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RESONANT INTERACTION BETWEEN A QUANTUM DOT AND A NARROWBAND LASER: SPECTROSCOPY AND OPTICAL PUMPING OF A SINGLE SPIN

M. KRONER*, S. SEIDL*,†, B. D. GERARDOT†, B. Biedermann*, A. Badolato‡, P. M. Petroff‡, K. Karrai* and R. J. Warburton†

*Center for NanoScience and Department für Physik, Ludwig-Maximilians-Universität, 80539 München, Germany

†School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, UK

‡Materials Department, University of California, Santa Barbara, California 93106, USA

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Recently, a technique has been developed for performing laser spectroscopy on single self-assembled quantum dots. A summary is presented here of spectroscopy results concerning the charge- and magnetic-field dependent energies and selection rules of the fundamental exciton. In addition, it is also shown how the resonant interaction of a laser with a singly-charged dot can be exploited to pump the electron into one of the spin states. This involves applying a small magnetic field and an appropriate bias in order to suppress spin relaxation through the hyperfine interaction and through co-tunneling processes, respectively.

1. Introduction

The laser as a highly coherent light source has opened up countless possibilities in probing and manipulating the quantum states of atoms. There is ample motivation to apply these techniques to semiconductor quantum dots which have atom-like discrete energy levels and at low temperature, a highly coherent optical response. Historically however, laser spectroscopy of atoms has been performed on ensembles of identical atoms in order to boost the signal, for instance the resonance fluorescence or absorption coefficient. This is impractical for quantum dots because each quantum dot is unique, possessing its own particular morphology and optical resonance. Furthermore, quantum information processing is motivating many experiments on individual quantum states. For both these reasons, a pressing need is the development of laser spectroscopy techniques for single quantum dots. This is a challenging enterprise as the quantum dot size, a few nm, is much smaller than the optical wavelength, typically a micron. We describe here laser spectroscopy on a single self-assembled quantum dot. The initial experiments are purely spectroscopic in nature, establishing the energies, polarization-dependent selection rules,[1] and behavior in a magnetic field.[2] Recent experiments in another group have gone one

stage further by using the resonant interaction with the laser to manipulate a single electron in quantum dot.[3]. A single electron is trapped in a quantum dot[4] and a magnetic field is applied to lift the spin-up, spin-down degeneracy and to inhibit the electron spin relaxation mediated by the nuclear spins.[5,6] Under these conditions, the electron is pumped into the spin state which doesn't interact with the laser.[3] This represents an optical pumping experiment, equivalently a laser cooling of spin or a spin initialization step of a spin qubit. Results of an experiment similar to that in Ref. 3 are presented here.

2. Experimental Techniques for Single Dot Laser Spectroscopy

The crucial concept for our laser spectroscopy scheme is to detect a resonant interaction between the quantum dot and a narrowband laser through a reduction in the transmission coefficient. As in an absorption experiment, a resonant interaction is signified by a reduction in the transmission coefficient. Perhaps in a solid-state environment, absorption implies the conversion of optical energy into heat. In this sense absorption spectroscopy is something of a misnomer for our experiment as at low temperature, resonant optical excitation of a quantum dot does not create heat but rather causes light to be Rayleigh scattered.[7] The laser field generates a quantum dot field and the detector placed directly behind the sample measures the total intensity, i.e., a time average of the interference of the two fields. In an engineering language, a small signal, the quantum dot field, is detected by mixing it with a large signal, the laser field, a homodyne detection.

To implement this concept, we couple laser light from a tunable narrowband (1 MHz) laser into an optical fibre which we use as a spatial filter. The fibre output is then focused to a diffraction-limited spot on the surface of the semiconductor heterostructure with a colimating lens and objective with NA either 0.55 or 0.65. The transmission through the sample at 4.2 K is detected with an in situ, unbiased p-i-n diode and ex situ, low-noise current preamplifier. The dots are embedded in a vertical field effect transistor, with an n^+-layer as back contact and on-surface Schottky barrier as gate electrode. By applying a voltage to the Schottky barrier, we can exploit Coulomb blockade to control the electron number on a single quantum dot,[8] and for a given charge state, we can exploit the Stark effect to manipulate the resonance energy.[9] Typically, we perform our experiments by applying a dc and ac bias to the gate voltage for a constant laser wavelength. We measure the ac signal on the detector, recording it as we sweep through the resonance with the dc bias. The ac modulation is crucial as it enables us to reject noise in all but a small bandwidth of frequency.

