Voltage-Controlled Optics of a Quantum Dot

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(Received 17 June 2004; published 18 November 2004)

We show how the optical properties of a single semiconductor quantum dot can be controlled with a small dc voltage applied to a gate electrode. We find that the transmission spectrum of the neutral exciton exhibits two narrow lines with ~ 2 μeV linewidth. The splitting into two linearly polarized components arises through an exchange interaction within the exciton. The exchange interaction can be turned off by choosing a gate voltage where the dot is occupied with an additional electron. Saturation spectroscopy demonstrates that the neutral exciton behaves as a two-level system. Our experiments show that the remaining problem for manipulating excitonic quantum states in this system is spectral fluctuation on a μeV energy scale.

DOI: 10.1103/PhysRevLett.93.217401 PACS numbers: 78.67.Hc, 71.70.Gm, 73.21.La, 78.55.Cr

In order to realize semiconductor-based schemes for the generation of single photons [1], the generation of entangled photons [2], and photonics-based quantum information processing [3], it is necessary to generate and manipulate optically active quantum states. Although atoms, ions, and molecules are possible hosts for these processes, quantum dots have the advantage that the solid state matrix acts as a built-in trap. In addition, semiconductor heterostructure technology allows considerable control over the continuum states at energies above the quantum dot energy levels. We demonstrate here how this capability can be exploited to control the polarization, the oscillator strength, and the energy of the optical transitions in a single quantum dot simply with a dc voltage. In particular, we are able to turn an exchange energy on and off. This level of tunability has never been achieved previously on any quantum object, atom, molecule, or quantum dot. Furthermore, our experiments provide compelling evidence that the ground state exciton behaves as a highly coherent two-level system.

Significant progress in quantum dot optics has been made with natural quantum dots, localizing fluctuations in a disordered quantum well [4], leading to the recent demonstration of an optical CROT logic gate [5]. For our own experiments, we employ a self-assembled quantum dot which has a much stronger confining potential than a natural dot, leading to a significantly longer excitonic coherence time of several hundred ps [6,7]. The oscillator strength of a self-assembled quantum dot is about 10, smaller than that of a natural quantum dot [8], making the controlled creation and detection of excitonic quantum states very challenging. We report here the first detection of ground state exciton creation in a single self-assembled quantum dot through a reduction in the transmission coefficient. This is a very direct experimental technique as it does not involve a third state as is inevitably the case in a luminescence-based method. Furthermore, it profits from all the advantages of laser spectroscopy, namely, high spectral resolution, precise line shape determination, and direct access to the oscillator strength.

The InGaAs quantum dots are embedded in a GaAs matrix and emit around 1.3 eV. The dots are positioned 25 nm above a metalliclike layer (n^+-GaAs) and 150 nm beneath a Schottky barrier on the surface, allowing us to modulate the properties with the gate voltage, the voltage applied to the Schottky contact. The gate voltage induces a vertical electric field which shifts the exciton energy through the Stark effect [9]. By changing the potential of the quantum dot relative to the back contact, we can control the occupation. There is a pronounced Coulomb blockade both in the capacitance [10] and in the photoluminescence [11], allowing us to control unambiguously the electron number and excitonic charge, respectively. Optical absorption was measured with a narrow band (5 MHz) laser, detecting the transmission with a Ge *p-i-n* photodiode placed directly beneath the sample at 4.2 K, as shown in Fig. 1(a). The Gaussian beam from the laser was focussed to a 1.2 μm full-width-at-half-maximum (FWHM) spot. The dot density in our sample is $\sim 5 \times 10^9$ cm^{-2}, implying that several tens of dots lie in the focus. However, owing to the narrow laser line, quantum dots can be addressed individually. With the given spot size, a combination of the large sample refractive index and the homogeneous broadening implies that the change in transmission at resonance is small, and, therefore, as in single molecule [12,13] and single ion [14] transmission experiments, a differential technique was adopted. In addition to a dc voltage, a square wave voltage with peak-to-peak amplitude 100 mV was applied to the gate. Spectroscopy was performed by sweeping the dc gate voltage at constant laser energy measuring the dif-

