REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE	3. DATES COVERED (From - To)
	New Reprint	-

4. TITLE AND SUBTITLE

Picosecond optical spectroscopy of a single negatively charged self-assembled InAs quantum dot

5a. CONTRACT NUMBER

W911NF-08-1-0487

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

411359

6. AUTHORS

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5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES

University of Michigan - Ann Arbor
Regents of the University of Michigan
3003 S. State St
Ann Arbor, MI 48109 -1274

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSOR/MONITOR'S ACRONYM(S)

ARO

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

55014-PH-QC.2

12. DISTRIBUTION AVAILIBILITY STATEMENT

Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES

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14. ABSTRACT

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15. SUBJECT TERMS

SEMICONDUCTOR; SPIN

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Duncan Steel
UU	UU	UU	UU		19b. TELEPHONE NUMBER: 734-764-4469

Report Title

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REPORT DOCUMENTATION PAGE (SF298)
(Continuation Sheet)

Continuation for Block 13

ARO Report Number 55014.2-PH-QC
Picosecond optical spectroscopy of a single neg ...

Block 13: Supplementary Note

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Picosecond optical spectroscopy of a single negatively charged self-assembled InAs quantum dot

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(Received 8 July 2010; accepted 18 August 2010; published online 17 September 2010)

We demonstrate an experimental technique for the transient read-out of the optical transitions in a single negatively charged self-assembled InAs quantum dot (QD) using resonant picosecond optical pulses and control of the QD charge state. Observable phenomena include trion (negatively charged exciton) decay, Rabi oscillations between the electron and trion states and the precession of electron and heavy-hole spins about an externally applied dc magnetic field. © *2010 American Institute of Physics*. [doi:10.1063/1.3487783]

Self-assembled quantum dots (QDs) grown by molecular beam epitaxy provide a versatile system both for investigations into fundamental light-matter interactions[1,2] and for a number of device applications.[3–5] For both pursuits, it is often crucial to be able to characterize the transient behavior of individual dots to determine the time scales for excited state relaxation and resident carrier spin dephasing. For the most part, time-domain studies of single self-assembled QDs have been limited to either photoluminescence[6] or photocurrent[7] type measurements due to the difficulties imposed by the small optical dipole moments involved.[8] These approaches rely on either sufficiently isolated radiative decay channels or fast carrier tunneling rates for read-out. Transient absorption based read-out to enable nonlinear optical spectroscopy and control measurements provides an alternative approach free from such restrictions.

Here, we report an experimental technique utilizing electrical control of the QD charge state and phase-sensitive optical detection to perform time-resolved read-out of an InAs QD containing a single electron on picosecond timescales. At zero magnetic field, pump-probe studies show the excitation and decay of the trion (negatively charged exciton) and are used to demonstrate coherent control of the electron-trion transition. In the presence of an external dc magnetic field applied perpendicular to the QD growth axis (Voigt geometry), we observe the independent precession of the electron and heavy-hole spins, providing a possible means of extracting spin coherence times.

The sample under study contains a single layer of self-assembled InAs QDs grown on a GaAs substrate in a Schottky diode structure that enables selective control of the number of electrons in a given dot via an externally applied bias voltage. An Al mask containing apertures approximately 1 μm in diameter allows for transition energy selective optical excitation of single QDs. The sample is placed in a magnetic He-flow cryostat to achieve operating temperatures of approximately 5 K. A tunable 76 MHz Ti:sapph laser generating mode-locked sech pulses approximately 2 ps in width is used as the excitation source.