Example data in Fig. 1 show the transmission coefficient as a function of dc bias for a single quantum dot recorded with an integration time of 0.1 s per point. There is a clear resonance with Lorentzian line shape. In this particular experiment, the contrast at resonance is 2.4%, the linewidth is 2.0 μeV (480 MHz), close to the radiative limit which corresponds to about 0.8 μeV for these dots based on a

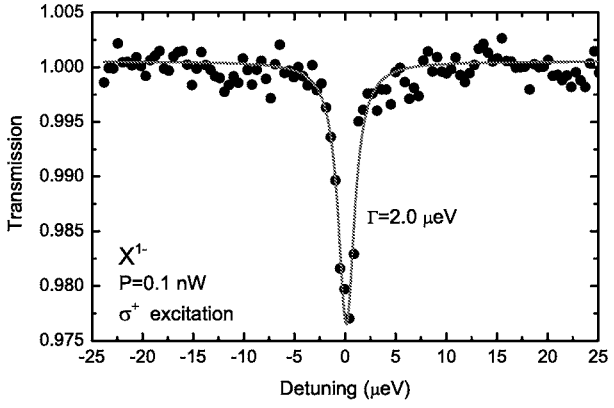

Fig. 1. Laser spectroscopy on a single self-assembled InAs/GaAs quantum dot: transmission coefficient recorded versus detuning. The laser wavelength was a constant 950 nm with power 0.1 nW, focused to a diffraction-limited spot of FWHM 380 nm, achieved using a NA = 0.65 objective and glass ($n = 2.0$) hemispherical solid immersion lens in contact with the sample. The detuning was achieved with the Stark effect in a vertical electric field with the dc bias chosen to select the X^{1-} resonance. The sample temperature is 4.2 K. The solid line represents a fit to a Lorentzian line shape with FWHM linewidth 3.3 μeV.

radiative lifetime of 800 ps. The increase in lifetime above the theoretical minimum may well be related to a spectral fluctuation, a random walk in the dot resonance energy, rather than reflect a dephasing process.[1]

3. Laser Spectroscopy of a Single Quantum Dot

3.1. *Spectroscopy in zero magnetic field*

Fig. 2 shows laser spectroscopy recorded on a single neutral exciton. The high spectral resolution of the experiment reveals that the neutral exciton, X^0, resonance consists of two closely-spaced transmission dips with linear orthogonal polarizations (Fig. 2), whereas the charged exciton resonance, X^{1-}, consists of just one transmission dip (Fig. 1). The origin of the X^0 splitting is the so-called fine structure arising from the exchange interaction between the electron and hole. The pure exciton angular momentum states with $L_z = \pm 1$ are mixed by the anisotropic part of the electron-hole exchange, resulting in two eigenstates, $\frac{1}{\sqrt{2}}(|+1\rangle \pm |-1\rangle)$, which couple to the vacuum state with different linear polarizations, E_x, E_y. The splitting arises therefore through a departure from ideal symmetry; this is most likely a shape anisotropy but it is important to note that the lack of inversion symmetry in the underlying lattice also induces a fine structure, albeit with a magnitude thought to be smaller than the one observed in the present experiment.[10] In the case of X^{1-}, the exciton state consists of a spin-paired electron doublet occupying the lowest energy electron shell and a single hole. The net electron spin is zero, eliminating the exchange interaction with the hole, and removing the fine structure.

