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Absorption spectroscopy of single InAs self-assembled quantum dots

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Abstract

Excitonic transitions of single InAs self-assembled quantum dots were directly measured at 4.2 K in an optical transmission experiment. We use the Stark effect in order to tune the exciton energy of a single quantum dot into resonance with a narrow-band laser. With this method, sharp resonances in the transmission spectra are observed. The oscillator strengths as well as the homogeneous line widths of the single-dot optical transitions are obtained. A clear saturation in the absorption is observed at modest laser powers.

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Keywords: Quantum dots; Absorption spectroscopy; Relaxation dynamics

Since their discovery, self-assembled semiconductor quantum dots have received increasing interest, both as a unique system for studying the physics of quasi-zero dimensional nanostructures, and for their potential photonics applications. An abundant literature exists on their optical properties, including the photoluminescence from single dots. Nevertheless, it is still a challenge to measure directly the optical absorption of an individual quantum dot (QD). Transmission experiments on single ions [1] and single molecules [2] revealed the difficulties inherent to this type of measurement in which the desired optical signal is buried in the orders of magnitude larger probing light beam. Recently, the development of sub-wavelength resolution near-field optical microscopes has enabled a measurement of the absorption of a "natural" QD a single island in a GaAs/AlGaAs quantum well with a very good signal-to-noise ratio [3,4]. However, measurements obtained on single InAs QDs showed very weak signals [5]. This behavior is due to the fact that the oscillator strength of such InAs dots is one order-of-magnitude weaker [6] than that of natural dots leading to a weaker absorption. The information accessible through absorption, namely the oscillator strength and the homogeneous linewidth of electronic states, is extremely valuable but difficult to obtain directly by other means. Such quantities can be inferred from the wavelength dependence of the photoluminescence intensity, but to interpret the results, a model must be assumed to

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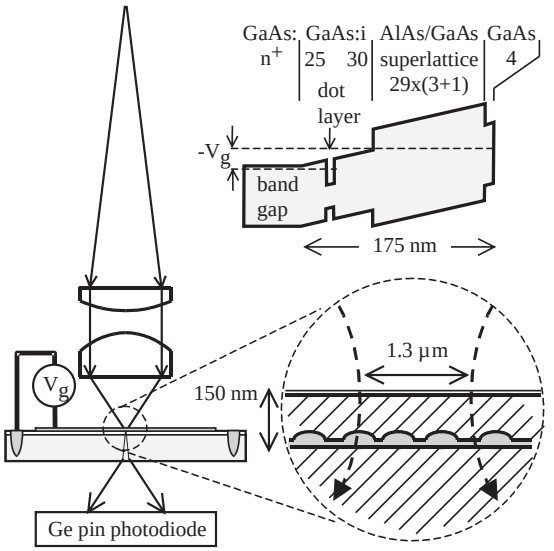

Fig. 1. Transmission experiment setup showing the microscope objective, the sample and the detector. The band structure is also shown (numbers in nm). The laser spot has a Gaussian profile with a full-width at half-maximum FWHM = 1.3 µm.

account for the intradot relaxation dynamics and ground state radiative recombination [7–9].

In this paper we show that excitonic absorption of a single InAs QD can be detected using Stark-shift spectroscopy in a transmission geometry. The signal-to-noise ratio is good. The experiment yields not only the oscillator strength but also information on the relaxation dynamics.

The dots investigated in this work were self-assembled by Stranski–Krastanow growth using molecular beam epitaxy. The dot layer is located inside a field-effect-structure, namely in a region between a 5 nm semitransparent NiCr gate film and a metallic highly n-doped GaAs layer, which we refer to as the back contact. The details of the growth can be found elsewhere [6,10]. The dot layer is separated from the back contact by a 25 nm thick undoped GaAs layer, which acts as a barrier for electron tunneling from the back contact into the dots, while the electrons are prevented from tunneling to the gate electrode by a 100 nm thick GaAs/AlAs superlattice blocking barrier. The schematic band structure in the dot layer is depicted in Fig. 1. The number of electrons in the dot is controlled electrostatically by applying a gate voltage V_g between the gate and the back contact ($-1\ \mathrm{V} < V_g < 1\ \mathrm{V}$). Coulomb blockade is very strong such that the number N of electrons in the dot is constant within a large window ΔV_g [6,11]. In our sample N ranges from 0 to 6 and ΔV_g is typically between 100 and 400 mV. A manifestation of dot charging is detected optically through the discrete energy shifts (typically a few meV) in the exciton emission energy whenever an electron is added to the dot [6,11]. Coulomb interaction as well as exchange interaction in charged excitons has been thoroughly investigated by measuring photoluminescence as a function of V_g [11]. An effect central to this work is that within the voltage window ΔV_g where the dot charge is constant, the excitonic energies can be shifted by up to about 200 µeV by the Stark effect. We use this effect to tune the exciton energy into resonance with the photon energy of a narrow band laser beam. Another important consideration is that complete filling of an electronic shell in the dot quenches the exciton absorption because of Pauli-blocking [6]. This effect can be used to turn the corresponding exciton absorption on and off [6].

