Stark-shift modulation absorption spectroscopy of single quantum dots

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Excitonic interband optical transitions within single InAs self-assembled quantum dots have been directly observed in a transmission experiment at 4.2 K. Using Stark shift, the excitonic energy levels of a single quantum dot are tuned into resonance with a narrow-band laser line. The Stark shift is also modulated at low frequencies. Relative changes in transmission can be detected this way down to one part per million. The oscillator strength as well the homogeneous linewidth of the transition is obtained. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1609243]

Optical transmission and reflection spectroscopy count among the most versatile and routine methods for characterizing gases, liquids, and solids. However, when it comes to investigate volumes of material as small as to contain only a single quantum absorber (or scatterer), most experiments rely on fluorescence (i.e., luminescence) spectroscopy techniques.[1] However, direct absorption measurements would be desirable since they allow one to quantitatively access the optical polarizability. Such experiments were conducted on single ions[2] and single molecules[3] but revealed a poor signal-to-noise ratio. The difficulty lies in the fact that the desired optical signal is detected in transmission along with the orders of magnitudes larger probing light beam whose fluctuations dominates the noise level. Recently, the absorption of single natural quantum islands in GaAs/ AlGaAs quantum wells was measured in transmission near-field microscopy experiments where excitonic interband transition lines were obtained with a very good signal-to-noise ratio.[4,5] This was made possible not only because these types of dots have a very large optical oscillator strengths but also that the use of a near-field optical probe allowed most photons to interact efficiently with a single quantum dot. Similar transmission measurements were reported on single self-assembled InAs quantum dots but showed very poor signals.[6] This is because, first, the oscillator strength f of InAs dots is one order of magnitude weaker[7] so that the total amount of scattered light is reduced in proportion to f. Second, the radiative decay rate is reduced also in proportion to f, imposing the use of weaker light intensities in order to not saturate the absorption. The signal-to-noise level reduces consequently as $1/f^2$ when one operates just below saturation; a condition necessary here in order to be able to measure enough signal. We present, in this letter, an optical method allowing direct transmission measurements on single InAs self-assembled quantum dots.

The InAs dots investigated in this work were grown self-assembled using the Stranski–Krastanov growth by molecular-beam epitaxy as described elsewhere.[8] The dots are separated from a highly doped GaAs layer, the back contact, by 25 nm of intrinsic GaAs which acts as a barrier for electron tunneling from the back contact into the dots. The electrons are prevented from tunneling to the gate electrode, the metalized sample surface, by a 100 nm thick GaAs/AlAs superlattice blocking barrier. The whole structure forms a field-effect device so that the dots can be both controllably charged with electrons[7] and have their energies tuned by the Stark effect.[9] The electron occupation of the dots is controlled by applying a voltage, V_g, between the gate and back contact and monitored with the capacitance of the device.[7] Here, we measured single dot absorption when the majority dot occupation was between 1 and 2 electrons on average. However, the occupation of each measured single dot could not be independently identified. From the ensemble capacitance, we estimated the dot density to be (5 ± 1) $\times10^9$ cm^{-2}. The InAs dots are about 20 nm in diameter with a height of about 7 nm. Our ensemble optical transmission measurements showed that the allowed dot excitonic transitions peaked at 1.09 eV for the ground-state levels and at 1.17 eV for the first-excited states. The spectral width of the absorption band was 30 meV due to inhomogeneous broadening inherent to ensemble measurements. The fact that excitonic transition can be Stark shifted at fixed electron occupation[9] is central to this work.

Figure 1(a) shows the experimental setup. The beam of a narrow band (1 MHz) tunable external-cavity semiconductor diode laser is focused onto the sample surface using an aspheric objective lens with a numerical aperture of 0.55. The size of the Gaussian focused spot width was 1.3 μm [full width at half maximum (FWHM)] as measured at 4.2 K with a laser wavelength of 1.06 μm. The transmitted light was detected using an unbiased Ge *p-i-n* diode placed directly behind the sample and a commercial current–voltage preamplifier. The objective lens, the sample, and the detector were located at 4.2 K in a He bath cryostat. This miniature transmission microscope was constructed to be mechanically stable so that a single quantum dot could be investigated for weeks at a time without losing it from focus. Ideally, the laser wavelength should have been tuned to 1.14 μm in order

