Inversion recovery measurements of exciton fine-structure beats in a single quantum dot

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Abstract. We present a method for time-resolving the coherent precession of the spin of a single neutral exciton confined to an InAs/GaAs quantum dot. A pair of circularly polarized π pulses are used to pump and probe the exciton spin, rather than a pair of interferometrically stable π/2-pulses in a quantum interference type experiment, faster, technically simpler, measurements of the small 15 µeV exciton fine-structure can be made in the photocurrent regime. A small change in the fine-structure splitting with vertically applied electric-field is observed.

1. Introduction
Semiconductor quantum dots possess a small anisotropy due to a combination of bulk asymmetries [1], spin-orbit coupling [2], piezo-electric, and dot elongation [3]. This anisotropy breaks the spin degeneracy of the neutral exciton, resulting in a small fine-structure splitting in the energies of the bright exciton states.

The fine-structure splitting is a property of practical importance, being the main limitation on the fidelity of dot-based polarization entangled photon sources [4], and also spin preparation techniques based on exciton ionization [5]. This motivates strong efforts to suppress the fine-structure splitting using post growth annealing [6], lateral magnetic-fields [7], lateral electric-fields [8, 9, 10], and optical Stark shifts [11]. The fine-structure splitting can be measured using polarization resolved single dot cw spectroscopy techniques such as micro-PL [2, 7, 3, 12], transmission [11], or photocurrent [13]. The fine-structure splitting can be deduced from the exciton spin beats observed in ensembles of dots [14, 6, ?, 15], or at a single dot level using quantum interference [16, 13], and recently four-wave mixing techniques [17].

Here we employ the inversion recovery method [18] to observe the exciton spin precession of a single quantum dot using photocurrent detection. Compared to quantum interference based techniques for observing the fine-structure beats [16, 13], an inversion recovery method has a number of technical advantages. It is a simpler technique that does not require a stabilized interferometer, has a faster acquisition time, and the signal is less sensitive to the pulse-area, and therefore more robust against the mechanical instability of the setup.

2. Principle of experiment
The bright exciton is composed of an electron with $m_J = \mp 1/2$, and a heavy-hole of $m_J = \pm 3/2$, and is created using light of $\sigma_{\pm}$ circular polarization respectively. The electron-hole exchange
interaction couples the exciton spin-states, to form linearly spin-polarized energy-eigenstates $|\alpha\rangle = (|\uparrow\rangle \pm |\downarrow\rangle)/\sqrt{2}$, where the label $\alpha = x, y$. These states are separated in energy by the fine-structure splitting $h\omega_{fs}$, and have linearly polarized optical selection rules.

A pair of picosecond laser pulses resonantly excite the neutral exciton transition with a pulse-area of $\pi$. The pulses arrive with a time delay of $\tau$ between them. Since the time-duration of the laser pulses is short compared with the period of the fine-structure beat, the pulse drives a Rabi rotation between the crystal ground-state $|0\rangle$, and the exciton spin up/down state selected by the $\sigma_\pm$ circular polarization of the pulse respectively. For a pulse-area of $\pi$ the pulse inverts the occupations of these two states. Initially the dot is in the crystal ground-state. At time $t = 0$, the first $\sigma_+$-polarized pulse prepares a spin-up exciton. This is a superposition of the $x,y$ energy eigenstates, which beat due to their energy separation: $|\psi\rangle = \cos(\omega_{fs}t/2)|\uparrow\rangle + \sin(\omega_{fs}t/2)|\downarrow\rangle$.

The exciton spin freely evolves due to the fine-structure beats and electron tunneling for a time $\tau$.

The absorption of the second pulse at time $t = \tau$ depends on the population of the exciton spin state that is probed. The $\pi$-pulse inverts the occupations of the crystal-ground-state, and the exciton spin state selected by the polarization of the laser pulse. This results in a change in the final exciton population, and therefore a change in photocurrent that is proportional to the population inversion between the selected exciton spin and the crystal-ground state. Hence, a photocurrent signal that oscillates between the exciton spin up/down states due to the fine-structure beat is observed.

A calculation of the measured photocurrent per pulse-pair is: $PC = e\nu_{rep}\int_0^\infty \Gamma_t(t)\rho_{XX} dt$, where $\rho_{XX} = (\rho_{\uparrow\uparrow} + \rho_{\downarrow\downarrow})$ is the total exciton population. The density-matrix operator is defined as: $\rho_{\alpha\beta} = |\alpha\rangle \langle \beta|$, with a corresponding expectation value of $\langle \rho_{\alpha\beta} \rangle$. The probability that the state $\langle \alpha = 0, \uparrow, \downarrow \rangle$ is occupied at time $t$ is: $\rho_{\alpha\alpha}(t)$. $\Gamma_t$ is the tunneling rate, and $\nu_{rep}$ is the repetition rate of the laser, which for this experiment gives $e\nu_{rep} = 12.18$ pA. The integral can be split into before, and after the probe pulse. The first integral is proportional to the change in the exciton population during $(0 < t < \tau)$: $PC_1(\tau) = e\int_0^\infty \Gamma_t(t)\rho_{XX}(t) dt = e\nu_{rep}\eta(f - \rho_{XX}(\tau))$, where $\eta$ is the photocurrent detector efficiency, and $f$ is fidelity of the inversion operation performed by the laser pulse.

