages, the radiative excitonic recombination is quenched due to field ionization [13]. The onset voltage of the X^0 PL emission is $V_g = -0.70$ V and the energy is centered at $E_{PL}(X^0) = 1.2719$ eV. The spectrum recorded at $V_g = -0.62$ V with an integration time of 120 s is shown in Fig. 3a. At $V_g = -0.50$ V the emission line jumps to lower energy indicating a transition from the neutral exciton state X^0 to the singly charged

state X^{1-}. We also observed higher charged excitons X^{2-} and X^{3-} at more positive gate voltages. From the gate voltage sweep we unambiguously identify the PL energy and the voltage interval of the neutral exciton X^0.

With applied gate voltage the X^0 energy exhibits a shift due to the Stark effect in the vertical electric field. The dependence of the PL energy is found to be a quadratic function of V_g, as anticipated for a polarizable dipole in an electric field [14]. Fig. 1b shows the quadratic fit function to the Stark-shifted PL energy corrected for the bias shift [15] in the absence of nonresonant pump light and limited to the voltage interval -0.70 to -0.45 V. The linear component is determined by the permanent dipole moment, the quadratic component by the polarizability of the confined exciton and both are in good agreement with values discussed in Ref. [14]. In the given voltage range of the X^0 emission line, a linear approximation of the energy dispersion with a slope of $+(1.3 \pm 0.1)$ μeV/mV can be made for the sake of simplicity without introducing significant errors.

In order to perform excitonic interband transition experiments on the ground state of the selected single QD, we set the laser wavelength to match the X^0 recombination energy. The fine tuning of the energy level into resonance was achieved by sweeping the gate voltage (Fig. 1). A superimposed square wave modulation signal with a frequency f and a small modulation amplitude δV allows for the measurement of the differential optical transmission [9]. The modulation frequency was set to 77 Hz. Due to the large active area ($\sim$20 mm^2) and therefore high ohmic resistance of the Ge diode at 4.2 K, its dynamic range was limited to about 100 Hz. Biasing the photodetector did not improve the performance. The laser power was set to 1.6 nW as measured on the Ge detector, corresponding to 0.145 W/cm^2 intensity within the focal spot. The gain mode of the SRS preamplifier was set to low-noise and the gain factor to 10^8 V/A. Using the lock-in settings of 100 μV for the sensitivity, 20 dB for the gain and 20 s for the time constant, a gate voltage sweep was performed. The DC-calibrator ramped up the voltage from -0.55 to -0.45 V in steps of 100 μV with a time interval adjusted to the lock-in integration time constant. A contrast ratio of maximum absorption strength to noise was typically 10.

A typical spectrum obtained with modulation amplitude of $2\delta V = 5$ mV peak to peak (p–p) is shown in Fig. 3b. The abscissa scale of the energy detuning is the same as in Fig. 3a for the recombination line detected with a standard PL setup. The representation demonstrates the advantage of the differential optical transmission technique in terms of spectral resolution. In fact, the spectrometer limited resolution of 150 μeV in the case of our PL setup is reduced down to 1.3 μeV rms by two orders of magnitude. The fine structure splitting of the ground state can be clearly resolved. A zoom-in representation is given in Fig. 3c, also showing a differential transmission spectrum recorded at $2\delta V = 1$ mV p–p modulation amplitude. Two absorption peaks separated by the fine structure splitting energy $\Delta_1 = (30.1 \pm 0.1)$ μeV can be resolved. We have investigated the polarization dependence of the resonances and found them to be linearly polarized in-plane with orthogonal orientations. The splitting energy and the polarization property are characteristic for confinement potentials with a weak in-plane asymmetry. Values for the fine structure splitting of 20–50 μeV observed for natural QDs have been reported earlier [16]. For self-assembled QDs grown under similar conditions, the splitting of the ground state could not be resolved but an upper boundary was given to 50 μeV [12]. Higher values (150–200 μeV) for the fine structure splitting were reported for strongly asymmetric self-assembled InGaAs QDs in Ref. [17].

