Coherent Optical Writing and Reading of the Exciton Spin State in Single Quantum Dots

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We demonstrate a one-to-one correspondence between the polarization state of a light pulse tuned to neutral exciton resonances of single semiconductor quantum dots and the spin state of the exciton that it photogenerates. This is accomplished using two variably polarized and independently tuned picosecond laser pulses. The first "writes" the spin state of the resonantly excited exciton. The second is tuned to biexcitonic resonances, and its absorption is used to "read" the exciton spin state. The absorption of the second pulse depends on its polarization relative to the exciton spin direction. Changes in the exciton spin result in corresponding changes in the intensity of the photoluminescence from the biexciton lines which we monitor, obtaining thus a one-to-one mapping between any point on the Poincaré sphere of the light polarization to a point on the Bloch sphere of the exciton spin.

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Coherent manipulation of quantum states is a critical step towards applications in quantum information processing. The atomiclike spectrum of semiconductor quantum dots (QDs) and their compatibility with modern microelectronics make them promising candidates for forming the building blocks of these future technologies. In particular, they form an excellent interface between flying photonic qubits and anchored matter-spin qubits. The coherent properties of spins of confined carriers and pairs of carriers (excitons) in QDs have been demonstrated by various experimental ways over the years [1–9]. The fundamental optical excitations of QDs, the exciton and pair of excitons (biexciton), have been proposed [10] and demonstrated [3] as coherent physical realizations of qubits and quantum logic gates.

In this Letter, we demonstrate a new method for initializing ("writing") the spin state of a QD confined exciton in *any* coherent superposition of its eigenstates by a single, resonantly tuned, polarized picosecond light pulse. Likewise, we show that the spin state of the initialized exciton can be determined ("readout") using a second, delayed, polarized picosecond light pulse, resonantly tuned into specific biexcitonic resonances. Figure 1(a) is an energy level diagram which schematically describes the processes involved in writing and reading the excitonic spin state. The first polarized laser pulse is resonantly tuned into either a ground or an excited exciton state to photogenerate an exciton. From the excited state, the exciton rapidly relaxes nonradiatively to its ground state, while preserving its initial spin (as demonstrated below).

In order to relate the polarization of the light to the spin state of the photogenerated exciton we note that the angular momentum projection of right-(left-) hand circularly polarized light R (L) in the direction of propagation is 1 (-1). Upon electron-heavy hole (e-h) pair generation, the electron spin ($\frac{1}{2}$) is oriented downward (upward) while the heavy-hole spin ($\frac{3}{2}$) is oriented upward (downward) such that the total angular momentum projection is conserved. We associate the spin state of such a pair with the polarization of the light by defining $|R\rangle = \Uparrow\downarrow$ ($|L\rangle = \Downarrow\uparrow$). Here a spin up (down) electron is denoted by $\uparrow$ ($\downarrow$) and a spin up (down) heavy hole is denoted by $\Uparrow$ ($\Downarrow$). With this notation

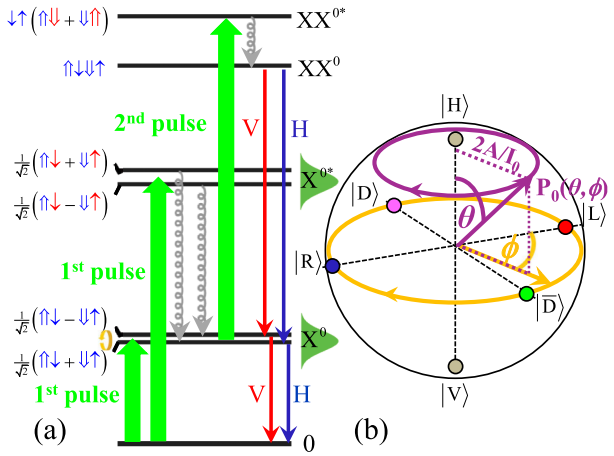

FIG. 1 (color online). (a) Schematic description of the writing and reading processes. Horizontal lines describe the states' energies. The spin wave functions of these states are depicted to the left. $\uparrow$ ($\Downarrow$) represents spin up (down) electron (hole). The blue (red) arrow represents carrier in the ground (excited) energy level. Upward (green) arrows denote resonantly tuned light pulses; first, short (long) to a ground (excited) exciton and second, to an excited biexciton state. Curled downward arrows denote nonradiative relaxation and a blue (red) downward arrow radiative H (V) polarized recombination. The linewidth of the laser pulses are given by the curves to the right. (b) A Bloch sphere representation of the exciton spin state initiated by the polarized laser pulse. The point $P_0(\theta, \phi)$ represents an arbitrarily polarized spin state. A and I_0, relate to the biexciton PL intensity [see Fig. 3(a)].