Fig. 1 shows a schematic of the experiment. We used an all optical fiber confocal microscope operating at 4.2 K to focus the beam of a tunable external cavity diode laser onto the sample ($1050 < \lambda < 1070$ nm). The laser manufacturer specifies a linewidth of 300 kHz during a 5 s measurement time. The laser beam is focused onto the sample surface where it has a Gaussian profile with a 1.3 µm full-width at half-maximum spot size. An unbiased Ge photodiode placed immediately behind the sample detects the transmitted light. The photocurrent is measured with a low-noise commercial current amplifier operating at room temperature. In our experiment, the laser wavelength is constant and the exciton energy in the dot is tuned by sweeping V_g. This avoids the problems associated with a fluctuating intensity when sweeping the laser. Although these fluctuations can be as small as 1%, they are unfortunately much larger than the amplitude of the transmission resonances we are seeking. We modulate the Stark shift with a square wave at 77 Hz in order to tune the excitonic transition in and out of resonance with the laser line. The demodulated transmission signal represents the change in light power ΔP on the detector induced through the Stark effect. Integrating with a 5 s time

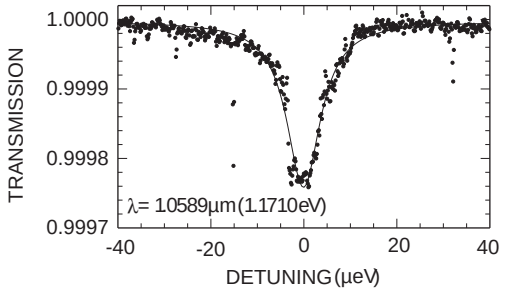

Fig. 2. A typical optical transmission resonance of a single quantum dot. The detuning energy is given with respect to the laser photon energy. To Stark-shift the resonance, V_g was swept from -0.595 to -0.555 V.

constant gives a low noise trace, limited by the current amplifier noise of 60 fA/Hz$^{1/2}$. The DC part of the signal is also measured and gives the total transmitted power P. Hence, the experiment provides $\Delta P/P$. In order to obtain the maximum possible ΔP, the amplitude of the square-wave modulation was set to give an excitonic energy shift of $\sim$100 μeV, larger than the resonance line width of $\sim$10 μeV. A typical transmission resonance is shown in Fig. 2.

The dots studied here are about 20 nm in diameter with a height of about 6 nm. Differential transmission measurements performed on the dot ensemble [6] showed that the ground state of the excitonic interband transition peaked at 1.09 eV, and that the first excited states have their maximum absorption at 1.17 eV. In our case, we set the probing photon energy at 1.17103 eV ($\lambda = 1.0589$ μm) in order to measure the resonance of the first excited states. We refer to this transition as p–p since in the strong confinement regime both the electron and the hole of the exciton occupy their respective p-state.

The areal dot density was estimated from ensemble capacitance measurements to be $(5 \pm 1) \times 10^9/\mathrm{cm}^2$ indicating that on average 90 dots are located in the probing spot area. Nevertheless, in this high-resolution spectroscopy experiment, only a few of these dots show a transmission resonance. This is due to the inhomogeneous broadening of 30 meV in the ensemble spectrum [6]. Since the typical tuning range through the Stark shift does not exceed 0.5 meV, the probability of measuring a dot absorption at any given laser wavelength located in the range of the ensemble absorption is $\sim(0.5\ \mathrm{meV}/30\ \mathrm{meV}) \times 90\ \mathrm{dots} = 1.5$ dots.

The amplitude of the transmission peak in the raw data of Fig. 2 is not the full amplitude of the transmission resonance. The tunable laser used here has a broadband background emission that corresponds to 63% of the measured power so that only 37% of the photons can contribute to the resonance. Furthermore, only half of these photons have the polarization corresponding to the transition selection rule. As a result, only about 18% of the total photons are active. Hence the true differential transmission of the dot is $\Delta T/T = \Delta P/(0.19 \times P) = 5.4 \times \Delta P/P$ in Fig. 2.

