Rabi splitting and ac-Stark shift of a charged exciton

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The Rabi splitting of the negatively charged exciton in a single InGaAs quantum dot is observed in resonance transmission spectroscopy. We use a pump laser excitation to drive strongly the unpolarized trion transition in a quantum dot and detect its modified absorption spectrum with a second weak probe laser. By tuning the pump laser near resonance, we observe an ac-Stark effect dispersion, with a power dependent Rabi splitting on resonance, both signatures of a strongly coupled two level system. Although the pump and probe laser fields are resonant with the same transition, we do not observe all features in the Mollow spectrum. We combine the results of pump probe with saturation spectroscopy data to deduce the individual contributions to the low power linewidth. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2837193]

One of the most attractive features of self-assembled quantum dots (QDs), embedded in a field effect device, is that they can be charged with single electrons or holes.[1] Spectroscopy on a single QD allows a detailed study of the optical properties of the emission (photoluminescence) (Refs. 2 and 3) and absorption[4–8] of neutral and charged QDs. The singly charged QD is particularly important since its resident electron can serve as a spin qubit.[7,9] A major prerequisite for coherent spin control in a QD is the few level nature of the electronic states. The observation of Rabi oscillations of the neutral exciton transition has strengthened the "artifical atom" picture of a single InGaAs QD.[10] Very recently, optical pump and probe experiments on a neutral QD led to the observation of the Rabi splitting in the frequency domain[11,12,12] and to a Mollow absorption spectrum[11] as observed on atoms.[13,14] However, spin control is best achieved on a charged exciton.[7] Here, we present a resonant pump and probe experiment on the negatively charged exciton revealing optical Stark shift dispersion as well as Rabi splitting at optical resonance.

The sample contains self-assembled InGaAs QDs whose emission wavelength has been shifted to around 950 nm by *in situ* annealing during growth. The dot layer is embedded in a field effect structure[1] consisting of a n-doped back contact, 25 nm below the QDs, and a metallic semitransparent NiCr top gate, 276 nm above the back contact. To prevent leakage currents in this structure, a blocking barrier was grown between the dot layer and the sample surface. It consists of an AlAs/GaAs superlattice beginning 10 nm above the QD wetting layer. By applying a voltage between gate and back contact, we can charge the dots with single electrons by tunneling from the back contact. Further, the resonance energies of the excitonic transitions are modified by the quantum confined Stark effect.[2,5] We perform resonant laser spectroscopy in transmission, where gate voltage modulation is used to apply lock-in detection.[6] The sample is mounted in a fiber-based, diffraction-limited confocal microscope that is immersed in He exchange gas in a liquid helium bath cryostat at 4.2 K. We use a continuous wave (cw) pump laser and a separate cw probe laser. The lasers are linearly polarized perpendicularly to each other. The trion (X^{1-}) transition is unpolarized at zero magnetic field,[4] such that the choice of polarization does not affect the outcome of the experiment. Behind the sample, a lens collimates the transmitted light. In order to separate the pump and probe transmission signals, a polarizing beam splitter divides the light into the two orthogonal polarizations and each polarization is detected by a photodetector. The setup is shown in Fig. 1(a). Due to losses in the collection path after collimation, we collect less power on the detectors P_{det} than experienced by the QD: $P_{\text{dot}}=pP_{\text{det}}$. This factor we measure to be $p=11$, the values for the power given below are the powers at the location of the quantum dot. The polarization sensitive detection setup allows us to increase the pump intensity up to 40 times the probe laser intensity without significantly affecting the probe detector. For all experiments, the probe laser power is kept at an intensity that lies below optical saturation of the transition:[4,15] $P_{\text{probe}}=9$ nW. We keep the pump laser photon energy constant in the middle of the X^{1-} charging plateau[5] with a power 20 times the power of the probe laser. The probe laser photon energy is scanned through the QD trion resonance for different gate voltages.

In Fig. 1(b), transmission spectra are shown for different gate voltages and hence for different exciton detunings from the pump laser photon energy [$\Delta=(E_X-E_{\text{pump}})\propto V_G$, with E_X the energy of the unperturbed X^{1-} transition]. The transmission, measured in the probe channel, is plotted as a function of the detuning of the probe laser with respect to the pump laser at different gate voltages. The plots clearly show that the single Lorentzian resonance detected by the probe laser at large negative Δ develops a splitting as $|\Delta|$ decreases and

