Coherent optical writing and reading of the exciton spin state in single quantum dots

Y. Benny,[1] S. Khatsevich,[1] Y. Kodriano,[1] E. Poem,[1] R. Presman,[1] D. Galushko,[1] P. M. Petroff,[2] and D. Gershoni[1,*]

[1] *The Physics Department, Technion - Israel institute of technology, Haifa, 32000, Israel.*
[2] *Materials Department, University of California Santa Barbara, CA, 93106, USA.*

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We demonstrate a one to one correspondence between the polarization state of a light pulse tuned to excitonic 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 atomic-like 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 work we demonstrate for the first time that the spin state of a QD confined exciton can be initialized ("written") in any coherent superposition of its eigenstates by a 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.

In Fig. 1(a) we use an energy level diagram to schematically describe the processes involved in writing and reading the excitonic spin state. The first polarized laser pulse photogenerates an excited exciton. We resonantly tune the laser into an excited exciton state so as to prevent scattered laser light from blinding the detectors.

In order to relate between 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 (1/2) is oriented downward (upward) while the heavy-hole spin (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 $|\mathrm{R}\rangle = \Uparrow\downarrow$ ($|\mathrm{L}\rangle = \Downarrow\uparrow$). 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 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 $\bar{\mathrm{D}}$, respectively) and the spin state of the photogenerated pair are given by: $|\mathrm{H}\rangle = 1/\sqrt{2}(\Uparrow\downarrow + \Downarrow\uparrow)$; $|\mathrm{V}\rangle = -i/\sqrt{2}(\Uparrow\downarrow - \Downarrow\uparrow)$; $|\mathrm{D}\rangle = \mathrm{e}^{-i\pi/4}/\sqrt{2}(\Uparrow\downarrow + i\Downarrow\uparrow)$ and $|\bar{\mathrm{D}}\rangle = \mathrm{e}^{i\pi/4}/\sqrt{2}(\Uparrow\downarrow - i\Downarrow\uparrow)$. The direction in space of the H polarization coincides with the direction of the natural major axis of the QD [3]. These 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 the L direction, and a component perpendicular to that plane, parallel to the rectilinear H-V axis. Thus, two angles define an arbitrary polarization: the angle ϕ and the angle θ, which defines the azimuth between the polarization and the H direction ("north pole") of the Poincaré sphere. 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, $|\mathrm{H}\rangle$ and $|\mathrm{V}\rangle$, respectively.

The photogenerated exciton relaxes nonradiatively into its ground state by phonon emission, while maintaining its spin state [11, 12]. A resonantly tuned H (V) polarized laser pulse photogenerates an exciton in its symmetric (antisymmetric) spin eigenstate $|H\rangle$ ($|V\rangle$). The exciton then remains in its eigenstate until it radiatively recombines. Since, however, the two eigenstates are not degenerate [3], they evolve in 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 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 pulse excitation. Such a pulse initiates the two spin eigenstates with equal probabilities. The initiated spin then precesses with time counter clockwise. This precession is described by the angle ϕ which linearly increases with time. After quarter of a period ($\phi = \frac{\pi}{2}$) the spin reaches the $|\bar{\mathrm{D}}\rangle$ state. After half a period ($\phi = \pi$) it reaches the $|\mathrm{R}\rangle$ state, and at three quarters of a period ($\phi = \frac{3\pi}{2}$) it reaches the $|\mathrm{D}\rangle$ state. It completes the cycle after one period T by returning to the $|\mathrm{L}\rangle$ state. The purple circle on the sphere describes precession of an exciton initially photogenerated with spin state $P_0(\theta, \phi)$ by arbitrarily polarized light pulse.

