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

Erik D. Kim,1,a) Katherine Truex,1 Yanwen Wu,1 A. Amo,1 Xiaodong Xu,1 D. G. Steel,1,b) A. S. Bracker,2 D. Gammon,2 and L. J. Sham3

1H. M. Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan 48109-1120, USA
2Naval Research Laboratory, Washington, DC 20375-5347, USA
3Department of Physics, University of California, San Diego, California 92093-0319, USA

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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 interactions1,2 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 photoluminescence6 or photocurrent7 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:sapphire laser generating mode-locked sech pulses approximately 2 ps in width is used as the excitation source.

We first perform Stark-shift modulation absorption studies9 on a selected QD with a narrow-bandwidth continuous-wave (cw) optical field to determine the range of trion absorption energies $E_{\text{trion}}$ as a function of the sample bias voltage $V_{\text{dc}}$, as plotted in Fig. 1(a). This range is limited by the range of $V_{\text{dc}}$ 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 and the QD without the pulse train. The absorption $T$ is

$$T(v, V_R) = |r|^2 + 2\Re\{r| + 2\Im\{r|,$$

where $|r|$ is the overlap integral of the pulse with the field radiated by the QD and $\Re\{r|$ and $\Im\{r|$ give the real and imaginary parts of this overlap integral.

![Fig. 1](image-url)
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, allowing 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 $\sim 4$ kHz square-wave between $V_L = -1$ V and $V_R$, the former value serving to shift the QD resonance frequency, and $\tau_\pm$ is the Heaviside function, and $\sin^2(\theta_{pu}/2)$ is the trion population generated by a pump of pulse area $\theta_{pu}$. Since each transition contributes equally to the QD radiated field, transmission measurements of the probe pulse yield $\Delta \alpha$ values proportional $\Delta \alpha$ when accounting for the signal background.

Figure 2(a) plots Isig versus the probe field for a train of resonant pulses as a function of power and $\pi$ 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 $855 \pm 74$ ps, consistent with previously reported values. Further, 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 post-sample pump filtering. We observe these Rabi oscillations by measuring the difference in Isig between a positive delay $\tau_+$ and negative delay $\tau_-$, i.e., $\Delta \alpha = \alpha_{\pi} + \alpha_{2\pi/5}$, as a function of pump pulse area. These difference measurements account for the power dependent offset in Isig 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 $\sim 8$ 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 $\sigma_+$ 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 $\Delta \alpha$ encountered by the time-delayed $\sigma_-$ polarized probe pulse in the $|z\rangle$ to $|t\rangle$ transition.

As with zero-field studies, an expression for $\Delta \alpha$ may be obtained by solving for the density matrix elements of the 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 $\Gamma_e$. The subsequent $V$-polarized probe pulse, cross polarized to enable post-sample 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 density matrix elements of the
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

$$\Delta \rho(t) = -1/2 + (1/4)\Theta(t)\sin^2(\theta_{pu}/2) \times \left\{ e^{-t/T_2^{(h)}} \cos(\omega_{h}t) - e^{-t/T_2^{(e)}} \cos(\omega_{e}t) \right\}, \tag{2}$$

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. $I_{s}(\tau_{d})$ 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 and the heavy-hole 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 whose optical transition energies may be sufficiently shifted via an external voltage.

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