HTML ABSTRACT + LINKS

Spin-selective optical absorption of singly charged excitons in a quantum dot

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(Received 19 October 2004; accepted 25 April 2005; published online 24 May 2005)

We report high resolution laser absorption spectroscopy of a single InGaAs/GaAs self-assembled quantum dot embedded in a field-effect structure. We show experimentally that the interband optical absorption to the lower Zeeman branch of the singly charged exciton is strongly inhibited due to spin (Pauli) blockade of the optical transition. At high magnetic fields the optical absorption to the upper Zeeman branch dominates the absorption spectrum. We find, however, that the spin blockade is not complete and a 10% leakage remains at high magnetic fields. Applying a gate voltage to empty the dot of its resident electron turns the spin blockade off. This effect is observed at 1.5 K and up to 9 T. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1940733]

The coherence time of an excess electron spin strongly confined in a quantum dot (QD) structure is expected to be orders of magnitude longer than the typical time scales required for its coherent manipulation.[1–4] Motivated by this observation, several groups have proposed to use single QD spins as quantum bits (qubits),[5] and to manipulate, couple, and measure individual spins using either transport[4] or optical techniques.[6] In the case of self-assembled InGaAs QDs with strong confinement along the growth direction, the lowest energy optical transitions are those arising from the excitation of a $J_z=+3/2(J_z=-3/2)$ valence electron to a $S_z=+1/2(S_z=-1/2)$ conduction state. If the QD already has an excess conduction electron, only one of these optical transitions is allowed; the other is spin (Pauli) blocked.[7] In contrast, a neutral QD with asymmetric confinement potential always has a pair of exciton transitions. It has been suggested that Pauli blocking of absorption or fluorescence can be used to implement high efficiency all-optical single-spin measurements[6,8] and conditional spin dynamics.[8]

In this letter, we report resonant absorption measurements on a single QD charged with a single excess electron in the regime of Pauli blocking. We observe that for high magnetic fields ($B>5$ T) where the electron is (with high probability) in the lowest energy $S_z=+1/2$ state, the absorption of a left-hand-circularly polarized (σ^-) laser is suppressed by a factor of 10 as compared to that of a right-hand-circularly polarized (σ^+) laser. In contrast, we observe that both σ^+ and σ^- transmission dips have equal strength for a neutral QD, irrespective of the applied magnetic field. Using the fact that the strength of σ^+ and σ^- absorption in a charged QD is a measure of the spin polarization of the resident electron, we were able to determine the electron g factor by fitting the magnetic field dependence of the transmission data. Our results represent a step towards all-optical coherent spin measurement.

The InGaAs dots investigated in this work were self-assembled in the Stranski-Krastanow growth mode by molecular beam epitaxy. The QDs are separated from a highly n doped GaAs back contact by 25 nm of intrinsic GaAs which acts as a tunnel barrier. The electrons are prevented from tunneling to the gate electrode, the metalized sample surface, by a 110-nm-thick GaAs/AlAs superlattice blocking barrier. The whole structure forms a field-effect device. The electron occupation of the dots is controlled by applying a gate voltage and monitored with the QD photoluminescence charging diagrams.[9] An individual QD is identified to be spectrally separated by typically more than 5 meV from neighboring dots assuring that the observed features originate unambiguously from a single QD. We used high resolution laser spectroscopy and a low temperature confocal microscope to measure the differential transmission through the QDs. This technique[10–12] gives optical spectra as shown in Fig. 1. The spectral resolution is much less than the QD absorption width. A dip in the transmission is obtained when the resonantly scattered laser light is diverted away from the photodetector placed immediately behind the sample.

In the absence of a magnetic field, a negatively charged exciton (X^{1-}) strongly confined in a QD shows a single unpolarized absorption peak in the optical interband spectrum, as demonstrated in Fig. 1 with high resolution transmission spectroscopy. In the case of X^{1-}, electrons are forced to fill the lowest energy s shell such that the total electron spin is 0 in accordance with the Pauli principle. As a consequence, the exchange interaction between the confined electrons and the hole is suppressed so that the optical spectrum is free of fine structure features irrespective of the asymmetry of the con-

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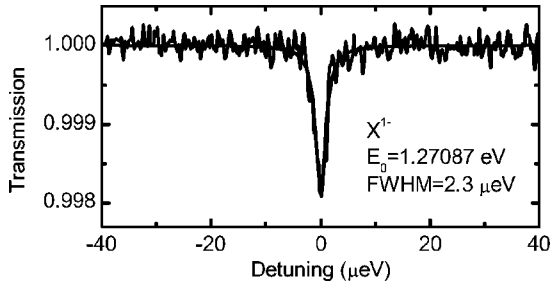

FIG. 1. High resolution laser spectroscopy of the transmission spectrum to a singly negatively charged exciton X^{1-} confined in a single self-assembled InGaAs quantum dot. The spectrum is taken at 4 K and $B=0$ T. The data are fitted to a Lorentzian peaked at $E_0=1.27087$ eV and with 2.3 μeV full width at half maximum.

fining potential.[12,13] This is not the case for the neutral exciton X^0: the electron-hole exchange leads generally to a splitting into two linearly polarized dipole-allowed optical transitions.[12,14,15] For the dot investigated here the measured neutral exciton splitting was 10 μeV.