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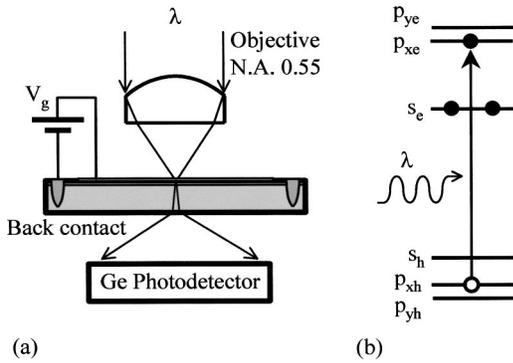[b]Electronic mail: karrai@lmu.de

FIG. 1. (a) Transmission measurement schematics (to scale). The Ge detector is placed directly behind the sample at 4.2 K. (b) The optical absorption creates an exciton with its electron and hole in a *p* level of the InAs quantum dot. Here, we illustrate two electrons filling the *s* shell (i.e., $V_g = -0.5$ V).
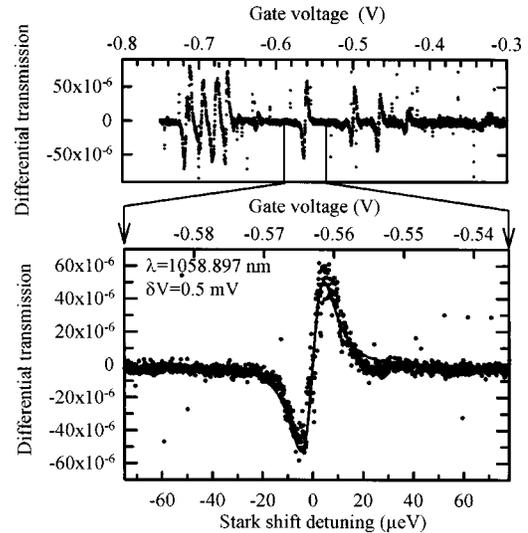

FIG. 2. Top: Differential transmission obtained by Stark-shift modulation of the excitonic dot absorption. The multiple lines originates from different excitonic transitions in several dots. Bottom: Modulated excitonic absorption line from a single optical transition in a single quantum dot.

to match the energy of the ground-state exciton transition. Our laser could not operate in such a wavelength regime. Instead, we tuned it to $\lambda \approx 1.06 \ \mu$m near the ensemble absorption peak energy at 1.17 eV of the first-excited excitonic state. The optical transition corresponds to the one that generates an exciton with both the electron and the hole occupying the *p* states, the first excited levels of the dot as shown in Fig. 1(b). A transmission spectrum is obtained using the Stark effect by sweeping the dot exciton transition energies across that of the probing photons which was kept fixed. The gate voltage is slowly ramped between -0.8 V and 0 V to in order to Stark shift the exciton energies. At the same time, a small square-wave modulation δV at a frequency of 77 Hz is superimposed onto the applied gate voltage. The ac part of the transmission photosignal is demodulated with a lock in. This way, a differential transmission signal is obtained only when a dot exciton energy is in resonance with the laser line. The dc part of the photosignal, typically 30 nW, was also measured during the experiment. The differential transmission is given quantitatively by the ratio of the demodulated photosignal to the dc signal and is limited by our current amplifier $[67 \text{ fA/(Hz)}^{1/2}]$ corresponding to a power equivalent noise of $\sim 150 \text{ fw/(Hz)}^{1/2}$. Our setup has a signal-to-noise ratio of $5 \times 10^{-6}/(\text{Hz})^{1/2}$. A reduction in this noise figure would be expected when operating at much higher frequencies. The data acquisition of a full spectrum is obtained in a few hours with an integration time constant of 5 s per data point.