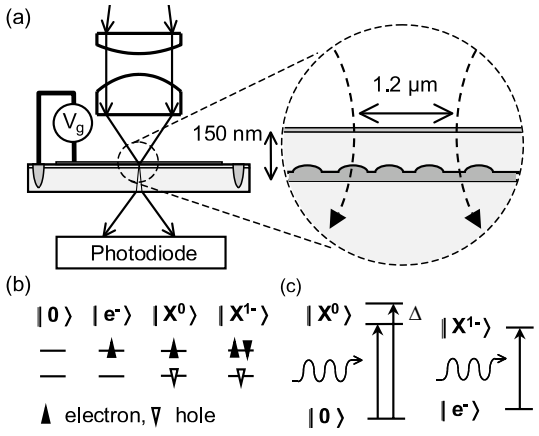

FIG. 1. (a) The optical microscope (to scale). Laser light is delivered with an optical fiber (not shown); the light is collimated and then focussed with an aspherical lens with numerical aperture 0.55 onto the sample. The FWHM spot size was measured to be 1.2 μm. Transmitted light is detected with an *in situ* Ge *p-i-n* photodiode. (b) Quantum mechanical states in a single quantum dot: $|0\rangle$ is the vacuum state, $|e^-\rangle$ the single electron state, $|X^0\rangle$ the neutral exciton state, and $|X^{1-}\rangle$ the singly-charged exciton state. (c) The level diagrams for the creation of a neutral exciton and a singly-charged exciton. The neutral exciton is split by Δ through the anisotropic electron-hole exchange interaction.

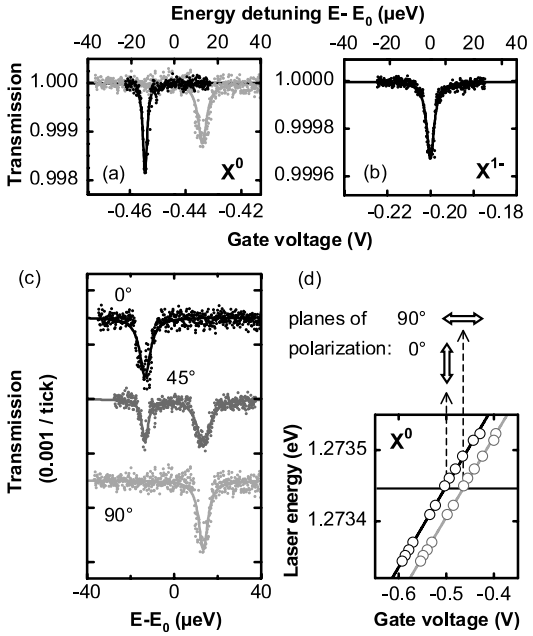

FIG. 2. (a) Differential transmission of the neutral exciton in a single self-assembled quantum dot. The two curves were recorded with orthogonal linear polarization. (b) Differential transmission for the singly-charged exciton X^{1-}. (c) Differential transmission for X^0 for three directions of the linear polarization. (d) Voltage-induced Stark-shift of the two resonance energies of the neutral exciton. At a fixed photon energy, the polarization can be switched between the two orthogonal linear orientations through the gate voltage. The detuning in (a), (b) and (c) was achieved at constant laser wavelength by sweeping the gate voltage. The sample was at 4.2 K. Note: Experimental data in Figs. 2(a)–2(c) were recorded on one quantum dot; those in Fig. 2(d), and in Figs. 3 and 4 were recorded on another quantum dot.

ferential transmission with a lock-in amplifier. Initially, the power was kept below 10 nW to avoid saturation effects. Figure 1(b) shows a schematic representation of the quantum mechanical states involved in the creation of the neutral exciton X^0 and the singly-charged exciton X^{1-}, and Fig. 1(c) denotes the levels involved in the optical transitions.