it is straightforward to show that the correspondence between the horizontal, vertical, diagonal, and cross-diagonal linear polarizations of the exciting light (H, V, D, and $\bar{D}$, respectively) and the spin state of the photogenerated pair are given by $|H\rangle = 1/\sqrt{2}(\Uparrow\downarrow + \Downarrow\uparrow)$, $|V\rangle = -i/\sqrt{2}(\Uparrow\downarrow - \Downarrow\uparrow)$, $|D\rangle = e^{-i\pi/4}/\sqrt{2}(\Uparrow\downarrow + i\Downarrow\uparrow)$, and $|\bar{D}\rangle = e^{i\pi/4}/\sqrt{2}(\Uparrow\downarrow - i\Downarrow\uparrow)$. The direction in space of the H polarization coincides with the direction of the natural major axis of the QD [3]. These spin states are described on the Bloch sphere of the exciton spin in Fig. 1(b). An arbitrarily elliptically polarized pulse is described by a point on the surface of the Poincaré sphere. Such a point can be viewed as having two components: a component on the equator plane (containing the L and D directions), deflected by an angle ϕ from L, and a component parallel to the rectilinear H-V axis. Thus, two angles completely define an arbitrary polarization, the angle ϕ and the angle θ between the polarization and the H-V axis. In a complete analogy, an arbitrary exciton spin state is described as a point on the Bloch sphere. The north and south poles of the Bloch sphere denote the exciton symmetric and antisymmetric eigenstates, $|H\rangle$ and $|V\rangle$, respectively. However, due to the anisotropic e-h exchange interaction, these eigenstates are not degenerate, even in the absence of externally applied magnetic field [3,11,12].

A resonantly tuned H (V) polarized laser pulse photogenerates an exciton in its symmetric (antisymmetric) spin eigenstate $|H\rangle$ ($|V\rangle$). When the pulse resonates with the excited state, the generated excited exciton relaxes nonradiatively into its ground state before recombination occurs. The exciton then remains in its eigenstate until it radiatively recombines. However, since the two eigenstates are not degenerate, they evolve at different temporal paces. Therefore, any other coherent superposition of these eigenstates precesses in time at a frequency given by the difference between the eigenenergies, divided by the Planck constant. Such excitation requires, however, a resonant pulse of spectral width which contains both eigenstates. For example, the orange circle on the equator of the sphere describes the evolution of an exciton spin excited by a resonant L polarized pulse excitation. Such a pulse initiates the two spin eigenstates with equal probabilities. The initiated spin then precesses clockwise with time, such that the angle ϕ equals $\frac{\pi}{2}$, π, $\frac{3\pi}{2}$, and 2π after $\frac{1}{4}$, $\frac{1}{2}$, $\frac{3}{4}$, and 1 period, respectively, while the spin state becomes $|\bar{D}\rangle$, $|R\rangle$, $|D\rangle$, and $|L\rangle$ again, respectively. The purple circle on the sphere describes precession of an exciton spin, photogenerated with $P_0(\theta, \phi)$ spin by an arbitrarily polarized pulse.

Delayed by $\Delta\tau$ from the first pulse, a second, polarized pulse, is then applied. This pulse is tuned to an excited resonance of the biexciton, to avoid scattered light from the detector. The probability to photogenerate a biexciton depends on the orientation of the exciton spin relative to the polarization of the second pulse, since the biexciton resonance used here contains two electrons in a singlet state [Fig. 1(a)]. Therefore, by monitoring the photoluminescence (PL) from the biexciton doublet as a function of $\Delta\tau$, one obtains direct information on the evolving exciton spin state. As schematically described by the point $P_0(\theta, \phi)$ in Fig. 1(b), the spin state of the initiated exciton can be determined by measuring the phase (thus ϕ) and the amplitude (thus θ) of the biexciton signal.