We first perform Stark-shift modulation absorption studies[9] on a selected QD with a narrow-bandwidth continuous-wave (cw) optical field to determine the range of trion absorption energies E_{trion} as a function of the sample bias voltage V_{dc}, as plotted in Fig. 1(a). This range is limited by the range of V_{dc} values for which the QD contains a single electron. Outside this voltage range, the QD optical transition energies are shifted by a few to several millielectron volts due to Coulomb shifts associated with the different QD charge states.[10]

We utilize these shifts to observe the interaction between the QD and resonant picosecond optical pulses incident along the QD growth axis. Specifically, we measure the time-averaged transmission of the pulses on a square-law detector. For a sample bias V_R within the trion absorption voltage range, each pulse generates an optical polarization within the QD. This polarization radiates an optical field that interferes with the transmitted pulse on the detector. The amplitude of this field is proportional to both the encountered difference in occupation probabilities between the optically coupled QD

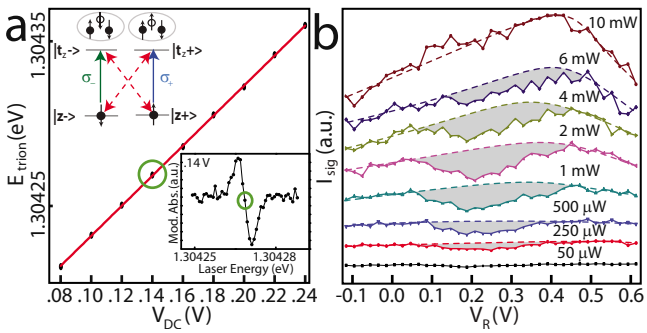

FIG. 1. (Color online) (a) E_{trion} as a function of V_{dc} at zero magnetic field where the solid curve is a linear fit. The upper-left inset shows the optical transitions involving the electron spin ground states and the trion states. The lower-right inset shows a modulation absorption scan taken at a sample bias of 0.14 V with a cw optical field and small voltage modulation. Since both the modulated Stark shift and the cw linewidth are smaller than the trion linewidth, the obtained signal is the derivative of the trion absorption line (see Ref. 9). The energy of the zero crossing point (circle) corresponds to the trion transition energy. (b) I_{sig} measurements (offset) for a single pulse train as a function of V_R for different average pulse powers. The shaded regions indicate the change in the transmission with respect to a voltage dependent background (dashed lines) due to detection of the QD radiated field.

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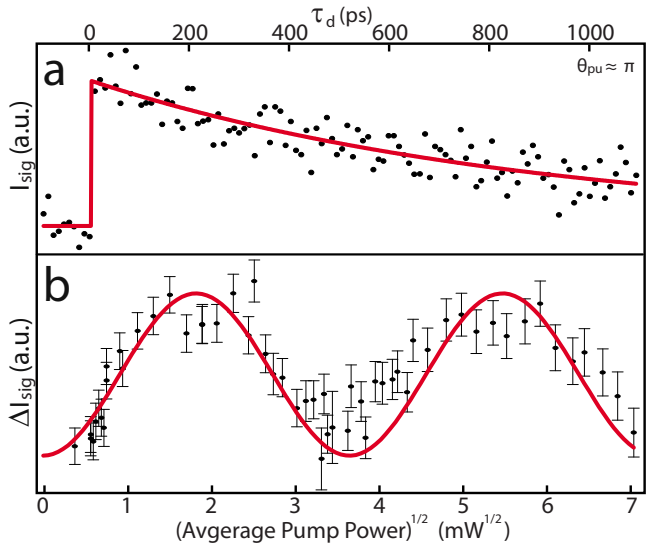

FIG. 2. (Color online) (a) $I_{sig}(\tau_d)$ measurements taken at zero magnetic field. The solid line is a fit of the data using the second term of Eq. (1). (b) ΔI_{sig} measurements as a function of the square root of the average pump power for $\tau_{\pm}=\pm 50$ ps and a probe pulse area of approximately $2\pi/5$. The solid line is an oscillatory fit of the data.

levels and the pulse area. For a sample bias V_L outside the trion absorption range, each pulse interacts negligibly with the QD due to the Coulomb shift of several pulse bandwidths, essentially quenching the QD radiated field. Thus, modulation of the sample bias between values V_L and V_R effectively modulates the interference signal, enabling the use of phase-sensitive homodyne techniques to detect the QD radiated field. Further, we note that the pulse repetition period of 13.2 ns is much longer than trion lifetimes measured in these systems,[6,11] thus permitting time-averaged measurements.