Figure 2(a) shows differential transmission spectra of a single quantum dot for voltages at which the photoluminescence exhibits only recombination of the neutral exciton. The gate voltage is converted into a detuning, $E - E_0$, by repeating the measurements at several known laser wavelengths; in a narrow range of voltage, the exciton energy varies linearly with gate voltage [Fig. 2(d)]. The neutral exciton exhibits two pronounced Lorentzian-shaped resonances, separated by $\Delta = 27$ μeV, as shown in Fig. 2(a) and 2(c). We find values of fine structure splitting Δ ranging from 10 μeV to 42 μeV for the ten different quantum dots measured in this sample. As shown in Fig. 2(c), the two neutral exciton lines are linearly polarized in orthogonal directions. The fine structure splitting Δ arises through the electron-hole exchange interaction [15], its magnitude indicating that a shape anisotropy makes the dominant contribution[16]. The main point is that the quantum dot exciton has a natural linear polarization basis. Our results show that we can exploit this feature to prepare very precisely the excitonic state of the quantum dot.

By controllably addressing the *x*- and *y*-polarized excitons, the dot can emit only one particular polarization. This is a very attractive concept for a single photon source as it allows control over the polarization of the emitted photon. However, without the electric field, the two polarizations have different energies such that there is an unwanted correlation between the polarization and the photon energy. With our field-effect device this correlation can be removed. Figure 2(d) demonstrates how the *x*- and *y*-polarized excitons can have exactly the same energy by applying two slightly different gate voltages, an ideal feature for a single photon source.

It would clearly be desirable to change also the basis of the quantum dot exciton to generate not just linearly polarized photons but also circularly polarized photons. Our structure allows us to do exactly this. By making the gate voltage slightly more positive, the electronic Coulomb blockade allows us to populate the quantum dot with a single electron in the absence of an optical excitation. In this case, as shown in Fig. 2(b), the doublet in the transmission spectrum is replaced by a single line. The reason for this is that the exciton generated by photon absorption now has a filled *s* shell [Fig. 1(b)] and, therefore, total electron spin zero such that the exchange

interaction with the spin-$\frac{3}{2}$ hole vanishes. While this argument has been invoked to explain several experiments on the fine structure of excitons [15,17,18], our own experiment provides the first proof that the neutral exciton's fine structure disappears on charging. It is now possible to control the polarization selection rules of the exciton by controlling the quantum state of the resident electron. Equivalently, a measurement of the spin state of the resident electron can be made with photon absorption. We demonstrate this concept by applying a magnetic field which splits the spin-up and spin-down electron states through the Zeeman energy. The exciton splits into two components with right- and left-handed circular polarizations, exactly as expected in the absence of an exchange interaction, as shown in Fig. 3(a). Again, by applying slightly different gate voltages, the right- and left-circular polarized excitons at a fixed magnetic field can have exactly the same energy. Alternatively, the two states can be tuned into resonance at constant voltage and laser energy with a magnetic field, as demonstrated in Fig. 3(b). Our experiment demonstrates, therefore, that charging with a single electron has a profound effect on the excitonic states, causing the features related to exchange to disappear. Furthermore, the absorption can be turned off completely by applying a voltage such that the dot contains two electrons [19]. In this case, the ground state is occupied with two electrons and further occupation is forbidden through the Pauli principle.