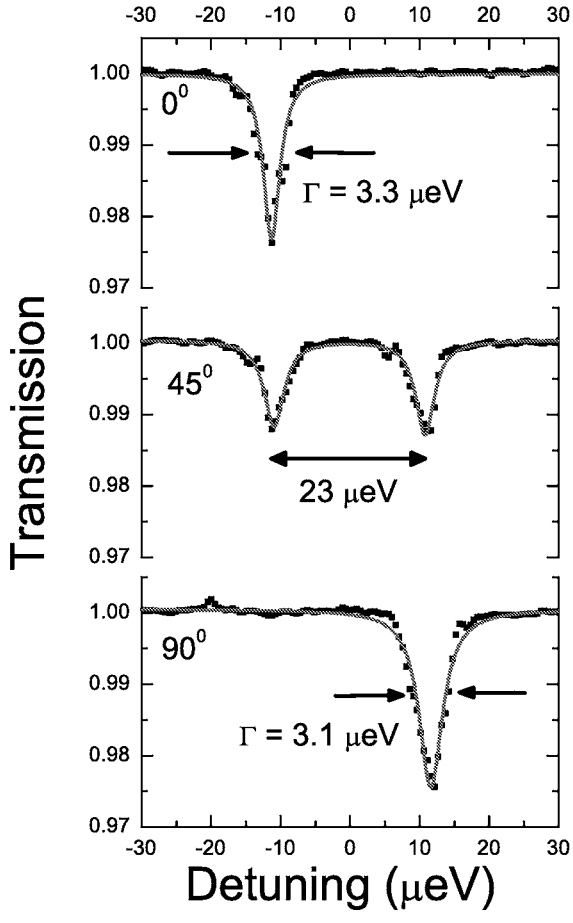

Fig. 2. Transmission laser spectroscopy on a neutral exciton X^0 in a single quantum dot at 4 K with linearly polarized laser light in three directions, 0, 45 and 90 degrees. The points are the measured data points; the solid lines fit to Lorentzian line shapes. The splitting is the fine structure, arising from the anisotropic electron-hole exchange interaction.

We find that the X^0 contrast is twice that of the X^{1-} resonance when the polarization is chosen to maximize the strength of one of the X^0 resonances.[11] The explanation is that in the case of X^{1-}, the resident electron has a spin which fluctuates randomly on the time scale of the experiment. This means that the dot is equally likely to be bright or dark with respect to the probing laser resulting in a 50% loss of contrast.

3.2. *Spectroscopy in an Applied Magnetic Field*

The selection rules can be manipulated by applying a magnetic field which we have explored in the Faraday geometry with the magnetic field aligned along the growth

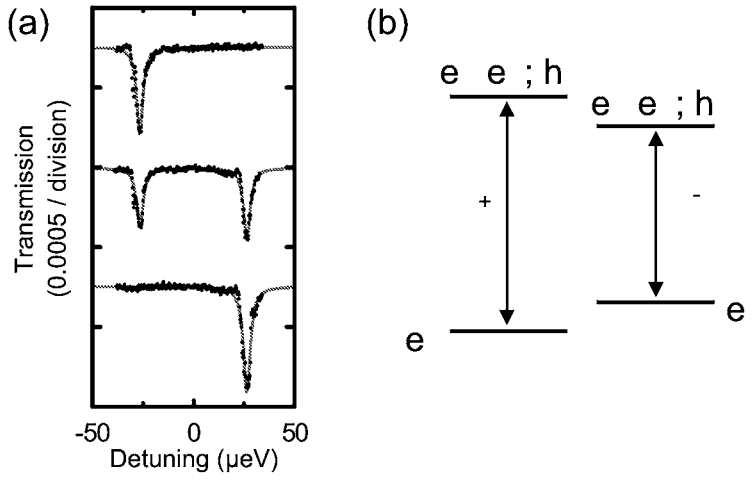

Fig. 3. (a) Laser spectroscopy on a single X^{1-} in a magnetic field. The single line, Fig. 2, splits into two as a result of the spin Zeeman effect. (b) Energy level diagram of a singly-charged quantum dot in a magnetic field.

direction of the sample. The magnetic field introduces a spin Zeeman term into the Hamiltonian. In the case of X^{1-}, the initial state of the optical transition is split by the electron spin Zeeman effect, and the final state is split by the hole spin Zeeman effect, Fig. 3(b). The X^{1-} resonance therefore splits into two with increasing magnetic field with the upper energy branch σ^+-polarized, the lower energy branch σ^--polarized, Fig. 3(a). For X^0, the initial state depends on both electron-hole exchange and electron and hole spin Zeeman effects. The net effect is that the splitting increases above the zero field splitting as the magnetic field increases with elliptically polarized lines. In the limit that the exciton Zeeman energy is larger than the zero field fine structure, the fine structure is a minor perturbation, and the two lines become circularly polarized, exactly like the X^{1-}. This is achieved for magnetic fields of $\sim$ 1 T for the dot in Fig. 3. The remarkable result is that the polarization selection rules of a quantum dot can be manipulated both by charging and by applying a magnetic field.