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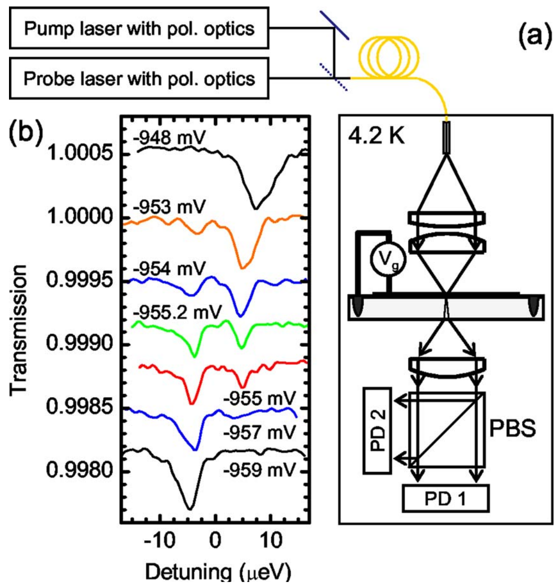

FIG. 1. (Color online) (a) Schematics of the polarization sensitive detection scheme. Two tunable narrow band lasers provide the pump and the probe beam whose polarizations can be controlled independently. The two beams are coupled into a single mode fiber that guides them into the cryogenic confocal microscope operating at 4.2 K. After the sample, the two transmitted beams are separated by a polarizing beam splitter (PBS) and detected on the two photodetectors PD1 and PD2. The graphs in (b) show the ac-Stark modified spectra measured with the probe beam for different detunings of the pump laser. This was achieved by keeping the pump laser photon energy constant and scanning the probe laser photon energy at different gate voltages. The pump and probe laser powers were $P_{\text{pump}}=202$ nW and $P_{\text{probe}}=9$ nW. The spectra have been shifted by an offset relative to 1 for clarity.

shows a doublet structure for $\Delta=0$. For positive Δ, the oscillator strength is completely transferred to the second resonance and the first one vanishes. In Fig. 2(b), the resonance energies of the observed transitions are plotted versus the applied gate voltage, while in (a) the X^{1-} resonance for zero pump power is shown. The behavior of the observed resonances follows a clear anticrossing with a strong pump laser.

In principle, the X^{1-} exciton represents a four-level system, with two electron spin ground states and two exciton spin states. However, without a magnetic field, the hyperfine interaction with the nuclei leads to rapid spin relaxation between the two electron spin states, comparable to the exciton spontaneous recombination rate.[16] In fields above about 0.3 T, relaxation via the hyperfine interaction is suppressed

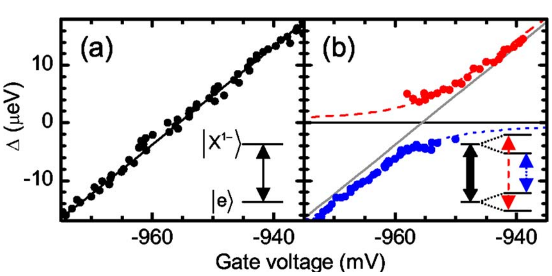

FIG. 2. (Color online) (a) Energy shift of the transmission resonance of the X^{1-} transition of a single QD as a function of the applied gate voltage for zero pump power. The inset shows the excited transition in the QD. (b) The measured resonance energies obtained by sweeping the probe photon energy while the strong pump laser photon energy is kept constant (horizontal line) are plotted as a function of the gate voltage. The gray line corresponds to the transition shown in (a). The dotted and dashed lines are a simple anticrossing behavior as expected from 1. The inset indicates the pump (black arrows) and probe (dotted and dashed arrow) transitions.

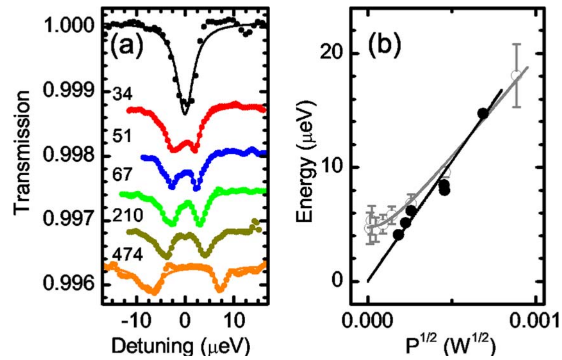

FIG. 3. (Color online) (a) The probe laser transmission spectra with pump laser on resonance, with increasing power (values in the figure in nW), as a function of the detuning between the probe and the pump laser photon energy. The spectra have been shifted vertically for clarity and are fitted by two Lorentzian resonances. (b) Splitting of the resonance measured with the probe laser as a function of the square root of the pump laser power (black) and the measured linewidth of the pump laser absorption spectrum (gray open circles). The splitting follows a linear behavior (black line), while the power broadening can be described by formula (2) (gray line).

