Fast Optical Preparation, Control, and Readout of a Single Quantum Dot Spin

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We propose and demonstrate the sequential initialization, optical control, and readout of a single spin trapped in a semiconductor quantum dot. Hole spin preparation is achieved through ionization of a resonantly excited electron-hole pair. Optical control is observed as a coherent Rabi rotation between the hole and charged-exciton states, which is conditional on the initial hole spin state. The spin-selective creation of the charged exciton provides a photocurrent readout of the hole spin state.

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The ability to sequentially initialize, control, and readout a single spin is an essential requirement of any spin based quantum information protocol [1]. This has not yet been achieved for promising schemes based on the optical control of semiconductor quantum dots [2]. These schemes seek to combine the picosecond optical gate speeds of excitons [3–6], with the potential for millisecond coherence times of quantum dot spins [7–9], by optically manipulating the spin via the charged exciton. This results in a system where the potential number of operations before coherence loss could be extremely high, in the range 10^{4-9}, and in a system compatible with advanced semiconductor device technologies. A number of important milestones have recently been reached, but these focus on the continuous initialization of an electron [10,11] or hole spin [12], detection of a single quantum dot spin [13,14], or optical control of ensembles of 10^{6-7} spins [15,16].

In this Letter, we demonstrate sequential triggered on-demand preparation, optical manipulation, and picosecond time-resolved detection of a single hole spin confined to a quantum dot, thus demonstrating an experimental framework for the fast optical manipulation of single spins. This is achieved using a single self-assembled InGaAs quantum dot embedded in a photodiode structure. The hole spin is prepared by ionizing an electron-hole pair created by resonant excitation. A second laser pulse then drives a coherent Rabi oscillation between the hole and positive trion states, which due to Pauli blocking is conditional on the initial hole spin state, key requirements for the optical control of a spin via the trion transition. Because of Pauli blockade, creation of the charged exciton provides a photocurrent readout of the hole spin state.

First we will describe the principle of operation. The qubit is represented by the spin states of the heavy hole ($J = \frac{3}{2}$), where logical states 0 (1), are the spin up (down) states ($m_J = \pm\frac{3}{2}$). Figure 1 shows an idealized quantum dot, embedded in an n-i-Schottky diode structure. An electric field is applied, such that the electron tunneling rate is much faster than the hole tunneling rate. The experiments use a sequence of two circularly polarized, time-separated laser pulses, with a time duration shorter than the electron tunneling time, labeled the "preparation" and "control" pulses. Figure 1 illustrates the steps (a)–(d) involved in the preparation and readout of the hole spin.

Preparation.—(a) The circularly polarized preparation pulse resonantly excites the ground-state neutral exciton transition ($0 - X^0$), driving a Rabi rotation through an

