Coherent Writing of the Dark Exciton State Using One Picosecond Long Optical Pulse

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Abstract: We demonstrate a one to one correspondence between the polarization of a picosecond optical pulse and the coherent spin state of the long lived dark exciton that it deterministically photogenerates in a single quantum dot.
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The ability to coherently control and exploit matter two level systems is essential for realizations of future technologies based on quantum information processing (QIP). Optical approaches, in particular, are preferred since they are state-selective, require no contacts, and are ultrafast. Spins of charge carriers in semiconductors play an important role among the matter venues of choice since they dovetail with contemporary leading technologies in general and those of light sources and detectors in particular. For these reasons, optical writing, readout and control of spins in semiconductor quantum dots (QDs) have been the subjects of many recent works. Indeed, semiconductor QDs provide an excellent interface between single photons and isolated matter spins due to their superior coupling to light.

The fundamental optical excitation of semiconductors is the promotion of an electron across the forbidden band-gap from the full valence band to the empty conduction band. An absorbed photon, thereby, leaves in matter a valence band hole – conduction band electron pair with opposite spin directions, or a bright exciton. The bright exciton forms a spin integer (total spin 1) two level system (qubit) in the matter. We have demonstrated that, in strain-induced self-assembled quantum dots, the polarization of a resonantly tuned, single picosecond optical pulse can be used for deterministic writing the exciton spin qubit in any desired coherent state [1]. Such a process is not possible for single spins, where a few pulses are required to prepare the spin in an eigenstate and then a three step (Ramsey) rotation is needed for writing the spin state at will. Moreover, full coherent control ("rotation") of the bright exciton can be achieved by one single optical pulse [2], while single spins require two pulses and free precession in between [3]. These advantages of the bright exciton, however, are not very useful, since its lifetime is rather short (sub ns) limited by radiative recombination of the electron-hole pair.

Since light barely interacts with the electronic spin, an electron-hole pair with parallel spin directions is almost optically inactive. Such a pair is called a dark-exciton. The dark exciton is also a spin integer (total spin 2) qubit [4] but since it is optically inactive, its lifetime can be orders of magnitude longer than that of the bright exciton [5]. Very recently we have demonstrated that the (DE) lifetime (~1 μs) is radiative and that it maintains coherence over at least ~100 ns [6]. As such the DE has clear advantages over the bright exciton, provided that it can be externally accessed, despite its optical inactivity.

Here, we show for the first time that, just like the case of the bright exciton, the polarization of a resonantly tuned, single picosecond optical pulse can be used for high fidelity deterministic writing of the dark exciton qubit in any desired coherent state. Thus, the time needed to write the DE qubit state deterministically is 5-6 (4-5) orders of magnitude shorter than its lifetime (coherence time). Consequently, the dark exciton forms an excellent matter qubit.

In Fig. 1a, we schematically describe the relevant energy levels, spin wavefunctions and optical transitions between these levels. In Fig. 1b, we describe the sequence of pulses used in the experiment, where color coding is used to match the optical transitions in Fig. 1a. The first ps pulse (green upward arrow) tuned to an excited dark exciton state, and variably polarized as described by the Poincare sphere in Fig. 1c, writes the dark exciton. The absorption of a photon from the second, circularly polarized, 10 ns pulse (red upward arrow) probes the temporal evolution of the dark exciton spin. The absorption is measured by the intensity of the emitted biexciton photon (downward blue arrow). The measured degree of circular polarization of the emission as a function of time after the writing pulse and the polarization of the writing pulse are presented in Fig. 1d, 1e and 1f for $P(\theta,\phi)$= $P(0<\theta<2\pi, 3\pi/2)$, $P(0<\theta<2\pi,0)$ and $P(\pi/2,0<\phi<2\pi)$, respectively. In Fig. 1g, the temporal dependence of the degree of circular polarization is presented for various writing pulse polarizations. The selected polarizations $\vec{P}(\pi,0)=\hat{V}$ $\vec{P}(\pi/2,\pi/2)=\hat{\bar{D}}$ and $\vec{P}(\pi/2,\pi)=\hat{R}$ are presented by the blue, green and azure lines, respectively. One can clearly see in Figs. 1d-f that the initial amplitude and phase of the circular polarization oscillations correspond to the angles θ and ϕ, which describe the polarization of the writing pulse, thus establishing a one to one

correspondence between the writing pulse polarization and the dark exciton coherent spin state. The maximal degree of polarization that we obtain (~0.65) is compatible with the finite temporal resolution of our detectors (~450 ps) [4], evidencing high fidelity dark exciton writing.