but in our device, spin relaxation is enabled by a spin-swap process with the back contact, a cotunneling phenomenon.[7] This means that there is no pronounced optical pumping of the spin and no chance of any long-lived ground state coherences in this particular case. We therefore attempt to describe these experiments with a two-level model, including a dephasing term γ_{nr} to represent the spin relaxation. As we show, this approach yields convincing agreement with the experimental data. The anticrossing in Fig. 2(b) can be described for a strongly coupled two-level system by[17]

$$E_{\pm} = \frac{1}{2}(\Delta \pm \sqrt{\Delta^2 + 4|W|^2}). \tag{1}$$

Here, $E_{\pm}$ gives the resonance energies of the two resonances of the strongly driven two level system, depicted by the dotted and dashed lines in Fig. 2(b). W quantifies the coupling strength, the Rabi energy $W=\hbar\Omega$ which is power dependent: $\Omega^2=2\alpha_0\gamma_0 P_{\text{pump}}/\hbar\omega_{\text{pump}}$.[18] The parameters α_0 and γ_0 describe the coupling of the two-level transition to the light field. α_0 is the low power limit of the transmission contrast, and $\gamma_0=\gamma_{\text{nr}}+\gamma_{\text{sp}}$ is the dephasing rate for low power, given by the sum of nonradiative rate γ_{nr} and the spontaneous recombination rate $\gamma_{\text{sp}}=1/\tau_{\text{sp}}$, where τ_{sp} is the spontaneous lifetime.[15,18] In the present experiment, the probe and the pump field are resonant with the same transition. This is very similar to a recently reported experiment carried out on the neutral exciton.[11] However, while for the neutral exciton, a Mollow absorption spectrum with gain was observed, as expected for a strongly driven two level system,[13,14] in the present experiment, there was no sign of gain in the spectrum. In order to lift the spin degeneracies, we repeated the experiment in a magnetic field of 1 T and found the same results as in zero magnetic field, namely, absence of gain and the central peak. This point is not fully understood but we note that gain and the central peak are sensitive to pure dephasing processes which are larger for the charged exciton than for the neutral exciton because of the hyperfine interaction, the interaction of the electron spin with the nuclear spins.

In Fig. 3(a), probe laser spectra are shown for different pump laser intensities. The resonance resembles a Lorentzian for zero pump power, but develops a splitting that increases with the square root of the pump laser intensity. In Fig. 3(b), the measured splitting is plotted as a function of the square root of the pump power (black dots) and fitted by a linear dependency (black line). This experiment measures the Rabi energy which depends on γ_0. To determine γ_{sp} and γ_{nr} separately, we glean additional information from saturation spectroscopy with a single laser where it is known that the measured linewidth increases with increasing laser power.[15,18,19] The linewidth follows the power-broadening relation:

$$\Gamma = 2\hbar\gamma_0\left(1 + \frac{P}{\hbar\omega\gamma_{sp}}\alpha_0\right)^{1/2}, \qquad (2)$$

with the spontaneous recombination rate γ_{sp}. In this formula, two limits can be distinguished. First, for vanishing laser power P, the linewidth is power independent: $\Gamma = 2\hbar\gamma_0 + c$. The constant c represents the broadening of the spectra due to spectral fluctuations in the vicinity of the QD.[4,7,11] Second, for large power, the linewidth increases with the laser power according to $\Gamma \simeq 2\hbar\gamma_0\sqrt{P\alpha_0/\hbar\omega\gamma_{sp}}$. Using the definitions given before for the Rabi frequency, this simplifies to $\Gamma \simeq \sqrt{2\gamma_0/\gamma_{sp}}\hbar\Omega$. In Fig. 3(b), the linewidth measured on the same trion transition in a one-laser experiment is plotted as a function of the square root of the laser power (gray open circles) and fitted by formula (2) (gray line). As expected, the linewidth converges for low powers toward a constant value $\Gamma = 2\hbar\gamma_0 + c$. Here, $\hbar\gamma_0 = 1.7$ μeV and $c = 1.3$ μeV. For large powers, the linewidths exhibit a slope that differs from that of the observed splitting in the pump-probe experiment which follows $2\hbar\Omega$ and we fit $\hbar\gamma_{sp} = 1.25$ μeV. This corresponds to a lifetime $\tau_{sp} = 0.52$ ns. This value agrees very well with that obtained by time resolved photoluminescence[20] on similar structures. We get furthermore the contribution of nonradiative decay to the linewidth $\hbar\gamma_{nr} = 0.45$ μeV. The mechanism responsible for this dephasing process is likely the hyperfine interaction. We note that in the present evaluation, the power loss in the detection arms does not introduce a systematic error since in both experiments, the single laser and pump-probe experiments were performed in the same setup.

In summary, we present a direct way to measure the Rabi splitting of a trion transition in a single QD and its dispersion when subject to a strong driving field. The data agree very well with a simple anticrossing model based on a two-level atom. By combining results from single laser and pump-probe experiments, we are able to infer the contributions of recombination, pure dephasing, and spectral fluctuations to the low power linewidth.

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