The transmission is modeled using a simple two level system with $\Delta T/T = -\alpha_0\gamma^2/(\omega^2 + \gamma^2)$. The signature of the optical resonance in transmission is a Lorentzian-shaped negative peak. Here, α_0 is the absorption at resonance, ω is the frequency detuning from the laser photon frequency, and $\gamma = \Gamma/2$ is the dephasing rate of the excitonic transition. The solid line depicted in Fig. 2 is the best fit to the experimental data obtained using the above expression with $\alpha_0 = 1.3 \times 10^{-3}$ and $\hbar\Gamma = 8.8$ μeV. The oscillator strength of a two-level optical transition is $f = 162\, n\phi^2\alpha_0\Gamma$ where Γ is in units of μeV, $n = 3.6$ is the optical index of the GaAs host material, and ϕ is the size of the focused Gaussian beam of the laser (the full-width at half-maximum, expressed in μm). We find $f \sim 12$ in good agreement with the ensemble experiment [6,12].

Once the exciton is photo-created in an excited state, it could relax its energy simply by re-emitting a photon resonant with the pumping field. With the measured oscillator strength we can estimate the radiative relaxation rate for spontaneous emission, $\Gamma_r = 2\pi ne^2 f/(3\varepsilon_0 m_0 \lambda^2 c)$. This leads to $\hbar\Gamma_r = 0.562$ μeV which is 20–30 times smaller than our measured line widths. A relevant point is that photoluminescence experiment at low pumping power shows hardly any emission from the p–p transitions. It is therefore unlikely that the lifetime of the excited state is radiation limited, and the origin of our resonance broadening must be of a different nature. Our narrowest measured line width is $\hbar\Gamma = 8.8$ μeV giving an upper limit of $\sim$75 ps for the non-radiative lifetime of the p–p excitonic resonance. Statistics performed on several tens of excitonic resonances obtained on nearby dots have shown homogeneous line widths ranging between 8.8 and 14 μeV, averaging around 12 μeV. This indicates that an exciton lifetime above 50 ps is

an intrinsic property of the QDs studied. The presence of long-lived excited states has been predicted as a consequence of the discrete nature of the density of states of QDs. In this model, relaxation is only possible when the energy difference between the states matches the available phonon energies [13]. However, this so-called phonon-bottleneck is not pronounced; in practice, there is strong emission from the QD ground state indicating that relaxation is efficient in these nanostructures. Relaxation lifetimes ranging between 2 and 90 ps have been reported for in InGaAs QDs depending on the size of the dot [14]. The conclusion of Ref. [14] is that relaxation times of just a few ps are associated with optical phonon emission. Relaxation times larger than ~20 ps are associated with acoustic phonon emission by the holes and this mechanism dominates the relaxation whenever the exciton has to relax by losing an energy less than the LO phonon energy. From transmission measurements performed on the dot ensemble, we know that the average separation between the ground and first excited transition is ~80 meV, well above the InAs LO phonon energy (~32 meV) [6]. Hence, one might expect that the homogeneous line width measured in our experiment is dominated by broadening due to optical phonon emission. However, this is not the case because in the range of applied gate voltage, the investigated dots are doubly charged [6]. After photon absorption, an electron–hole pair is created in the first excited state. If the s state is empty, the p state electron relaxes by emitting an LO phonon. However, in our case with a full s shell, electron relaxation is clearly forbidden because of the Pauli exclusion principle. Instead, the hole relaxes to its ground state (Fig. 3). This process occurs via low-energy acoustic phonon emission, a process that is much slower than LO phonon emission [14]. The line width of the transmission resonance is therefore determined by hole relaxation in this experiment. In this way, we determine a dot-dependent hole relaxation time of 50–75 ps.

Insight into the relaxation mechanism can be obtained by measuring the dependence of the absorption on the probing light power. We observe a dramatic reduction of the absorption peak for laser powers exceeding 0.5 W/cm^2. This quenching can be explained in terms of saturation. When a new exciton is created, the charge configuration of the single QD changes. The resulting Coulomb and exchange interactions shift

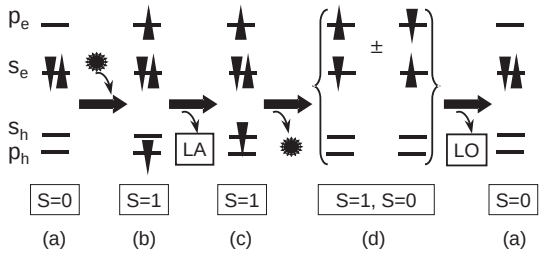

Fig. 3. Dot configurations for the cascade process showing a complete cycle between two absorption events for a doubly charged quantum dot. S indicates the total spin of the carriers in the dot.