The sample used in this Letter was grown by molecular-beam epitaxy on a (001)-oriented GaAs substrate. One layer of strain-induced InGaAs QDs was deposited in the center of a one wavelength microcavity [12,13]. For the measurements the sample was placed inside a metal tube immersed in liquid helium, maintaining temperature of 4.2 K. A $\times 60$ objective of 0.85 numerical aperture was used to focus the light on the sample surface and to collect the emitted PL. Two dye lasers, synchronously pumped at a repetition rate of 76 MHz by the same frequency-doubled Nd:YVO_4 (Spectra Physics-Vanguard^{TM}) laser were used for generating the resonantly tuned light pulses. The duration of the pulses were 10 ps and their spectral widths about 100 μeV. They were continuously tuned using coordinated rotations of two plate birefringent filters and an etalon. The polarizations of the pulses were independently adjusted by a polarized beam splitter and two pairs of computer-controlled liquid crystal variable retarders. The polarization of the emitted PL was analyzed by the same setup. The delay between the pulses was controlled by a moving retroreflector. The PL was filtered by a one-meter monochromator, and detected by either a silicon avalanche photodetector or a CCD camera.

Figure 2(a) shows polarization-sensitive PL spectra of a single QD. The neutral exciton (X^0) and biexciton

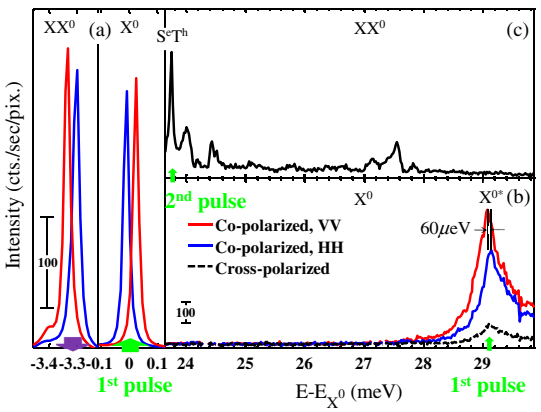

FIG. 2 (color online). (a) V (red) and H (blue) polarized PL spectra from the resonantly excited QD. (b) [(c)] Polarization-sensitive PLE spectra of the exciton [biexciton]. (c) was obtained while resonantly exciting the exciton as indicated by the green vertical arrow in (b). For the readout the second laser was tuned to the biexciton resonance marked by the vertical green arrow in (c), while the PL was monitored from the XX^0 doublet as marked by the downward purple arrow in (a).

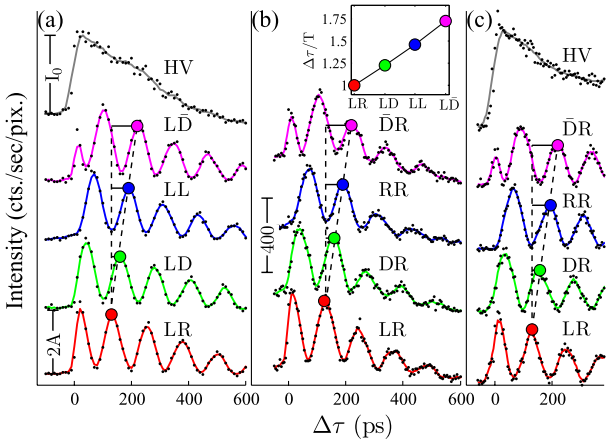

FIG. 3 (color online). (a) Emission intensity of the PL from the biexciton spectral lines as a function of the delay time between the pulse into the excited exciton resonance and the pulse into the biexciton resonance, for various pulse polarizations. The points are measured data and the solid curves are guides to the eye. The first (second) letter describes the polarization of the first (second) pulse. The phase and amplitude of the oscillations are related to the polarization of the first pulse [see Fig. 1(b)]. The phase differences between the various curves are summarized in the inset to (b). (b) Similar to (a) but for various polarizations of the first laser pulse and fixed R second pulse. (c) Similar to (b) but here the first pulse is tuned directly to the ground exciton states.