Figure 1(b) plots measurements of the time-averaged transmission for a train of resonant pulses as a function of V_R at different average powers. In each trace, the sample bias voltage is modulated with a ~4 kHz square-wave between $V_L=-1$ V and V_R, the former value serving to shift the QD off resonance by ~5 meV. The transmission is represented as I_{sig}, which is proportional to the detector photocurrent measured at the sample bias modulation frequency. Results reveal a voltage dependent background signal that becomes more pronounced at higher powers (dashed lines). The cause of this background is not entirely known at present but appears to arise from a broad continuum of optically active states that coincides with QD energies as seen in photoluminescence studies. On top of this background, a power-dependent dip (shaded region) in I_{sig} emerges as V_R is scanned through the trion absorption voltage range due to the detected QD field. This dip covers the entire trion absorption range (~150 μeV) since this range is much smaller than the bandwidth of each optical pulse (~1 meV). The area of the dip bears an oscillatory dependence on the pulse power consistent with a pulse driven trion Rabi oscillation, the frequency corresponding to a trion dipole moment of approximately 8 Debye.

We now utilize this technique in pump-probe studies to observe transient phenomena, setting V_R to the center of the dip of Fig. 2(b). Pulse delays τ_d are kept to values much smaller than the pulse repetition period to preserve the ability to perform time-averaged measurements. In studies without an external magnetic field, an *H*-polarized pump pulse is used to generate equal trion population in both transitions (taking both transition dipoles to be equal) that decays back to the electron spin ground states at a rate of Γ_t. The subsequent *V*-polarized probe pulse, cross polarized to enable postsample filtering of the pump, encounters the same occupation probability difference $\Delta\rho$ in each transition. An analytical expression for $\Delta\rho$ may be obtained by solving for the matrix elements of the density operator $\hat{\rho}$ for one of the trion transitions and has the form

$$\Delta\rho(t) = -1/2 + \Theta(t)\sin^2(\theta_{pu}/2)e^{-\Gamma_t t}, \tag{1}$$

where $-1/2$ is the occupation probability difference prior to excitation at 5 K, Θ is the Heaviside function, and $\sin^2(\theta_{pu}/2)$ is the trion population generated by a pump of pulse area θ_{pu}. Since each transition contributes equally to the QD radiated field, transmission measurements of the probe pulse yield $I_{sig}(\tau_d)$ values proportional $\Delta\rho(\tau_d)$ when accounting for the signal background.

Figure 2(a) plots $I_{sig}(\tau_d)$ for pump and probe fields of pulse area π and $2\pi/5$, respectively. The predicted exponential decay of the signal is clearly evident. Fitting the data to the second term of Eq. (1) yields a trion lifetime of 855 ± 74 ps, consistent with previously reported values. Further, from the second term of Eq. (1) we see that pump-driven trion Rabi oscillations may be observed for lower average powers incident on the detector as a result of postsample pump filtering. We observe these Rabi oscillations by measuring the difference in I_{sig} between a positive delay τ_+ and negative delay τ_-, i.e., $\Delta I_{sig}=I_{sig}(\tau_+)-I_{sig}(\tau_-)$, as a function of pump pulse area. These difference measurements account for a power dependent offset in I_{sig} that arises from the leakage of the pump to the detector. Two Rabi oscillations are plotted in Fig. 2(b) as a function of the square root of the average pump power for $\tau_{\pm}=\pm 50$ ps. An oscillatory fit of the data is given by the solid curve in Fig. 2(b), showing qualitative agreement between theory and experiment. These results confirm the dipole moment of ~8 D determined from the single pulse studies of Fig. 1(b).

Application of an external dc magnetic field in the Voigt geometry lifts the Kramer's degeneracy for the electron states and the trion states, enabling the observation of both electron and the heavy-hole spin precession. A resonant σ_+ polarized pump pulse serves to generate a stimulated Raman coherence between the spin states of both the electron and the heavy-hole via two-photon processes, thereby initializing oppositely oriented electron and heavy-hole spin polarization vectors along the optical axis. These spin polarization vectors then precess about the magnetic field axis at rates determined by the electron and heavy-hole in-plane g-factors, which generally differ. Over the course of the precession, the magnitudes of the optically induced spin polarization vectors decay in time. The rate of decay for the electron is determined by the electron spin dephasing time, while the rate of decay for the heavy-hole is determined primarily by the trion relaxation time. These decaying spin precessions manifest as damped oscillations in the value of $\Delta\rho$ encountered by the time-delayed σ_- polarized probe pulse in the $|z-\rangle$ to $|t_z-\rangle$ transition.

