Exciton-Spin Memory with a Semiconductor Quantum Dot Molecule

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We report on a single photon and spin storage device based on a semiconductor quantum dot molecule.
Optically excited single electron-hole pairs are trapped within the molecule, and their recombination rate
is electrically controlled over 3 orders of magnitude. Single photons are stored up to 1 μs and read out on
a subnanosecond time scale. By using resonant excitation, the circular polarization of individual photons
is transferred into the spin state of electron-hole pairs with a fidelity above 80%, which does not degrade
for storage times up to the 12.5 ns repetition period of the experiment.

Further development of the emerging fields of quantum computing and quantum communication require quantum interfaces between optical "flying" and solid-state "stationary" qubits that enable reversible transfer, storage, and manipulation of quantum information [1]. Semiconductors quantum dots (QDs) constitute promising candidates as quantum interfaces between light and matter. Indeed, optical absorption of photons to create an electron-hole pair (or exciton) provides a natural mechanism to transfer quantum information from optical fields into the solid state. Moreover, optical [2], electrical [3], and optoelectrical [4,5] coherent control of an exciton qubit in a QD have been successfully demonstrated. However, those approaches do not permit storage of quantum information, which limits their usefulness to the short lifetime of the exciton.

QD-based devices offering spin storage have been developed [6–8] but rely on the storage of single carriers. Therefore, only pure spin states corresponding to the spin eigenstates can be stored. Indeed, for any other exciton-spin state, the electron and the hole spins are entangled, and loss of one of the carriers makes it impossible to recreate the original photon state, which prevents any extension of those schemes to a quantum memory.

Exciton storage has been demonstrated in ensembles of quantum dot molecules (QDMs) [9], quantum posts [10], nanocrystals [11], natural QDs [12], and coupled quantum wells [13–15]. In those works the carriers were spatially separated for the storage operation. However, those works do not address the issue of transcribing and storing the polarization of a photon into the spin state of the spatially separated electron-hole pair, a necessary step towards a quantum memory for polarized light.

In this Letter, we present a novel approach which allows for initialization and storage of the exciton-spin state in a semiconductor QDM. We first demonstrate microsecond light storage at the single photon level. This is achieved by controlling the recombination rate of the electron-hole pair

through electric-field-dependent spatial separation in the QDM. We then extend our scheme to exciton-spin storage. Using a vertical magnetic field to lift the degeneracy of the exciton-spin states, we show high-fidelity transfer and storage of the circular polarization of photons into the exciton-spin state under resonant optical excitation, with no observable degradation of the fidelity of the operation with time.

The device used consists of a *p-i-n* heterostructure grown by molecular beam epitaxy. Two layers of InAs QDs separated by a thin GaAs layer were grown at the center of the intrinsic region made of a GaAs quantum well clad with a short period superlattice equivalent to $Al_{0.75}Ga_{0.25}As$ on each side, which prevents tunneling of the carriers out of the QDM region when a large vertical electric field is applied. Vertical alignment of the QDs to form QDMs naturally arises from the strain field generated by the growth of the first layer. Doping extends into the superlattice and allows application of an electric field along the growth direction. This *p-i-n* device is encased in a weak planar microcavity to enhance collection efficiency. We used standard photolithography and wet etching techniques to fabricate a diode with an area of $35 \times 60~\mu m^2$. An opaque metallic film with micron-sized apertures allowed single QDMs to be addressed optically. All our experiments were performed on samples cooled to <10 K in a liquid helium cryostat. A diode laser triggered by a pulse pattern generator of tunable period was used for above-band excitation. Resonant and quasiresonant excitation was achieved by using a wavelength-tunable Ti: sapphire pulsed laser of linewidth <30 μeV operating at a fixed period of 12.5 ns.

A schematic band diagram of the device is shown in Fig. 1(a). QDs from the top layer are strain-induced and therefore larger than QDs from the bottom layer. The electron (hole) energy states are consequently lower (higher) in the top QD than in the bottom QD. The exciton naturally relaxes into the top QD at zero electric field as it

is the lowest energy configuration ("direct state"). The top QD is positioned on the *p*-doped side of the diode structure so that applying an electric field in reverse bias separates the hole energy levels of the top and bottom QDs further. On the other hand, the electron energy levels from both QDs are tuned into resonance at a field F_0 and the electron wave function hybridizes over both QDs. Further increase of the electric field leads to further polarization of the electron into the bottom QD, the exciton being eventually delocalized over both QDs forming the molecule ("indirect state").