Our ability to write the optically inactive dark exciton with a short laser pulse is due to dark/bright exciton mixing induced by the reduced symmetry of the QD [7]. We exploit this mixing by applying a polarized optical pulse tuned to an excited DE resonance (green upward arrow) where this mixing is enhanced relative to the mixing in the ground state [6-7]. The photogenerated excited DE relaxes to its ground level via a spin preserving phonon (downward spiral grey arrows).

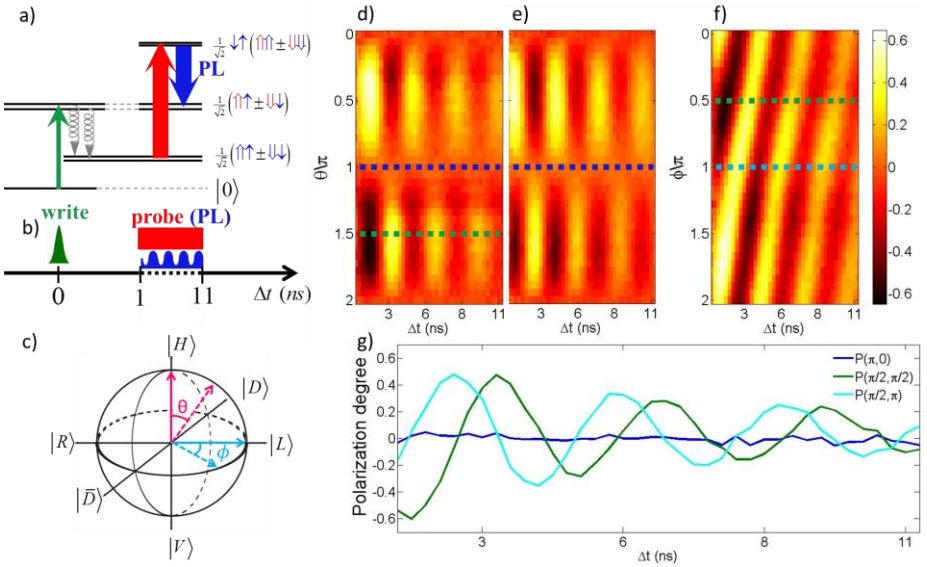

Fig.1 (a) Schematic description of the method for writing the DE spin and reading it out. Horizontal lines with spin wave functions to their left describe the energy of the relevant excitonic and biexcitonic states. ↑ (⇓) represents spin up (down) electron (heavy hole). The blue (red) arrow represents carrier in the ground (excited) energy level. Long vertical arrows denotes resonant optical transition – green from the vacuum to an excited dark-exciton excited by a few-picoseconds long pulse, red from the dark-exciton to a spin blockaded biexciton excited by a 10ns cw window and blue emission from the biexciton to the dark-exciton. Curly downward arrows represent non-radiative spin-preserving heavy hole transitions. Note that due to the 1.4 μeV energy splitting between the dark exciton eigenstates, coherent superpositions of its eigenstates precess in time with period of 3.09 ns. (b) The temporal sequence of the pulses in the experiment and the resulted emission. (c) The polarization of the optical pulse used for writing is represented on the Poincare sphere $\vec{P}(\theta,\phi)=\exp(-i\phi/2)\cos(\theta/2)\hat{H}+\exp(i\phi/2)\sin(\theta/2)\hat{V}$. (d-f) The degree of circular polarization of the biexciton emission as a function of the time from the writing laser pulse and the writing laser polarization as obtained by subtracting the PL excited by left-hand circular probe pulse from the PL excited by right-hand polarized probe pulse and dividing by their sum. The writing laser polarization was $\vec{P}(\theta,\phi)=\vec{P}(\theta,3\pi/2)$, $\vec{P}(\theta,\phi)=\vec{P}(\theta,0)$ and $\vec{P}(\theta,\phi)=\vec{P}(\pi/2,\phi)$ respectively. (g) Polarization degree as function of time from the writing laser pulse at $\vec{P}(\pi,0)=\hat{V}$, $\vec{P}(\pi/2,\pi/2)=\hat{\bar{D}}$ and $\vec{P}(\pi/2,\pi)=\hat{R}$ polarizations. The colors match the colors of the horizontal lines in (d-f). Writing in $\hat{\bar{D}}$ and $\hat{R}$ polarizations results in oscillations with maximal amplitude and relative expected delay of quarter period, while in $\hat{V}$ polarization there are no oscillations at all as expected from an eigenstate.

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