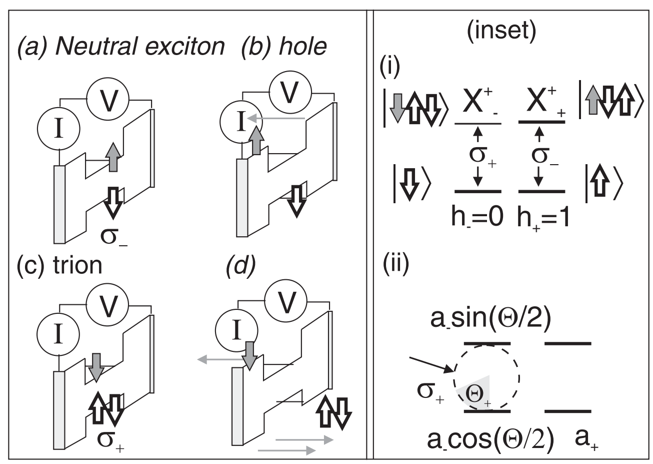

FIG. 1. Illustration of operating principle. *Preparation:* (a) Resonant excitation of the $0 - X^0$ transition creates a spin-polarized electron-hole pair. Filled (open) arrows are electron (hole), respectively. (b) Under applied electric field the electron tunnels from the dot, leaving a spin-polarized hole. *Readout:* (c) A circularly polarized π pulse creates a charged exciton only if the hole is in the target spin state. (d) Carriers tunnel from the dot, the creation of a charged exciton resulting in a change in the photocurrent proportional to the occupation of the target hole spin state. Inset: *Control* (i) Energy levels of heavy-hole or charged-exciton system which acts as two decoupled 2-level atoms: $h_\pm - X^+_\pm$. (ii) Driving a Rabi rotation with σ_+ circular polarization addresses the $h_- - X^+_-$ transition only.

angle equal to the pulse area of π [17]. This creates a spin-polarized electron-hole pair with near unit probability. (b) Under the action of the applied electric field the electron will tunnel from the dot, resulting in a photocurrent proportional to the final exciton population of up to one electron per pulse, which for a 76-MHz repetition rate is 12.18 pA [3,4]. Since the electron tunneling rate is much faster than for the hole, the electron tunnels from the dot leaving a spin-polarized hole.

Control.—(inset of Fig. 1) To control the $h - X^+$ transitions, the following control scheme is used. Because the spin lifetimes are long compared with the duration of the control laser pulse, the heavy-hole or charged-exciton 4-level system acts as two decoupled 2-level atoms, as illustrated in the inset of Fig. 1. The optical selection rules are a result of Pauli blocking, with each hole spin state coupling to a single auxiliary state: $|h_\pm\rangle - |X_\pm^+\rangle$. For a laser on resonance with the $h - X^+$ transition the control Hamiltonian is [17]

$$\hat{H} = \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_+ & 0 & 0 \\ \Omega_+ & 0 & 0 & 0 \\ 0 & 0 & 0 & \Omega_- \\ 0 & 0 & \Omega_- & 0 \end{pmatrix},$$

where $\Omega_\pm$ are the Rabi frequencies of the circular polarization components of the control laser, and the basis is $|\psi\rangle = (|h_-\rangle, |X_-^+\rangle, |h_+\rangle, |X_+^+\rangle)$. The control laser pulse implements the unitary operation $\hat{U}(\Theta_+, \Theta_-) = \exp(\frac{i}{\hbar} \times \int H dt)$:

$$\hat{U} = \begin{pmatrix} \cos(\frac{\Theta_+}{2}) & -i\sin(\frac{\Theta_+}{2}) & 0 & 0 \\ -i\sin(\frac{\Theta_+}{2}) & \cos(\frac{\Theta_+}{2}) & 0 & 0 \\ 0 & 0 & \cos(\frac{\Theta_-}{2}) & -i\sin(\frac{\Theta_-}{2}) \\ 0 & 0 & -i\sin(\frac{\Theta_-}{2}) & \cos(\frac{\Theta_-}{2}) \end{pmatrix},$$

where $\Theta_\pm = \int \Omega_\pm dt$ are the pulse areas of the circularly polarized laser components. Control of the phase of the hole spin could be achieved as follows. A σ_+ polarized control laser addresses one transition only. For an initial state of $|\psi\rangle = (a_-, 0, a_+, 0) \equiv (a_-, a_+)$, a σ_z gate imparting a relative phase shift of π between the hole spin states would be implemented when $\Theta_- = 0$, $\Theta_+ = 2\pi$:

$$\hat{U} \rightarrow \hat{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

The phase shift arising from a 2π Rabi rotation has been verified in four-wave mixing experiments on the neutral exciton of an interface dot [6]. Further discussion of this control scheme can be found in Refs. [17,18].

Readout.—(c) and (d) Creation of the charge exciton results in a change in the photocurrent signal, and hence a readout proportional to the probability that the hole is in the target spin state at that instant in time.