For the manipulation of quantum states, it is desirable to work with a highly coherent two-level system. Up until now, the two-level nature of a self-assembled quantum dot exciton has been probed by detecting Rabi oscillations either on the excited exciton, relaxation to the ground state exciton providing a convenient detection scheme [20], or on the ground state exciton in a vertical electric field, relying on electron and hole tunneling to generate a photocurrent [21]. In both cases, the detection process compromises the coherence, a problem we can avoid with saturation spectroscopy. The experiment involves measuring the maximum contrast in the transmission resonance as a function of laser power. Saturation corresponds to a reduction in the contrast. The results, plotted in Fig. 4(a), show how a laser power of just 100 nW is sufficient to saturate the neutral exciton transition. We consider two different interpretations, one involving a coherent two-level system, the other a shelving state. In the first scenario, a photon excites an exciton which remains in resonance with the laser such that a subsequent photon can stimulate recombination. In this case, saturation arises when stimulated emission dominates over spontaneous emission because in this limit the detector signal is unchanged in the presence of resonant absorption. In the second scenario, an exciton relaxes quickly after its creation into an intermediate shelving state where it is no longer resonant with the laser. In this case, saturation arises because the exciton has a finite lifetime. At high power the system spends a large fraction of the time in the shelving state where there is zero chance of photon absorption. The crucial differences between the two models are that saturation occurs at roughly a factor of 3 higher power in the shelving model than in the two-level model and that the functional dependencies of the absorption on power are completely different. Applying the density-matrix formalism to a two-level system, we find the elegant result for the maximum absorption $\alpha(P)$ as a function of laser power P: $\alpha(P)/\alpha(0) = 1/[1 + 2\alpha(0)\tilde{N}\tau/\hbar]$, where $\tilde{N}$ is the photon flux through the quantum dot plane, and τ is the radiative lifetime. We have determined $\tau = 800 \pm 100$ ps both from the lower power limit of the transmission spectroscopy and from direct measurements of the radiative decay allowing us to predict the behavior at high power. As shown in Fig. 4(a), this two-level analysis gives an excellent fit to the experimental data. Conversely, in the shelving model, $\alpha(P)/\alpha(0) = 1 - \exp\{-\hbar/[\alpha(0)\tilde{N}\tau]\}$. With $\tau = 800$ ps, the shelving model gives a very poor account of the saturation characteristics [Fig. 4(a)]. A fit can be obtained in the shelving model only by increasing the radiative lifetime to 2.4 ns, an unreasonably long lifetime, and even in this case, an exponential decay gives a much poorer fit to the data than the two-level model. Remarkably, therefore, the self-assembled quantum dot displays the saturation characteristics of a two-level system.

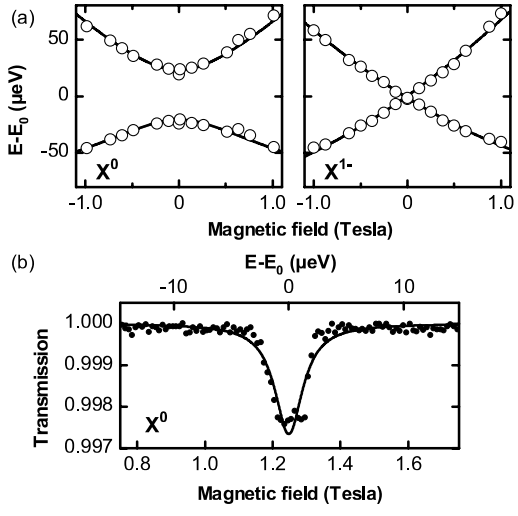

FIG. 3. (a) Magnetic dispersion of the neutral exciton X^0 (left) and the singly-charged exciton X^{1-} (right). The solid lines are fits to the data with detuning $\Delta E = \pm\frac{1}{2}g^*\mu_B B + \beta B^2$ for the X^{1-}, and $\Delta E = \pm\frac{1}{2}\sqrt{\Delta^2 + (g^*\mu_B B)^2} + \beta B^2$ for the X^0, taking the exciton g-factor $g^* = 1.8$, the anisotropic electron-hole energy $\Delta = 42$ μeV, and the diamagnetic shift $\beta = 8.7$ μeV/T^2. (b) Differential transmission versus magnetic field. The resonance corresponds to one branch of the spin-split neutral exciton.