The energy of the exciton state as a function of the electric field is accurately modeled by using the direct and indirect states as basis states, which leads to the Hamiltonian [16]

$$\mathbf{H} = E_0 + \begin{pmatrix} 0 & t \\ t & ed(F - F_0) \end{pmatrix}, \qquad (1)$$

where E_0 is the energy of the exciton confined in the top QD at zero electric field, F is the electric field applied to the structure, t is the tunneling rate of the electron from the top to the bottom QD, e is the charge of the electron, d is the distance between the QDs, and F_0 is the field at which electron energy levels are tuned into resonance. The eigenstates of this Hamiltonian are the symmetric (low-energy) and antisymmetric (high-energy) combinations of the basis states [17]. Calculated photoluminescence (PL) spectra of the emission from both eigenstates as a function of the electric field is shown in Fig. 1(b), where the dotted lines correspond to the energy of the direct and indirect states. Coherent tunneling of the electron results in an anticrossing between the eigenstates. The dipole moment ed of the

exciton in its indirect state results in a linear Stark shift of the emission energy.

Photon and exciton-spin storage exploits the long lifetime of the exciton indirect state originating from the electron-hole spatial separation. Figure 1(c) shows a schematic of the storage operation. At $t = 0$, a laser pulse creates an electron-hole pair which relaxes into the QDM. The electric field is such that the exciton is in an indirect state characterized by a long lifetime which prevents recombination of the electron and the hole during a storage time Δt. At $t = \Delta t$, a subnanosecond modulation of the electric field is applied to bring the exciton from the indirect to the direct state so that readout of the emission occurs naturally through recombination.

PL emission from the symmetric exciton state of the QDM used for photon storage is shown in Fig. 2(a) as a function of the electric field. Fitting using Eq. (1) gives values of $d = 8.2 \pm 0.05$ nm and $t = 7.9 \pm 0.05$ meV. The high-energy antisymmetric states of the QDMs that we studied were either not visible [as in Fig. 2(a)] or of weak intensity and therefore not suitable for storage experiments. We attribute this to effective thermalization of the exciton to the low-energy symmetric state. Missing spectral lines from QDMs due to thermalization effects have also been reported in other studies [18]. The exciton lifetime as a function of the electric field measured by using standard time-resolved spectroscopy ("direct measurement") is shown in Fig. 2(b). A rapid increase in lifetime is observed as the electron wave function progressively polarizes from the top to the bottom QD.

Using the method described in Fig. 1(c), we successfully stored the exciton for storage times up to 1 μs, corresponding to more than 1000 times the usual lifetime of an exciton in a single QD. The results are shown in Fig. 2(c) for storage times of 0.2, 0.4, 0.6, 0.8, and 1 μs. The intensity
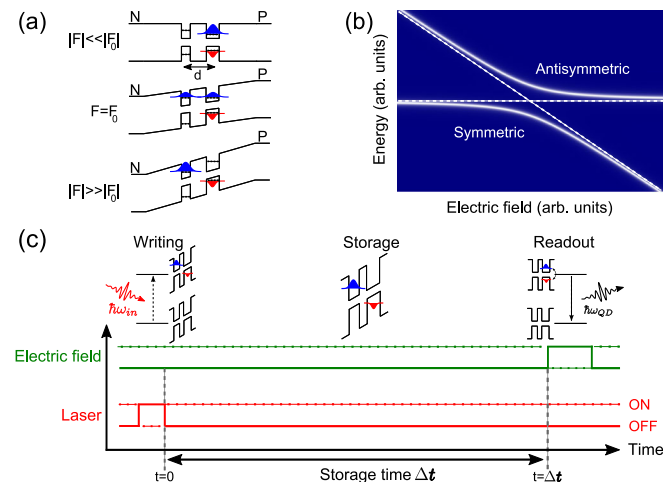

FIG. 1 (color online). (a) Schematic of the band structure of the QDM at various electric fields. The electron and hole wave functions for the low-energy eigenstate are illustrated, respectively, in blue and red. (b) Calculated PL spectra of the emission from the QDM eigenstates as a function of the electric field. The dotted lines correspond to the energies of the direct and indirect states as a function of the electric field. (c) The operating principle behind photon and exciton-spin storage.
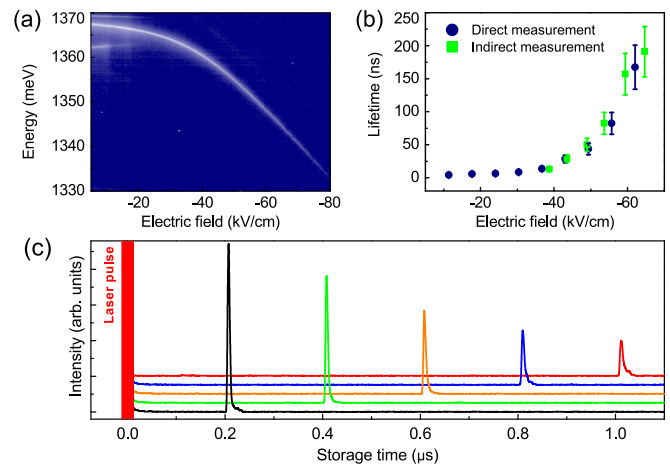

FIG. 2 (color online). (a) Electric-field dependence of the PL emission from the symmetric exciton state used for photon storage. (b) Exciton lifetime as a function of the electric field. (c) Readout signal after storage in an indirect state, at an electric field of -77 kV/cm, for storage times up to 1 μs.