Full details of the device can be found in Ref. [19], where inversion recovery measurements on this dot confirm that the neutral exciton coherence is limited by electron tunneling. Because of the electron-hole exchange interaction the exciton transitions are linearly polarized and have a fine-structure splitting of $h/(230 \pm 10\text{ ps})$. Resonant excitation in step (a) with circular polarization creates a spin-polarized exciton. A combination of the fine-structure beat and the time for the electron to tunnel leads to some loss of spin polarization. However, at the reverse bias of 0.8 V used in the experiment, the electron tunneling rate ($\Gamma_e^{-1} = 35$–40 ps) is fast compared with the period of the fine-structure beat, minimizing any loss of spin orientation, and is slow enough to observe weakly damped Rabi oscillations (see below). At the same time, the slow hole tunneling rate ($\Gamma_h^{-1} \sim$ ns) is much faster than the repetition rate of the laser ensuring the dot is initially in the crystal ground state.

Figure 2 presents one- and two-color photocurrent spectra to show the preparation and detection of a single hole spin. The lower trace of Fig. 2 presents the case of single pulse excitation. A single peak corresponding to the neutral exciton transition ($0 - X^0$) is observed, with line shape determined by the Gaussian pulse shape (FWHM = 0.2 meV).

In the case of two-color excitation, a preparation pulse with pulse area π is tuned to the $0 - X^0$ transition, to create a neutral exciton with a probability close to 1. The photocurrent is then recorded as a function of the detuning of the control pulse, which also has a pulse area of π. The middle traces of Fig. 2 show two-color photocurrent spectra for a time delay of 7 ps, much shorter than the electron tunneling time. For copolarized pulses there is a dip at the ($0 - X^0$)

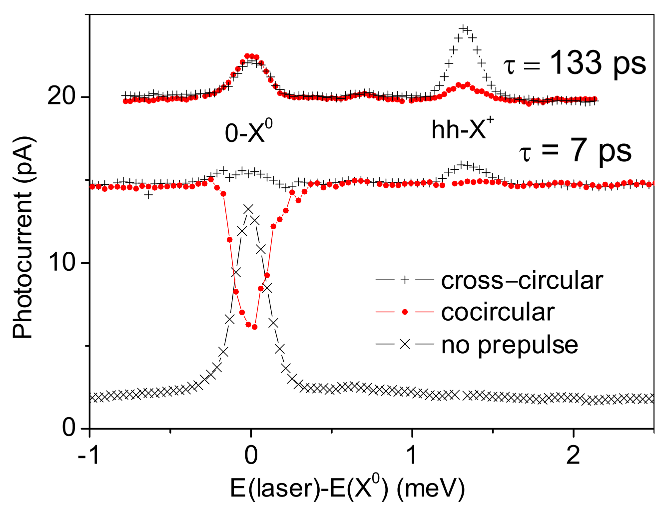

FIG. 2 (color online). Photocurrent versus laser detuning. (Lower part) Single circular polarized π pulse, with $0 - X^0$ peak. (Middle and upper parts) Two pulse spectra, with preparation pulse resonant with $0 - X^0$ transition, with short (long) time delay. The upper trace is offset for clarity. Note the emergence of the $h - X^+$ line at longer time delays for cross-polarized excitation.

transition, since the pulse pair is now equivalent to a 2π pulse. For the cross-polarized case there is only a very weak $(0 - X^0)$ feature, but importantly there is an additional peak at a detuning of $+1.32$ meV, corresponding to the heavy-hole to charged-exciton transition $(h - X^+)$ [20]. As the time delay increases, the electron tunnels from the dot, and the heavy-hole population grows, as seen in the upper traces of Fig. 2. At a time delay of 133 ps, which is much longer than the 35–40 ps electron tunneling time, the exciton is completely ionized, resulting in a weak polarization insensitive $(0 - X^0)$ peak and a stronger polarization sensitive $h - X^+$ peak.