The expected line shape is the derivative of a Lorentzian transmission curve. Details on the line shape and the fit function to the data points (solid line in Fig. 3c) have been discussed in Ref. [9]. We adopt the same notation and the fitting formalism for our analysis. The differential transmission signal is given by $\delta T = \alpha_0 \gamma^2 \{1/[(\omega+\delta\omega)^2+\gamma^2] - 1/[(\omega-\delta\omega)^2+\gamma^2]\}$. Here α_0 denotes the absorption value at resonance, ω is the Stark frequency detuning from the resonance, $\delta\omega$ is the adjustable energy modulation amplitude given by $\hbar\delta\omega = (1.3\ \mu\text{eV/mV})\delta V$ and γ is the dephasing rate of the excitonic transition. We obtained the full-width at half-maximum of the absorption resonance $\Gamma = 2\gamma$ for each of the two peaks as well as the strength $\alpha_0 = 0.0014 \pm 0.0001$. From the spectra we deduce for $\hbar\Gamma$, the linewidth in energy, values of (3.6 ± 0.2) and (5.4 ± 0.3) μeV for the two lines. To obtain the maximum absorption value at resonance,

we have to account for the fact that under the present excitation conditions the absorption amplitude still depends on the laser power. We find that the absorption reaches its maximum value 0.0037 for power densities in the focal spot below $\sim$0.15 W/cm^2. Moreover, the excitation is circularly polarized yet the transitions are linearly polarized, such that the value for the maximum absorption strength has to be doubled resulting in $\alpha_{max}\sim$0.007. In the low power limit, the linewidth is (2.3 ± 0.2) μeV, very close to values reported in Ref. [18]. Using the model of a two level system and again following the formalism described in Ref. [9], we derive for the oscillator strength of the transition $f\sim$13–15 in good agreement with values obtained from ensemble absorption measurements on InAs QDs [19].

In summary, we have established a new absorption technique for measuring the transition to the exciton ground state in single self-assembled QDs. Due to an improvement of the spectral resolution by more than two orders of magnitude, the fine structure splitting of the ground state energy level could be resolved. The homogeneous linewidth as well as the oscillator strength for the ground state transition were determined directly in the absorption spectrum.

We would like to thank the groups of J.P. Kotthaus and T.W. Hänsch at the LMU for the loan of equipment, likewise C. Kurtsiefer and J. Volz in the group of H. Weinfurter, LMU, for assistance in setting up fiber/web camera access to their wavemeter. We acknowledge the loan of low temperature positioners from Atto Cube Systems. Financial support for this work was provided in Germany by the DFG (SFB631) and in the UK by ESPRC. B. Alén was supported by the Marie Curie European training program.

References

[1] A. Imamoğlu, et al., Phys. Rev. Lett. 83 (1999) 4204;
E. Biolatti, R.C. Iotti, P. Zanardi, F. Rossi, Phys. Rev. Lett. 85 (2000) 5647;
P. Chen, C. Piermarocchi, L.J. Sham, Phys. Rev. Lett. 87 (2001) 067401.
[2] P. Michler, et al., Science 290 (2000) 2282;
Z. Yuan, et al., Science 295 (2002) 102.
[3] M. Pelton, et al., Phys. Rev. Lett. 89 (2002) 233602.
[4] D.J. Wineland, W.M. Itano, J.C. Berquist, Opt. Lett. 12 (1987) 389.
[5] W.E. Moerner, L. Karod, Phys. Rev. Lett. 62 (1989) 2535.
[6] J.R. Guest, et al., Phys. Rev. B 65 (2002) 241310.
[7] T.H. Stievater, et al., Appl. Phys. Lett. 80 (2002) 1876.
[8] T. Matsumoto, et al., Appl. Phys. Lett. 75 (1999) 3246.
[9] B. Alèn, F. Bickel, K. Karrai, R.J. Warburton, P.M. Petroff, Appl. Phys. Lett. 83 (2003) 2235.
[10] J.M. Garcia, et al., Appl. Phys. Lett. 71 (1997) 2014.
[11] R.J. Warburton, et al., Nature 405 (2000) 926;
M. Bayer, et al., Nature 405 (2000) 923.
[12] B. Urbaszek, et al., Phys. Rev. Lett. 90 (2003) 247403.
[13] D. Haft, et al., Appl. Phys. Lett. 78 (2001) 2946.
[14] R.J. Warburton, et al., Phys. Rev. B 65 (2002) 113303.
[15] J.M. Smith, et al., Appl. Phys. Lett. 82 (2003) 3761.
[16] D. Gammon, E.S. Snow, B.V. Shanabrook, D.S. Katzer, D. Park, Phys. Rev. Lett. 76 (1996) 3005.
[17] M. Bayer, et al., Phys. Rev. Lett. 8 (1999) 1748.
[18] M. Bayer, A. Forchel, Phys. Rev. B 65 (2002) 041308.
[19] R.J. Warburton, et al., Phys. Rev. Lett. 79 (1997) 5282.