After the first pulse, a second, polarized pulse delayed by $\Delta\tau$ from the first one is applied. The second pulse is tuned to an excited resonance of the biexciton, and is used to read the evolving spin state of the exciton. The absorption of the second pulse and the probability to photogenerate a biexciton depends on the orientation of the exciton spin relative to the polarization of the second pulse. The reason for this dependence is the fact that the absorption into a biexcitonic resonance depends on the relative spin orientation between the pairs of the carriers. Hence, by monitoring the intensity of the photoluminescence (PL) from the biexciton spectral line as a function of the delay between the two pulses, one obtains direct information on the evolving exciton spin state. As schematically described by the point P_0 in Fig. 1(b), the spin state of the initiated exciton can be determined by measuring the phase (thereby finding ϕ) and the amplitude (thereby finding θ) of the biexciton signal.

The sample used in this work 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 [13, 14]. For the optical measurements the sample was placed inside a sealed metal tube immersed in liquid Helium, maintaining temperature of 4.2K. A $\times$60 microscope objective with numerical aperture of 0.85 was placed above the sample and used to focus the light beams on the sample surface and to collect the emitted PL. Two dye lasers, synchronously pumped by the same frequency-doubled Nd:YVO_4 (Spectra Physics- Vanguard$^{\mathrm{TM}}$) laser were used for generating the resonantly tuned optical pulses. The repetition rate of the setup was 76 MHz corresponding to a pulse separation of about 13 ns. The duration of the pulses were about 10 ps and their spectral widths about 100 μeV. They could have been continuously scanned using coordinated rotations of two plate birefringent filters and an etalon. The polarizations of the pulses were independently adjusted by a polarized beam splitter (PBS) and two pairs of computer controlled liquid crystal variable retarders (LCVRs). The polarization of the emitted PL was analyzed by the same LCVRs and PBS. The delay between the pulses was controlled by a retroreflector on a translation stage. The PL, was filtered by a 1-meter monochromator, and detected by either a silicon avalanche photodetector or a CCD camera.

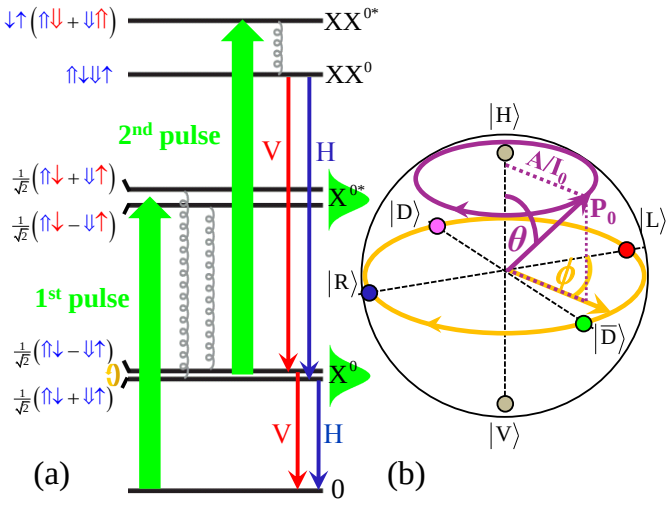

FIG. 1: (a) Schematic description of the writing and readout processes. Horizontal lines describe the states' energies. The spin wavefunctions associated with these states are depicted to the left. $\uparrow$ ($\Downarrow$) represents spin up (down) electron (hole). Blue (red) arrow represents carrier in the first (excited) energy level. Green arrows denote resonantly tuned light pulses, 1$^{\mathrm{st}}$ to an exciton and 2$^{\mathrm{nd}}$ to a biexciton excited state. Curled lines denote nonradiative relaxation and a blue (red) line radiative H (V) polarized recombination. The spectral width of the laser pulses are represented by the green curves to the right. (b) A Bloch sphere representation of the exciton spin states associated with the polarization of the initiating pulse. The point $\mathrm{P}_0(\theta, \phi)$ represents an arbitrarily polarized spin state. A and I_0, relate to the biexciton PL intensity, (see text).