The $(0 - X^0)$ and the $(h - X^+)$ features in the two pulse spectra have the opposite selection rules. For $(0 - X^0)$, the Coulomb interaction shifts the energy of the biexciton by -2.16 meV and out of resonance with the spectrally narrow laser pulse preventing the absorption of the cross-polarized control pulse. In the case of the positive trion, absorption of a copolarized pulse is forbidden by the Pauli exclusion principle, since it would result in two holes of the same spin, as shown in the inset of Fig. 1. By contrast, cross-polarized excitation of the positive trion results in a change in photocurrent proportional to the occupation of the target hole spin state. The energy separation between the X^0 and X^+ transitions is in close agreement with photoluminescence measurements.

From the amplitudes of the $h - X^+$ peaks for cross (4.2 pA) and copolarized (0.88 pA) excitation at a time delay of 133 ps, we deduce that when there is a hole, there is at least an 83% probability of the hole occupying the desired spin state. At 133 ps, there is also a $0 - X^0$ peak, indicating an approximately 20% probability of the dot occupying the crystal ground state, implying that no hole of either spin has been prepared, possibly due to radiative recombination of the neutral exciton. This demonstrates steps (a)–(d) in Fig. 1, showing preparation and detection of a single spin.

Figure 3(a) presents time-resolved measurements from which the heavy-hole population can be deduced. The preparation pulse creates a neutral exciton, while the control pulse of pulse area π, resonant with $h - X^+$, probes the population of the target hole spin state. For cross-circular excitation an exponential rise is observed as the electron tunnels from the dot, and the heavy-hole population increases until saturation [as illustrated in Fig. 1(b)]. The hole population has an exponential rise time of 40 ps, consistent with the electron tunneling time [19]. After the fast initial rise the hole population slowly decays with a lifetime in excess of 600 ps. Because of the electron-hole exchange interaction of the neutral exciton, the hole ends up with the opposite spin about 20% of the time, resulting in a slower rise of the copolarized signal [21]. This is the first time-resolved measurement of a single quantum dot spin with subnanosecond time resolution.

To demonstrate control of the $h - X^+$ transition, as depicted in the inset of Fig. 1, we study the Rabi rotation