In a high magnetic field and at low enough temperatures, the ground state of a singly charged QD is that of a spin-polarized electron in the lowest Zeeman level. As shown in Fig. 2(a), an interband photon can only be absorbed under the condition that the photo-generated electron is of opposite spin. In principle, at $T=0$ K this translates into a single absorption line in the optical spectrum irrespective of the applied magnetic field. In spite of the applied magnetic field, the exciton absorption should not show a Zeeman splitting, i.e., the optical transition at the lower exciton Zeeman energy should be spin blocked due to the Pauli principle independently of the photon polarization.

We have investigated Pauli blocking experimentally. Figure 2(b) shows transmission spectra in the presence of a

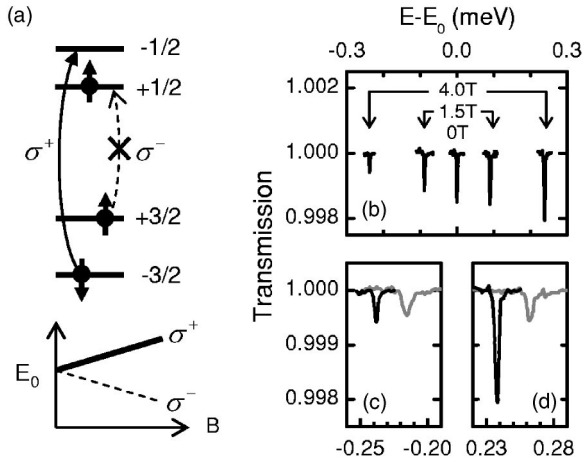

FIG. 2. (a) Schematics of the quantum dot Zeeman-split highest valence levels $J_z(-3/2,+3/2)$ and the lowest conduction levels $S_z(+1/2,-1/2)$ labelled with their respective angular momentum quantum numbers. The Zeeman energies are given by $\mu_B B(g_e S_z - 1/3 g_h J_z)$. Here the Landé factors are $g_e<0$ and $g_h<0$. The dipole-allowed transitions are shown with their circular polarizations σ^+ and σ^-. The low energy optical transition is Pauli blocked by the presence of the resident electron in the lowest conduction level. The schematic evolution of the absorption energy in magnetic field B is also shown. (b) Differential transmission spectra of the charged exciton in a single quantum dot at 1.5 K and magnetic fields of 0, 1.5, and 4.0 T. (c), (d) Low and high energy resonances of the charged exciton transition at 4.0 T for 1.5 K (black) and 12 K (gray). The small energy shift between the spectra is due to the slight dependence of the band gap on temperature. For all spectra the laser polarization was linear.

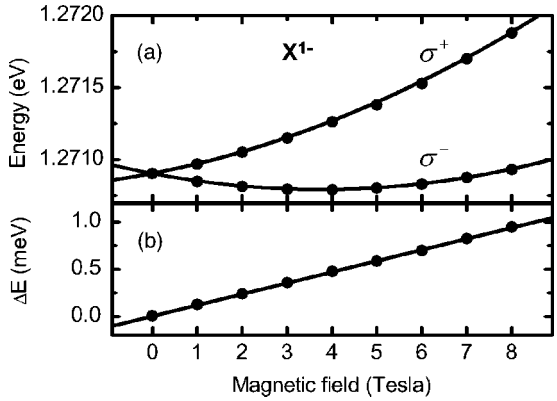

FIG. 3. (a) Magnetic field dispersion of the charged exciton at 4.2 K. Both circularly polarized branches are shown. The solid lines are fits to the data with $E=E_0\pm 1/2 g^* \mu_B B+\beta B^2$ taking the exciton g-factor $g^*=(g_e+g_h)=-2.0$ and the diamagnetic shift $\beta=7.9$ μeV/T^2. (b) Zeeman splitting of the two branches. The solid line is a linear fit to the data with a slope of 118 μeV/T.