The second integral is proportional to the exciton population after the $\alpha$-polarized probe has been applied: $PC_2(\tau) = e\nu_{rep}\int_\tau^{\infty} \Gamma_t(t)\rho_{XX}(t) dt = e\nu_{rep}\eta(f\rho_{00}(\tau) + \rho_{\alpha\alpha}(\tau) + (1 - f)\rho_{\alpha\alpha}(\tau))$. A comparison of the photocurrent signals measured for co and cross-circular excitation gives:

$$PC_{--} - PC_{++} = e\nu_{rep}\eta f(\rho_{\uparrow\uparrow}(\tau) - \rho_{\downarrow\downarrow}(\tau))$$

where $\Gamma_e, \Gamma_h$ are the electron and hole tunneling rates respectively, and $\Gamma_s \approx \Gamma_e$ is the relaxation rate of the exciton spin. The exciton spin should exhibit a damped-cosine behavior due to the fine-structure beat [14].

3. Experiments and discussion

The experiments were performed on an n-i-Schottky diode with a single layer of low density InAs/GaAs quantum dots. The results presented are from a single dot emitting at 951 nm in the high energy tail of the dot-distribution. The two identical pulses have a Gaussian shape, with a Fourier transform limited full-width half-maximum of 0.2 meV. The photodiode sits in a cold-finger cryostat, at a temperature of about 15 K. Further details of both the device, and the setup can be found in ref. [18].

A set of polarization resolved inversion recovery measurements at various gate voltages is presented in figure 1. As discussed earlier, a pair of circularly polarized laser pulses excite the
Figure 1. Inversion recovery measurements of exciton spin. (a) Photocurrent versus time-delay for on-resonant excitation of neutral exciton transition with two $\pi$ pulses. The red (black) traces correspond to co (cross) circular polarized excitation schemes. (b) The difference in photocurrent ($PC_{-+} - PC_{++}$) between cross and co-circular excitation, which is proportional to the exciton spin, versus time-delay. The traces are calculated using data in (a), and the red-lines show fits to data using eq. (1). The fitting parameters are displayed in (c,d). (c) The decay rate $\Gamma_x$, due to electron tunneling versus gate voltage. (d) Small change in exciton fine-structure with vertically applied electric-field.

dot on resonance with the neutral exciton transition with a pulse-area of $\pi$, and the resulting photocurrent is measured as a function of the inter-pulse time-delay. For co (cross) circular excitation the data are shown as red (black) traces in fig. 1(a) respectively. The pulse-area is calibrated by measuring a Rabi rotation [18].

The time-resolved data are presented in figs. 1(a,b). Figure 1(a) shows a comparison of the photocurrent versus time-delay measured for a pair of co and cross-circularly polarized laser pulses that oscillate in anti-phase. The oscillation is due to the fine-structure beat as discussed above. Figure 1(b) shows the difference between the photocurrents measured for cross and co-circular excitation, and are calculated using the data in fig. 1(a). The signal is proportional to the net exciton spin: $(\rho_{\uparrow\uparrow} - \rho_{\downarrow\downarrow})$, and exhibits a damped-cosine behavior, due to the precession of the spin and a decay due to electron tunneling that increases with applied gate voltage. The red-lines shows the fits to eq. (1) that are used to extract the fine-structure and the electron tunneling rate, which are presented in figs. 1(c) and (d) respectively.

The data presented in fig. 1(a) is symmetric with respect to time-delay, because the two pulses are identical except for their polarization, and the spin-insensitive photocurrent detection only responds to the relative polarization of the laser pulses. For near zero time-delay, where the pulses are not temporally overlapped, the photocurrent signal is low for co-circular excitation, because the two $\pi$-pulses act as a single $2\pi$-pulse and de-excite the exciton. Whereas for cross-
circular excitation the second pulse does not interact with the spin-up exciton prepared by the first pulse, resulting in a net photocurrent equivalent to one exciton. As the electron tunnels from the dot, the exciton spin state relaxes to a hole-state, resulting in the convergence of the co/cross-circular photocurrent signals as seen in fig. 1(a). Both photocurrent signals saturate at a time-delay that is long compared with all tunneling times, since each pulse creates an exciton that subsequently tunnels resulting in a net photocurrent equivalent to two excitons per pulse-pair for both co and cross-circular excitation.

When the pulses are temporally overlapped there is a peak (dip) in the photocurrent for co (cross) circular excitation respectively, as seen in fig. 1(a). In the case of co-polarized excitation, the interference of the pulses results in an effective single pulse excitation with a pulse-area varying between $0 - 2\pi$, resulting in a peak of non-zero net-photocurrent. For cross-polarized excitation, the temporally overlapped pulses act as a single pulse with a pulse-area of $\pi\sqrt{2}$, and a linear polarization with an orientation angle equal to the inter-pulse phase. Ideally this would create $\sin^2(\pi\sqrt{2}/2)$ excitons per pulse-pair, versus one exciton per pulse-pair when the pulses are no longer overlapped, resulting in the observed dip in photocurrent.

Figure 1(d) shows a small decrease in the exciton fine-structure splitting with increasing vertically applied electric-field. A small change of about 3 $\mu$eV is observed for a change in gate-voltage from 0.5-0.8 V, with an electric-field gradient of $dE_{fs}/dF = -2.9 \pm 0.2$ $\mu$eV.V$^{-1}$.µm. This is of a similar value to previous measurements using a micro-PL technique [19, 20]. We note that this change in the fine-structure splitting is small compared with the 80 $\mu$eV achieved using a lateral electric-field [8, 10]. We speculate that this may be due to a decrease in the overlap of the electron and hole wavefunction as the electric-field increases the electron-hole separation, resulting in a decrease in the electron-hole exchange energy.

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