A typical spectrum obtained this way is shown in Fig. 2. Figure 2 shows that a typical differential transmission spectrum was obtained by slowly sweeping the gate voltage between -0.8 V and 0 V. The ac modulation amplitude was $2\delta V = 1$ mV peak to peak. Several narrow lines are seen. Similar spectra are obtained on various regions of the sample. In all of the cases, no absorption line was found at $V_g > -0.3$ V. We determined from capacitance measurements on the dot ensemble that at this voltage, the *p* shell fills with electrons from the back gate.[9] Here, the modified Coulomb interaction lowers the exciton energy by 10 meV, shifting the absorption peak away from the laser line. Furthermore when the *p* shell is completely filled, a Pauli blocking of the transition will prevent absorption. The position and intensities of the resonances depend on the illuminated sample location. In the bottom part of Fig. 2(a), a single

resonance spectrum was measured with a high-resolution gate scan. The obtained line shape is the derivative of the expected transmission signal. This is the case when the amplitude of the Stark modulation is smaller than the homogeneous linewidth of the transition. In order to obtain a correspondence between the applied gate voltage and the Stark energy shift, the measurement was repeated at several fixed laser photon energies as shown in Fig. 3. We obtained a linear Stark-shift dispersion with a slope of $+(2.78 \pm 0.06) \ \mu$eV/mV. This linear dispersion was measured also on several other single resonances and was found to be very similar. The dots also have a quadratic dispersion term[9] that could not be resolved within the small range of energy shift measured here. We performed the differential transmission measurements with increasing amplitudes δV of the square-wave modulation. The result is shown in Fig. 4. The spectra evolve from a derivativelike absorption line into two well defined single resonances. In order to model the transmission
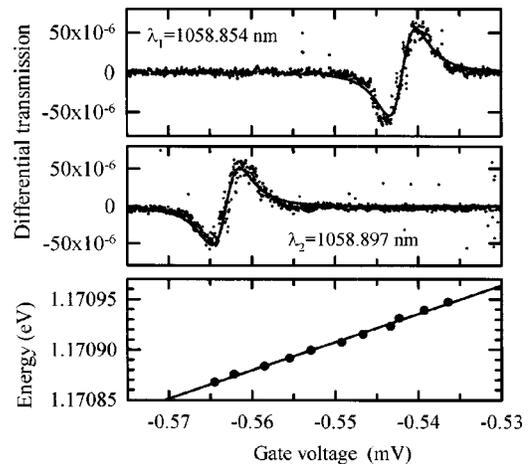

FIG. 3. Two top panels: Two absorption line Stark shifted measured at two different wave lengths on a same single quantum dot. Bottom panel: The photon energy plotted against the resonance position gives a precise calibration of the Stark-shift energy dependency. ac (77 Hz) modulation $2\delta V = 1$ mV.
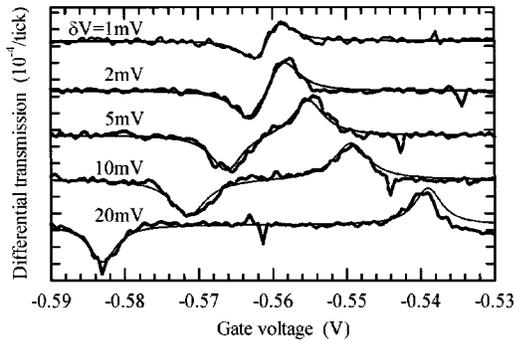
FIG. 4. Dependency of the Stark-shift modulated absorption on the amplitude of the gate voltage modulation (indicated). As the amplitude of the square-wave gate modulation exceeds the absorption linewidth, the signal evolves into two separate absorption lines. The line fit is obtained using $\hbar\Gamma = 14.5\ \mu$eV and $\alpha_0 = 2.2 \times 10^{-4}$ and increasing modulation δV (i.e., $\delta\varepsilon$).