At larger magnetic fields, the upper energy X^{1-} branch gains in strength over the lower energy branch.[2]. The explanation is that the electron spin in the initial state of the optical transition occupies preferentially the lower energy spin state simply through Maxwell–Boltzmann statistics. This provides a way to estimate the electron g-factor (the spectroscopic splitting measures the exciton g factor not the bare carrier g factors); it also provides a brute force method of initializing the electron spin. However, this is unattractive as at 4 K, very large magnetic fields are required to achieve a high spin polarization. This is not only technically demanding but also problematic for a spin qubit as the rate of phonon-related spin relaxation processes increases as B^5.[12]

4. Optical Pumping of a Single Electron

The electron spin relaxation time in a quantum dot is limited by the hyperfine interaction with the nuclear spins at low magnetic fields (up to a few hundred mT)[6,5] and by phonon-related processes at high fields[12] with a minimum at modest fields lying somewhere around $\sim$ 1 ms. In our device, a co-tunneling process can also relax electron spin. In this process, the quantum dot electron spin exchanges its spin with the spin of an electron close to the Fermi energy in the Fermi sea.[13] While this process can be strongly suppressed by operating at the center of the charging plateau,[13] our current thinking is that for the devices of Fig. 3 (25 nm tunnel barrier), co-tunneling limits the spin relaxation time to a few hundred ns even in modest magnetic fields. However, this process can be controlled through the electron tunneling time which we have recently adjusted by increasing the thickness of the tunneling barrier, the barrier separating dot and back contact. This small change to the device causes the X^{1-} contrast to behave in quite a different way.

Figure 4 shows the contrast in the X^{1-} transmission spectroscopy as a function of bias. The resonance exists in a bias window corresponding to the bias range in which the single electron forms the lowest energy initial state, simply as a consequence of the Coulomb blockade. At zero magnetic field, the X^{1-} resonant contrast is large; in magnetic field, the X^{1-} contrast remains large at the edges of the plateau but is

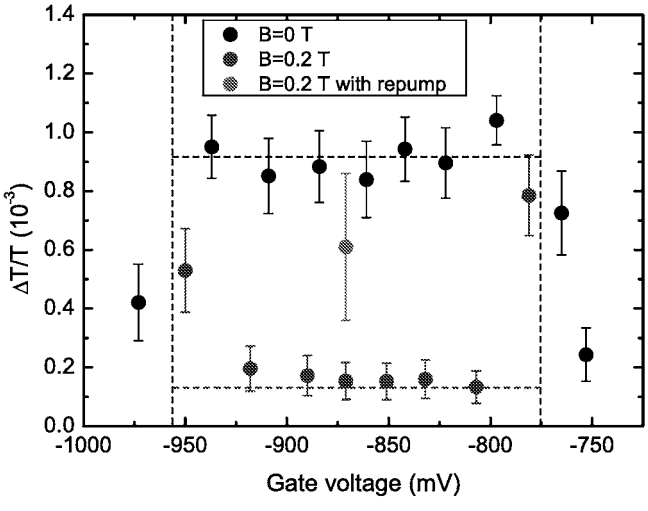

Fig. 4. Contrast at the X^{1-} resonance in the transmission coefficient as a function of bias, V_g. The vertical dashed lines mark the charging voltages. Data are shown for $B = 0$ (black points) and for $B = 0.2$ T (blue points). The horizontal solid lines represent the averaged contrast away from the charging voltages. The red point marks the contrast when the dot is pumped with two lasers, one tuned to the σ^+ resonance and the other to the σ^- resonance, with equal intensities such that the total intensity is the same as for the single laser experiment.

suppressed in the center of the plateau. Our interpretation is an optical pumping process: the laser, tuned to the σ^+ resonance, pumps the electron into the spin-down state where it cannot scatter light, leading to a loss of contrast. At the edges of the plateau, spin swap with the continuum makes this process very inefficient; at the center of the plateau, spin swap is much reduced, and optical pumping becomes effective. We can verify this interpretation by pumping both spin transitions simultaneously. We use a second laser with σ^- polarization tuned in energy such that it comes into resonance with the dot at exactly the same bias as the first laser with σ^+ polarization. With both lasers, we find that the transmission contrast reappears. This is important as it demonstrates for instance that the electron remains