Fig. 2(a) shows polarization-sensitive PL spectra of a single QD. The neutral exciton (X^0) and biexciton (XX^0) are each indicated with its cross linearly polarized doublet. Fig. 2(b) shows a polarization sensitive PL excitation (PLE) spectrum of the exciton line using one laser source and a PLE spectrum of the biexciton line using two laser sources. The spectral position of the first resonant laser, which "writes" the exciton spin state, is marked on the PLE spectrum of the exciton by a green vertical arrow. This spectrally broad resonance, 29 meV above the exciton line, is due to phonon assisted absorption [15]. It shows high degree of linear polarization memory as judged by the difference between the co- and cross-linearly polarized PLE spectra. Such memory is indicative of spin preserving relaxation [11, 12].

The PLE spectrum of the biexciton line was obtained by scanning the frequency of the second laser, while the first laser is resonantly tuned into the excitonic resonance. The spectral position of the second resonant laser

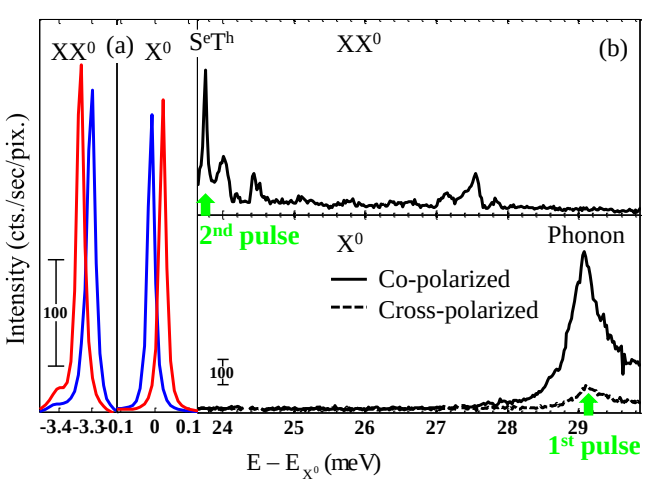

FIG. 2: (a) Linear vertical (red) and horizontal (blue) polarized PL from a resonantly excited single QD. (b) Polarization sensitive PLE spectra of the exciton (bottom) and that of the biexciton (up). The latter was obtained while resonantly exciting the exciton as indicated by the first green vertical arrow. For readout the second laser was tuned to the biexciton resonance marked by the second vertical green arrow.

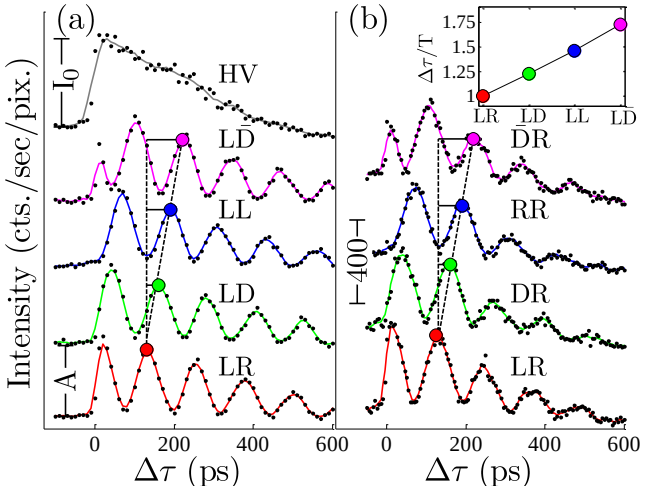

FIG. 3: (a) Emission intensity of the PL from the biexciton spectral line (XX^0) as a function of the delay time between the pulse into the exciton resonance and the pulse into the biexciton resonance for various pulse polarizations. The points are measured data and the solid curves are guides to the 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(a)]. 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.

(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 [11, 12]. Therefore, while in the first case the biexciton is best excited through the exciton eigenstates by co-linearly polarized photons, in the latter, the excitation requires a cross-linearly polarized pair [12].