magnetic field. At $T=1.5$ K and under magnetic field two well-resolved transmission resonances are observed. Both lines correspond to the two optical transitions shown in Fig. 2(a). The energy positions of the resonances are plotted in Fig. 3(a) showing clearly a behavior quadratic in field, consistent with the exciton diamagnetic shift.[16] The energy splitting, plotted in Fig. 3(b), is proportional to the applied magnetic field consistent with a Zeeman splitting of the charged exciton. We have conducted the transmission measurements as a function of the light polarization. At finite magnetic fields the two dipole-allowed transitions have right and left circular polarizations. Figures 2(b)–2(d) show the corresponding spectra using linearly polarized light. At low temperatures, the strength of the low energy resonance decreases with increasing magnetic field while the opposite is true for the higher energy resonance. Figure 4(a) shows the relative weight of the resonances as a function of magnetic field. For each magnetic field the polarization was optimized to maximize independently each of the two resonances. At high magnetic field it can clearly be seen that the absorption of the higher energy branch dominates. The low energy branch is dramatically inhibited, saturating at high field at a level of one tenth of the stronger peak, without ever completely disappearing [Fig. 4(b)]. The high magnetic field limit confirms

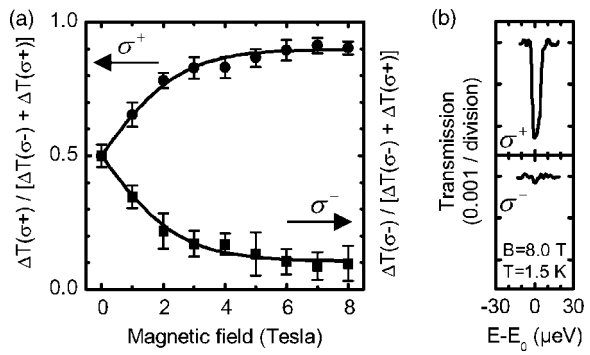

FIG. 4. (a) Evolution of the relative resonance intensities with magnetic field for both circular polarizations. The solid lines are fits to the data with electron g-factor $|g_e|=1.8$ as described in the text. The temperature was 1.5 K. (b) Differential transmission spectra of the right-hand circular (upper panel) and left-hand circular (lower panel) transition at a magnetic field of 8.0 T.

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roughly the expected Pauli blocking behavior in that one resonance clearly prevails.

A further confirmation of the Pauli blockade picture is obtained by increasing the temperature to $T=12$ K. In this case, the spectra obtained at $B=4$ T show two Zeeman-split absorptions with comparable peak strengths [Figs. 2(c) and 2(d)]. The interpretation is that the resident electron is thermally activated and occupies both spin states with about equal probability. Lowering the temperature to 1.5 K clearly favors the higher Zeeman branch confirming that the spin polarization of the resident electron is the source of the Pauli blocking. A final consistency check is obtained by setting the gate voltage such that the dot is freed of its resident electron. The neutral exciton transition is then favored and under the same magnetic field and temperature conditions we observe that the lower Zeeman branch regains its initial strength, exactly as expected.

At $T=1.5$ K the degree of spin polarization depends on the magnetic field. We have used a two-level Boltzmann statistical distribution for the spin state occupation. Considering that the relative strength of both resonance peaks is a measure of the spin polarization of the resident electron, we fitted the evolution of the data in magnetic field, as shown in Fig. 4(a). As a fit parameter we used a Landé factor for the resident electron of $|g_e|=1.8\pm0.2$. This value is about twice as large as the one reported for InGaAs/GaAs self-assembled QDs in Ref. 15. However, several other dots emitting around 1.3 eV close to the center of the inhomogeneous QD energy distribution exhibit an electron g factor of 0.6 as measured at 4.2 K. The deviation of the g factor from dot to dot is not surprising since the value strongly depends on the individual In content and strain distribution. To obtain an optimal fit, we needed to introduce empirically a limit to the maximum achievable electron spin polarization of about 90% in order to account for the fact that the lower energy absorption branch does not vanish at high fields, as shown in Fig. 4(b). This result indicates that the Pauli blockade of the optical transition is not complete as ideally predicted. It was also observed for the dots with the electron g factor of 0.6 consistent with the temperature and the Zeeman energy. At the present time, the precise origin for the apparent limitation of the Pauli blocking is still unknown and requires more detailed measurements and analysis.

In conclusion, using high-resolution transmission spectroscopy we have confirmed that a singly negatively charged exciton confined in a single self-assembled QD has a dominant absorption line at the higher Zeeman energy branch as a result of the spin polarization of the resident electron. Unexpectedly, at high fields the absorption at the lower Zeeman branch, although drastically inhibited as expected from Pauli blocking, is still not completely extinguished.

The authors would like to thank A. O. Govorov for helpful discussions. Financial support for this work was provided in Germany by the DFG Grant No. SFB 631, in Switzerland by NCCR Quantum Photonics Grant No. 6993.1, and in the UK by the EPSRC.

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