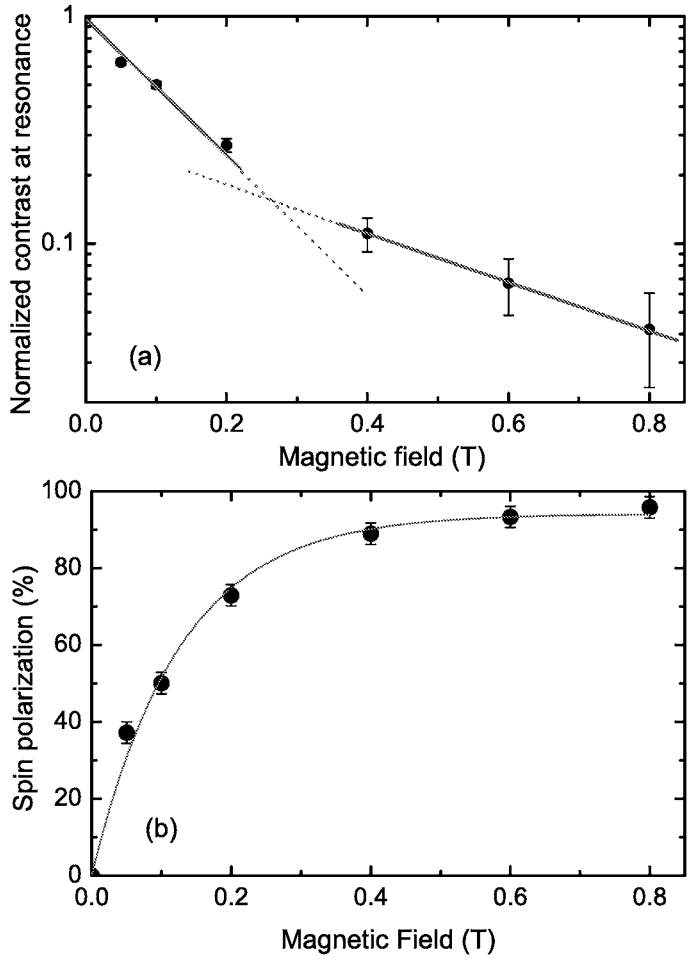

Fig. 5. (a) Resonant contrast vs. magnetic field for a single X^{1-}. The data have been normalized to the contrast at zero magnetic field. The solid lines are exponential fits to the data in two regimes of magnetic field. (b) Spin polarization versus magnetic field for the same data as in (a).

in the dot, enabling us to relate the loss of contrast in the single laser experiment to electron spin.

Our results show that the spin polarization is 96% and that the signal recovers to ~65% of its zero field value on repumping. The ETH group has reported a spin polarization as high as 99.8% with total recovery on repumping.[3] In both experiments, the fidelity has a strong dependence on field below 1 T, Figure 5, with hints of a double exponential behavior in our data. This is caused in large part by electron spin relaxation through the hyperfine interaction with the nuclei.

This scheme for optically pumping a single spin requires the optically-induced spin flip rate, γ_o, to be much larger than the electron spin relaxation rate, γ_e. The optical spin-flip mechanism could be a spin-flip Raman transition via the exciton state[3] or possibly very weak spontaneous emission from $X^{1-} \uparrow$ to $e \downarrow$, allowed perhaps by a small in-plane magnetic field. Presently, we have insufficient experimental data to distinguish between the two. The polarization of 96% implies that $\gamma_e/\gamma_0 \sim 0.01$; further work needs to determine these rates independently.

5. Conclusions and Outlook

Laser spectroscopy of a single quantum dot has been used to determine the line shapes, the linewidth, the selection rules and the energy splittings of the fundamental optical excitations of a single quantum dot. While this remains a demanding experiment, recent results have increased the contrast at resonance to 6% and allowed us to reduce the integration time to ~100 ms while still achieving acceptable signal:noise. By using this sensitive probe, we find that the quantum dot is uniquely tunable: its energies can be controlled with an electric field through the Stark effect; its natural polarization basis can be changed from linear to circular with a magnetic field; and spin relaxation can be controlled over orders of magnitude with both applied bias and an applied magnetic field. The resonant interaction with the laser allows us both to probe and to pump the quantum dot; a laser cooling of spin represents the first profound example of using the laser as a pump.

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