(XX^0) are each indicated with its cross-linearly polarized doublet. Figure 2(b) shows polarization-sensitive PL excitation (PLE) spectra of the exciton doublet using one laser source. Figure 2(c) shows PLE spectrum of the biexciton doublet using two laser sources. The spectral position of the first resonant laser, which "writes" the exciton spin state, was either tuned to the exciton PL doublet [Fig. 2(a)] or to its broad resonance [Fig. 2(b)] as marked by (green) upward arrows. This strong, linearly polarized resonance, about 29 meV above X^0, originates from the first single hole and second single electron state [Fig. 1(a)]. Its width is due to resonant coupling to the first electronic level via one optical phonon [14]. The lifetime of this excited state as judged by its linewidth (~ 1 meV) is much shorter than its precession period as judged by the energy difference between its H and V colinearly polarized components [60 μeV—Fig. 2(b)]. The relaxation here, resonantly mediated by optical phonons, is much faster than that between the corresponding heavy-hole levels, where acoustic phonons mediate it [15,16]. The initial spin is thus, predominantly preserved in the relaxation.

The PLE spectrum of the biexciton doublet [Fig. 2(c)] was obtained by scanning the frequency of the second laser, while the first laser was tuned into excitonic resonance. The spectral position of the second resonant laser (the readout) is indicated by the (green) vertical arrow on the biexciton PLE spectrum. Full characterization of the two-photon biexciton PLE spectrum will be published elsewhere. The particular resonance used here, is due to a ground state two-electron singlet and a ground and excited state two-heavy holes triplet [S^eT^h—Fig. 1(a)]. This resonance differs from the conventional biexciton transition in which both carrier pairs form singlets in their respective ground levels. Here, the heavy-hole pair form a triplet [15,16]. Therefore, while in the first case the biexciton is excited through the exciton eigenstates by the colinearly polarized photon pair, in the latter, the excitation requires a cross-linearly polarized pair [16]. We use the latter resonance and not the ground biexciton state, to avoid blinding the PL detector.

In Fig. 3 we plot the PL emission intensity from the biexciton doublet as a function of the delay time between the two laser pulses, for various combinations of the two pulses polarizations. The first (second) capital letter denotes the polarization of the first (second) laser pulse. The uppermost black curves in Figs. 3(a) and 3(c) present photogeneration of a biexciton by a cross-linearly polarized second pulse. The biexciton PL signal has the maximum immediately after the first pulse and it decays exponentially as the exciton radiatively recombines. The rest of the curves in Fig. 3(a) show the excitation (writing) of the exciton by L pulse and various polarizations of the second pulse which excites the biexciton ("reads" the exciton spin). In these cases, the first photon polarization lies on the equator of the Poincaré sphere, and it generates a coherent superposition of the two exciton spin eigenstates with equal probabilities. Since these eigenstates are energetically separated by 34 μeV, the exciton spin state precesses in time along the equator of its Bloch sphere with a period $T = h/(34\ \mu\text{eV}) = 122$ ps. The differences between the various curves are only in their relative phases. This dependence is summarized in the inset to Fig. 3(b). The inset to Fig. 3 presents the delay times $\Delta\tau$ on which the second maxima are observed in each one of the four curves in Fig. 3(a), in units of T. One clearly sees that there is a constant phase shift of a quarter of a period between the various polarizations of the second readout pulse. The probability to photogenerate a biexciton by a second pulse depends on the pulse polarization with respect to the exciton spin polarization. Therefore, the second pulse effectively "projects" the exciton spin state onto the complementary direction. This means that a R (L) pulse projects the spin onto the $|L\rangle$ ($|R\rangle$) state and a $\bar{D}$ (D) pulse projects the spin onto the $|D\rangle$ ($|\bar{D}\rangle$) state. Similarly, a H (V) pulse projects the spin onto the $|V\rangle$ ($|H\rangle$) state. Thus, the state of any polarized exciton spin can be determined by the polarization of the initial pulse and a second projective light pulse. Similar behavior is observed when one varies the polarization of the writing pulse while keeping fixed the polarization of the readout pulse, as we show in Figs. 3(b) and 3(c). Figure 3(c) clearly shows that there is no difference in the exciton spin evolution when it is excited into the ground or into the excited state.