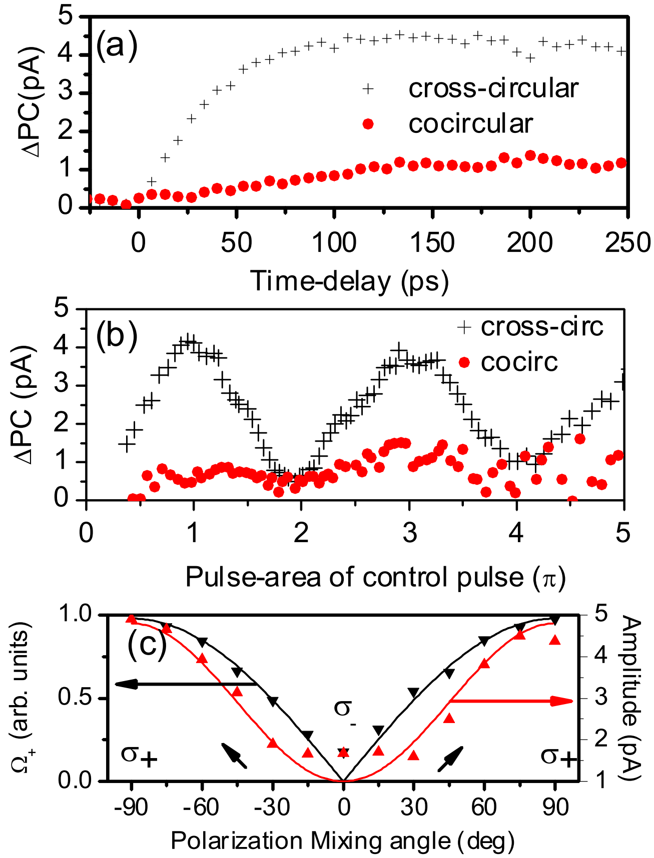

FIG. 3 (color online). (a) Time-resolved measurements of heavy-hole population. Photocurrent versus time delay between prepulse and a control pulse resonant with the $h - X^+$ transition. (b) Rabi rotation of the $h - X^+$ transition, where creation of the charged exciton is conditional on the heavy-hole spin state. The π pulse provides a readout of the hole spin state. (c) Polarization dependence $(\Omega_-, \Omega_+) \equiv \Omega[\cos(\alpha), \sin(\alpha)]$ of the $h - X^+$ Rabi rotation. (▼) Inverse period of $h - X^+$ Rabi rotation versus polarization angle α of the control pulse, for a σ_- preparation pulse. The line shows the $|\sin(\alpha)|$ dependence confirming the independence of the $h_\pm - X^+_\pm$ transitions. (▲) Amplitude of Rabi rotation for σ_+-polarized control pulse versus the polarization angle of the preparation pulse.

of the transition. Figure 3(b) shows the photocurrent versus pulse area of the control pulse, at a time delay of 133 ps. Two pulses are incident on the sample: the preparation pulse and a control pulse of variable pulse area resonant with the $h - X^+$ transition. A background photocurrent linear in power has been subtracted [3,4]. For cross-circular excitation more than two periods of a weakly damped Rabi oscillation are observed. For cocircularly polarized excitation, the Rabi rotation is suppressed. The results in Fig. 3(b) demonstrate a Rabi rotation of a charged exciton conditional on the initial spin state. Previous reports of a Rabi oscillation of a charged exciton were for *uninitialized* spins, in both an ensemble of quantum dots [15] and for an excited state charged exciton of unknown charge [22].

To confirm that the 4-level $h - X^+$ system behaves as two decoupled two-level optical transitions, which are rotated by $\hat{U}$ when excited by the control laser, we studied the polarization dependence of the $h - X^+$ Rabi rotation. In the first experiment a σ_- preparation pulse is used to create an initial state which is predominantly $|h_-\rangle$: $|\psi\rangle \approx (1, 0, 0, 0)$. The Rabi rotation is then measured as a function of the polarization of the control pulse, defined as $(\Omega_-, \Omega_+) \equiv \Omega[\cos(\alpha), \sin(\alpha)]$. The amplitude of the Rabi rotation is almost constant, but the inverse period is equal to the $|\sin(\alpha)|$ amplitude of the σ_+ component of the Rabi frequency of the control pulse, as seen in Fig. 3(c). This demonstrates that the $|h_\mp\rangle$ state only interacts with $\sigma_\pm$ polarized light.

In the second experiment, the polarization of the control pulse is fixed at σ_-, and the Rabi rotation is measured as a function of the polarization of the preparation pulse. The period of the Rabi rotation is constant, but the amplitude exhibits a $\sin^2(\alpha)$ dependence reflecting the occupation of the $|h_-\rangle$ state, as shown in Fig. 3(c). This demonstrates that the polarization of the preparation pulse can be used to control the initial populations of the hole spin states $|h_\pm\rangle$, in the mixed state. The distinct $|\sin(\alpha)|$ and $\sin^2(\alpha)$ dependencies of these measurements further confirm that $\hat{U}$ is a good approximation of the action of the control pulse.

Armed with tools for the sequential initialization and readout of a single spin, a number of future experiments are now possible. For example, a magnetic field in the Voigt configuration may be used to achieve an arbitrary phase shift on a single spin [23]. To observe the precession of the hole spin a preparation and readout pulse sequence would be applied. The data presented here strongly suggest that when a third circularly polarized control pulse with a pulse area of 2π is applied, an operation $\hat{U}(0, 2\pi) \approx \hat{\sigma}_z$ will induce a relative phase shift of π between the hole spin states, resulting in a phase jump in the spin precession. The phase shift can then be controlled using the detuning of the laser [24].

To summarize, using a photodiode structure we demonstrate sequential initialization, coherent optical control, and photocurrent readout of a single hole spin. This work establishes an experimental platform for investigating optical control of single quantum dot spins, which marries the ultrafast coherent control of excitons with the long coherence times of spin based qubits.

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Note added.—Following submission of this work, Mikkelsen *et al.* [25] reported the coherent precession of a single electron spin.

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