In Fig. 3 we plot the PL emission intensity from the biexciton spectral line, XX^0, 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 upper most black curve in Fig. 3(a) presents photogeneration of a biexciton by a cross linearly polarized second pulse. The biexciton PL signal has 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 phogenerates 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 of 121.6 ps ($T = h/34\ \mu eV$). The differences between the various curves are only in their relative phases. This dependence is summarized in the inset to Fig. 3(b). The delay times $\Delta\tau$ on which the second maxima are observed in each one of the four curves in Fig. 3(a) is plotted in units of T. One can clearly see that there is a constant phase shift of a quarter of a period between the various polarizations of the second "readout" pulse. When a R pulse is used for the readout, the second maximum occurs exactly one period after the excitation. When a D pulse is used the second maximum lags by quarter of a period. Readout using a L pulse leads to phase shift of half a period, and finally readout with a $\bar{D}$ pulse results in phase shift of three quarters of a period.

In each spin state, the probability to photogenerate a biexciton by a second resonantly tuned light pulse depends on the polarization of the light pulse with respect to the exciton spin polarization. Effectively, the second pulse "projects" the exciton spin state onto the complementary spin 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 ob-

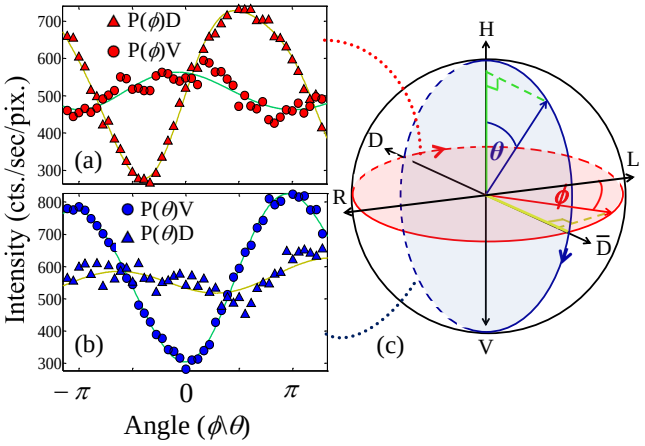

FIG. 4: (a) [(b)] Emission intensity of the PL from the biexciton (XX^0) line as a function of the polarization angle ϕ (θ) as defined in (c), for D (triangles) and for V (circles) readout pulses. Solid lines are guides to the eye. The delay time between the two pulses was T=121.6 ps.

served when one varies the polarization of the writing pulse while keeping fixed the polarization of the readout pulse, as we show in Fig. 3(b). Our experimental measurements demonstrate unambiguously that a resonantly tuned H, V, D, $\bar{\text{D}}$, R or L picosecond pulse photogenerates (writes) an exciton with initial spin state $|\text{H}\rangle$, $|\text{V}\rangle$, $|\text{D}\rangle$, $|\bar{\text{D}}\rangle$, $|\text{R}\rangle$, or $|\text{L}\rangle$, respectively.

In Fig. 4 we present yet another set of measurements in which we demonstrate arbitrary positioning of the exciton spin on its Bloch sphere by a different method. 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 $\Delta\tau = \text{T}$. 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 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-$\bar{\text{D}}$ axis (θ in the H-R plane). The readout in both cases is performed with a D pulse (triangles) and with a 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 ϕ, 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 [12]. In a complementary set of measurement which we present in Fig. 4(b) the opposite behavior is observed. Here the angle θ is continuously varied. 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 LCVRs and in the alignment of the setup axes relative to those of the QD. Fig. 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 an excitonic resonance and the initial spin state of the photogenerated exciton. We directly map the polarization of the light pulse, as represented by arbitrary point on the Poincaré sphere, to exciton spin state as represented by a point on the Bloch sphere. For this we use a second, delayed polarized optical pulse tuned to particular biexcitonic resonances. 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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* Electronic address: dg@physics.technion.ac.il

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