signal, we assumed a homogeneously broadened absorption Lorenzian line shape. In this model, the nonmodulated transmission is given by $T = 1 - \alpha_0 \gamma^2/(\omega^2 + \gamma^2)$, where α_0 is the absorption at resonance, ω is the Stark frequency detuning from the resonance, and γ is the dephasing rate of the excitonic transition. The FWHM of the absorption resonance is $\Gamma = 2\gamma$. In our experiment, we modulate the energy of the transition in a square-wave form with an adjustable peak-to-peak amplitude $2\hbar\delta\omega$. Within a modulation period, the transmission switches between $T(\omega - \delta\omega)$ and $T(\omega + \delta\omega)$. The ac transmission signal is then given by the difference $\delta T = \alpha_0 \gamma^2 \{1/[(\omega - \delta\omega)^2 + \gamma^2] - 1/[(\omega + \delta\omega)^2 + \gamma^2]\}$ as shown in Fig. 4. In the limit of small modulation amplitudes, such $\delta\omega \ll \gamma$, the differential transmission simplifies to $\delta T \approx 4\alpha_0 \gamma^2 \omega \delta\omega/(\omega^2 + \gamma^2)^2$. In this limit, the FWHM defined herein is $\Gamma = \sqrt{3}\Delta\omega$, where $\hbar\Delta\omega$ is the energy separation between the position of the minimum and the maximum of the resonance. The maximum absorption is given by $\alpha_0 = \Delta T \Delta\omega/(3\delta\omega)$, where ΔT is the peak-to-peak value of the differential transmission signal. Here, applying an energy modulation $\hbar\delta\omega = 1.39\ \mu$eV (i.e., $\delta V = 0.5$ mV), we measure $\hbar\Delta\omega = 8.3\ \mu$eV and $\Delta T = 1.0 \times 10^{-4}$. We deduce then $\hbar\Gamma = 14.5\ \mu$eV and $\alpha_0 = 2.0 \times 10^{-4}$. At large modulation amplitudes $\delta\omega \gg \gamma$, the signal develops into two well resolved Lorenzian shaped lines separated in energy by $2\hbar\delta\omega$. In this mode of operation, a more direct measurement of the optical resonance is obtained. In particular, both Γ and α_0 can be extracted directly from the FWHM and the amplitudes of the peaks. The position of the resonances are shifted by $+\delta\omega$ and $-\delta\omega$. The strength of the resonance $\alpha_0 = 2.0 \times 10^{-4}$ obtained experimentally is not the dot absorption yet. The measured spectral emission of our laser shows a broadband background superimposed on a narrow line, an effect inherent to laser diodes. The nonresonant background accounts for ~62% of the total intensity measured in the dc photosignal. Furthermore, a selection rule on the photon state of polarization must be obeyed. Since we could not observe significant changes in the strength of the signal when rotating

the laser linear polarization by 90°, we deduce that only half of the photons are contributing to the transition. The absorption of active photons should be then $\alpha_{\text{Max}} \sim 4 \alpha_0 = 8.0 \times 10^{-4}$. In order to relate the oscillator strength f of the transition to α_{Max} and Γ, we use the absorption of a two-level system given by $\alpha(\omega) = [\hbar e^2 f/(2\gamma A m_0 \varepsilon_0 c n_{\text{eff}})]\gamma^2/(\omega^2 + \gamma^2)$, where $\alpha(0) = \alpha_{\text{Max}}$, A is the area of the illuminated spot[10] and $n_{\text{eff}} = n = 3.6$ is the optical index of the GaAs matrix in which the dots are imbedded.[11] We deduce that the oscillator strength of the measured optical transition is $f \sim 12 \pm 3$ in good agreement with what we previously measured in an ensemble measurement[7,11] on samples from the same wafer. The expected rate of spontaneous emission is related to f by $\Gamma_{\text{sp}} = 2\pi n e^2 f/(3\varepsilon_0 m_0 \lambda^2 c)$, namely, $\hbar\Gamma_{\text{sp}} = 0.563\ \mu$eV or, equivalently, a relaxation time of $1/\Gamma_{\text{sp}} = 1.17 \times 10^{-9}$ s. In contrast, $1/\Gamma$ measured here is 26 times shorter. Our interpretation is that the exciton is created in its first-excited state and decays in its ground state faster than $1/\Gamma_{\text{sp}}$ through acoustic phonon emission. We anticipate that the resonant absorption of the long-lived ground-state exciton should be dominated by spontaneous photon emission. In this limit, the peak absorption strength is given by $\alpha_{0\text{sp}} = (3/2\pi)(\lambda/n)^2/A = 0.021$.

In conclusion, we demonstrated that resonant transmission high-resolution spectroscopy can be performed on self-assembled single InAs quantum dots. We introduced a measurement for studying single dot dephasing processes and Coulomb interaction-induced fine structure of excitons.

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[10] $A = 1.13\phi^2$ for a Gaussian spot, where ϕ is the spot width at half intensity.

[11] The effective optical index n_{eff} takes into account the proximity of the sample surface so, in fact $(n+1)/2 < n_{\text{eff}} < n$. In R. J. Warburton, C. S. Dürr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Phys. Rev. Lett. **79**, 5282 (1997) we used $n_{\text{eff}} = (n+1)/2$ where we assumed the dots located at the sample surface.