As with zero-field studies, an expression for $\Delta\rho$ may be obtained by solving for the density matrix elements of the

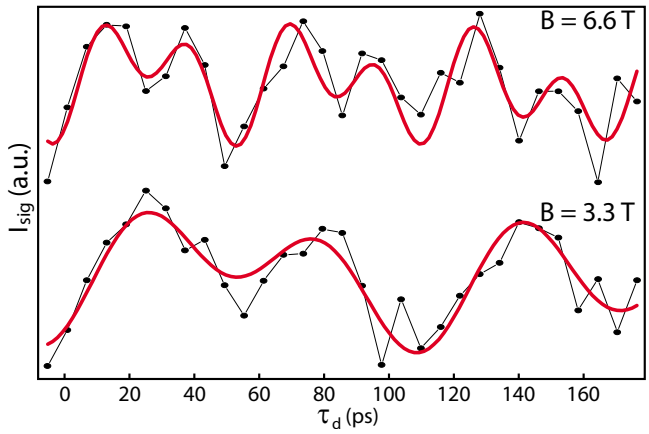

FIG. 3. (Color online) $I_{sig}(\tau_d)$ measurements taken at magnetic field (B) values of 3.3 T (bottom) and 6.6 T (top). As the pump-probe delay range is short compared to previously observed electron and heavy-hole coherence times, the data are fit to Eq. (2) taken in the limit of very long spin coherence times (solid curves).

system, though here all four levels must be considered due to the presence of the magnetic field. In this case, $\Delta\rho$ has the form

$$\Delta\rho(t) = -1/2 + (1/4)\Theta(t)\sin^2(\theta_{pu}/2) \times \{e^{-2\Gamma_t t}[2 - e^{-t/T_2^{h*}}\cos(\omega_h t)] - e^{-t/T_2^{e*}}\cos(\omega_e t)\}, \quad (2)$$

where $-1/2$ is the occupation probability difference prior to excitation, $\omega_{e(h)}$ is the electron (heavy-hole) spin precession frequency and $T_2^{e(h)*}$ is the inhomogeneously broadened coherence time of the electron (heavy-hole) spin due to the fluctuating nuclear spin environment in the dot. $I_{sig}(\tau_d)$ measurements at external magnetic field values of 3.3 and 6.6 T are plotted in Fig. 3, exhibiting two-frequency modulations caused by the electron and heavy-hole spin precessions. The limited range of pump-probe delays in Fig. 3 prevents accurate extraction of the electron and heavy-hole spin coherence times, as this range is much shorter than reported coherence times for both the electron[12] and the heavy-hole[13] in similar dots. Hence, the data are simply fit to Eq. (2) assuming no spin dephasing (solid curves of Fig. 3). The agreement between the fitting and the data suggests spin coherence times that are much longer than the pump-probe delay values considered. From the fitting, we obtain electron and heavy-hole g-factor magnitudes $|g_e|$ and $|g_h|$ of 0.378 ± 0.007 and 0.202 ± 0.006, respectively, in agreement with separate frequency domain measurements of the electron and heavy-hole Zeeman splittings (data not shown).

We have presented an experimental technique enabling the optical characterization of single self-assembled QDs on picosecond timescales. Use of this technique with methods such as balanced detection may provide improved signal to noise ratio to allow for the extraction of spin coherence times. Further, this technique is also readily applicable to other systems such as quantum dot molecules[14] whose optical transition energies may be sufficiently shifted via an external voltage.

This work was supported in part by ARO (IARPA, W911NF-08-1-0487), AFOSR (FA9550-09-1-0457), ONR (NSA), DARPA (W911NF-09-1-0406), and NSF (PHY 0114336 and PHY 0804114).

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