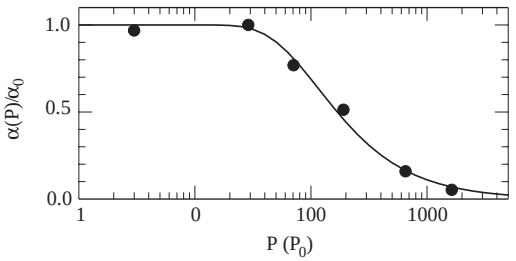

Fig. 4. Absorption at the excitonic resonance divided by the maximum absorption in the limit of low laser power plotted against laser power. The laser power is scaled relative to P_0 which we estimate to be 1 ± 0.5 nW.

the exciton absorption energy by an amount many times larger than the homogeneous line width [11]. The excitation laser wavelength is no longer in resonance with the interband optical transition and a new photon cannot be absorbed until the original configuration has been restored. This takes a certain time τ. During time τ, the medium is transparent to the incoming light. As the laser power increases, many photons strike the sample when the dot is transparent such that the time averaged absorption cross-section is effectively reduced. The probability of the system decaying into its initial state after an absorption event at $t = 0$ behaves with time t as $e^{-t/\tau}$. The absorption cross-section $\alpha(P)$ at an arbitrary excitation power is then given by $\alpha(P) = \alpha_0(1 - e^{-\hbar\omega/\alpha_0\tau P})$ where $\alpha_0 = \alpha(0)$ is the absorption in the limit of very low illumination power. In Fig. 4, we represent the evolution of the maximum absorption contrast as a function of the excitation power for a single excitonic resonance. The measured power is expressed in units of P_0 where $P_0 \sim 1 \pm 0.5$ nW within our measurement uncertainty. The solid line represents the best fit to the exponential

law indicated above. Taking $\alpha_0 \sim 5.4 \times 10^{-4}$, the fit yields an average cycle time of $\tau \sim 3$ ns. This is about 40–60 times larger than the dephasing time obtained from the homogeneous broadening of the transmission resonance. This result strengthens our hypothesis of non-radiative broadening for the optical p–p transition. It might be argued that the tunneling of electrons from the first excited state through the barrier plays a role in the relaxation dynamics. This is not the case however since the photoluminescence from the doubly charged exciton in its ground state is very intense, showing that the electron in the p state is long lived [11]. Our results suggest that a cascade of relaxations takes place, as depicted in Fig. 3. The dot has initially two electrons in its ground state (a). It then absorbs a photon which generates an electron and a hole in their respective excited states (b). Hole relaxation then takes place by acoustic phonon emission depicted by the transition (b) to (c). This process determines the exciton dephasing time of 50–75 ps. The subsequent step in the cascade, (c) to (d), shows a radiative recombination process which occurs with a typical radiative lifetime of ~800 ps as determined from time-resolved photoluminescence measurements on these samples. The system is then left with two electrons, one in the s shell and one in the p shell, which form either a singlet or a triplet state, in complete analogy with the first excited state of a He atom. Relaxation of this state (d), returns the system to its initial state (a). Only when the dot returns to state (a) can the dot absorb another photon. The saturation experiment implies that the cascade process takes about 3 ns which is larger than the radiative lifetime. There is therefore a process in the cascade that is slower than the radiative rate. We propose that the last process, (d) to (a), represents the bottleneck in the cascade process. The singlet state can relax its energy through emission of an LO phonon as no spin flip is required. We have measured this relaxation time to be 2 ps from the line width of the emission from a doubly-charged exciton [11]. This process is therefore not the limiting one. However, the triplet state must transfer its total spin $S = 1$ to the lattice in order to relax and this state is therefore long lived [11]. In the cascade of Fig. 3, both the singlet and triplet state are equally probable. Considering several cycles, the time the dot spends in a non-resonant state is essentially determined by the slowest process, namely the relaxation of the triplet state. Our measurements suggest that this process has a typical relaxation time of about 3 ± 1 ns.

In conclusion, we have demonstrated a method of measuring absorption on a single dot with high spectral resolution. It clearly opens the way for studying dephasing processes and Coulomb interactions of neutral as well as charged excitons in a single dot. We also show that the absorption can be saturated at laser power densities exceeding 1 W/cm^2. By studying the saturation of the absorption, we argue that we can determine the spin-flip relaxation rate of electrons in the excited state.

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