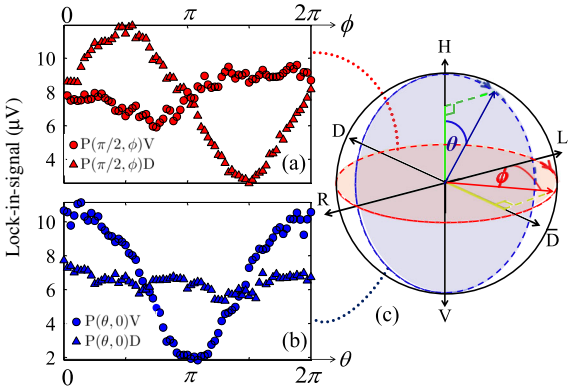

FIG. 4 (color online). (a) [(b)] Biexciton doublet PL intensity (locked-in to the second laser) as a function of the polarization angle ϕ [θ] as defined in (c), for D (triangles) and for V (circles) polarized readout pulses. $\Delta\tau$ between the two pulses was $T = 122$ ps.

This proves that the nonradiative relaxation is mostly spin conserving. Our experimental measurements demonstrate unambiguously that a resonantly tuned H, V, D, $\bar{D}$, R, or L picosecond pulse photogenerates (writes) an exciton with initial spin state $|H\rangle$, $|V\rangle$, $|D\rangle$, $|\bar{D}\rangle$, $|R\rangle$, or $|L\rangle$, respectively.

In Fig. 4 we present a set of measurements in which an arbitrary initial positioning of the exciton spin on its Bloch sphere is demonstrated. The figure describes writing of the exciton spin state by continuous variation of the polarization of the first pulse along a given circle on the Poincaré sphere while leaving the temporal delay between the pulses fixed at $\Delta\tau = T$. The polarization of the readout pulse is left fixed, either in a normal direction to the plane of variation of the first pulse or in that plane. In the first case, one expects the biexciton PL signal to remain constant since the spin projection on the probe direction is constant, independent of the in-plane angle. In the second case, however, the spin projection on the probe direction is expected to undergo maximal periodic oscillations resulting in the largest amplitude of oscillations of the signal of the biexciton. In one set of measurements [Fig. 4(a)] the polarization angle is varied about the V-H axis (ϕ in the L-D plane) and in the other set [Fig. 4(b)] the angle is varied about the D-$\bar{D}$ axis (θ in the H-L plane). The readout in both cases is performed with D pulse (triangles) and with V pulse (circles). The observed oscillations can be described as change in the magnitude of the projection of the Bloch vector along the direction of the polarization of the readout pulse. The points on the equator, the circle that is defined by varying ϕ, while leaving $\theta = \pi/2$ have the same projection (equal to 0) on the V direction. Therefore, almost no oscillations are observed in this case. The projection on the D direction, however, undergoes maximal variations with the angle ϕ. Indeed, large periodic oscillations in the signal are observed. Clear maxima (minima) in the intensity are obtained when the Bloch vector of the exciton is antiparallel (parallel) to that of the probe. As expected for this particular biexcitonic resonance, maximum absorption is obtained for cross-linear polarizations [16]. In a complementary set of measurements [Fig. 4(b)] the opposite behavior is observed. Here the angle θ is continuously varied, while $\phi = 0$. Now, maximal oscillations occur for the V readout pulse and diminishing oscillations for the D readout pulse. The small oscillations observed in the latter are probably due to small inaccuracies in the calibration of the liquid crystal variable retarders and in the alignment of the setup axes relative to those of the QD. Figure 4 demonstrates that the spin state of the exciton can be prepared at any point on the Bloch Sphere [Fig. 4(c)], in correspondence to the elliptic polarization of the writing light pulse on its Poincaré sphere.

In summary, we establish clear correspondence between the polarization of a light pulse tuned to excited or ground excitonic resonances, and the initial spin state of the photogenerated exciton. We directly map the polarization of the light pulse, as represented by a point on the Poincaré sphere, to the exciton spin state as represented by a point on the Bloch sphere. For this we use a second, delayed polarized pulse tuned to a particular electronic-singlet, biexcitonic resonance. The second pulse projects the excitonic spin state onto a predetermined direction, providing thus a way for reading the exciton spin.

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