The FWHMs of the neutral excitons are 2.3 μeV and 5.0 μeV in Fig. 2. Other dots give similar results, although it is not always the case that one resonance is broader than the other. The smallest linewidth we have

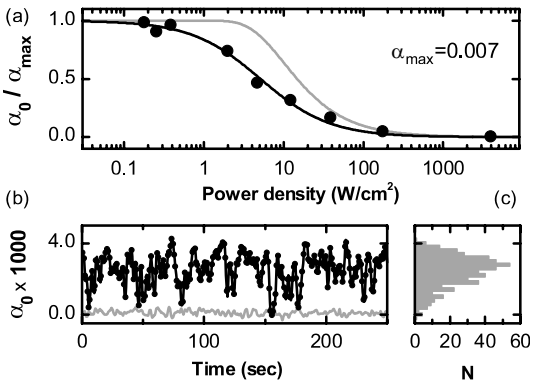

FIG. 4. (a) The differential absorption at resonance plotted against laser power. The solid black line is the result expected for a two-level system taking a radiative lifetime of 800 ps. The solid gray line models the power dependence of an absorbing system including an intermediate shelving state, again taking a radiative lifetime of 800 ps. (b) The differential absorption signal as a function of time at zero detuning (black) and several linewidths away from the exciton resonance (gray). (c) Histogram of the differential absorption amplitude signal at zero detuning.

recorded so far is 1.6 μeV. In the ultimate limit of lifetime broadening, the coherence time is equal to the radiative lifetime, corresponding to a FWHM of 0.8 μeV for our dots. Hence, although our resonances are very sharp, we have not yet achieved the lifetime broadening limit. In order to probe the broadening mechanism we measured the absorption as a function of time for constant laser energy, starting with the quantum dot in exact resonance with the laser. The results are plotted in Fig. 4(b) which shows also the transmission signal measured when the laser energy is several linewidths away from the exciton resonance. On resonance, the absorption signal has quite large fluctuations, whereas, off resonance there is just small random noise. The time constant for this particular experiment was 1 s. The experiment demonstrates that there are fluctuations in the absorption spectrum of the quantum dot on this time scale. The results are entirely consistent with a temporal fluctuation of the resonance position. The absorption signal at constant laser energy fluctuates as the dot moves in and out of resonance. The statistical nature of the fluctuations is shown in the histogram of absorption amplitude, Fig. 4(c). Assuming that on a much shorter time scale, the resonance is lifetime broadened with a width of 0.8 μeV, the data in Fig. 4(c) allow us to deduce that spectral fluctuation contributes approximately 0.5 μeV to the resonance linewidth. Integrating for longer times, as was the case in the data of Figs. 2 and 3 [time constant 50 s in Figs. 2(a) and 2(c) and 2 s in Figs. 2(b) and 3(b)], the resonance is broadened further, between about 1 and 3 μeV, by the spectral fluctuation. On the time scale of the experiment, the back contact is unlikely to be the origin of the spectral fluctuation given its high conductivity. Instead, preliminary results suggest that the spectral fluctuation is less in a magnetic field, implying perhaps that an interaction between the electronic spin and the nuclear spins in the quantum dot is important.

In summary, we have demonstrated that a self-assembled quantum dot can be used to prepare high-fidelity excitonic states with an unprecedented degree of tunability. The tuning is achieved simply through a voltage. This property was demonstrated by applying high resolution laser spectroscopy to a single quantum dot. Our results demonstrate that the remaining factor limiting the exploitation of these results for the controlled manipulation of quantum states is spectral fluctuation on a μeV energy scale.

We would like to thank Jörg P. Kotthaus, Harald Weinfurter and Ian Galbraith for helpful discussions. Financial support for this work was provided in Germany by DFG Grant No. SFB 631 and in the UK by the EPSRC and The Royal Society.

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