I of the readout emission decays with storage time Δt due to the finite lifetime τ_s of the indirect state. This decay is given by $I(\Delta t) = I_0 e^{-(\Delta t)/(\tau_s)}$, where I_0 is the intensity measured at $t = 0$. The lifetime of the storage state can therefore be measured indirectly by fitting the decay to an exponential. This was done at different electric fields, and the corresponding measurements are shown in Fig. 2(b) and compared to the measurements obtained by standard time-resolved spectroscopy. Agreement between both methods confirms the storage of the exciton in its indirect state. The maximum storage time was limited only by the pulse pattern generator used to trigger the laser pulse, which had a maximum operating period of 1.1 μs. Storage times of several seconds can be expected by using our scheme and engineering QDMs so that longer lifetimes can be achieved [9].

To store exciton spins in a single QDM by using our operating scheme, both resonant optical excitation and a vertical magnetic field are required. Resonant excitation will allow for high-fidelity transfer of the polarization of a photon into the exciton-spin state [3], while a vertical magnetic field will lift the degeneracy of the exciton-spin states and define two Zeeman eigenstates with circular polarizations and long spin lifetimes [6,7].

In Fig. 3(a), PL emission from the QDM used for exciton-spin storage is shown as a function of the electric field. Fitting using Eq. (1) provides values of $d = 8.15 \pm 0.05$ nm and $t = 6 \pm 0.5$ meV. We also performed PL excitation spectroscopy at various electric fields, and two clear resonances were found. Their energies are plotted as a function of the electric field in Fig. 3(a). The first resonance was found at 36.7 ± 0.6 meV from the exciton energy. A similar resonance at the same energy was found as well for other QDMs, indicating resonant excitation

through the creation of a 1-LO phonon in GaAs, the expected energy of which is ≈ 36.4 meV [19]. The second resonance appears at 55.2 ± 2.1 meV and is attributed to the p shell of the exciton state. Consistent with this assignment, we found the position of this resonance varied from QDM to QDM. The lifetime as a function of the electric field was measured by using standard time-resolved spectroscopy, and results are shown in Fig. 3(b). The lifetime levels out around 1 μs for this particular QDM, which is related to the minimum overlap of the electron-hole wave functions reached in the indirect state.

The behavior of our QDM under a vertical magnetic field was also studied. The energy of the exciton-spin states as a function of magnetic field B [20] is given by $E(B) = E_0 + \gamma_1 B + \gamma_2 B^2 + \cdots$, where $|\gamma_1| = g\mu_B/2$ provides a direct measurement of the exciton g factor g (μ_B is the Bohr magneton, and the sign of γ_1 is given by the angular momentum ± 1 of the spin states) and γ_2 is the diamagnetic coefficient and where higher order terms can be neglected for the fields at which we are working ($B < 4T$). Measurements of the exciton g factor and the diamagnetic coefficient γ_2 as a function of the electric field are shown in Figs. 3(c) and 3(d). No electric-field dependence of the g factor is found for electron tunneling, as electron g factors are similar in bulk GaAs (-0.44) and InAs QDs (-0.6) [21]. However, a strong dependence of the diamagnetic coefficient as a function of the electric field is found, and an order of magnitude difference is measured between the direct and indirect states. The diamagnetic coefficient is a measure of the in-plane electron-hole separation, which in turns depends on the lateral confinement and Coulomb interaction [20]. When the electron tunnels from the top to the bottom QD, the lateral confinement does not change significantly since both QDs have similar sizes. The increase in γ_2 in the indirect state therefore provides a measure of the reduced Coulomb interaction. γ_2 saturates at high electric field in the indirect state, where the electron-hole pair is fully spatially separated.

Circular spin eigenstates are created by applying a vertical magnetic field such that $g\mu_B B \gg s$, where s is the fine structure splitting of the exciton eigenstates. For our QDM, s in the direct state at zero electric field is 76.5 ± 0.1 μeV, and circular eigenstates are therefore created for $B > 2T$. Exciting resonantly into the QDM while applying a vertical magnetic field allows for high-fidelity transfer of the photon circular polarization into the exciton-spin state. The fidelity of this transfer is defined as the degree of polarization of the exciton emission $F = (I_{\text{co}} - I_{\text{cross}})/(I_{\text{co}} + I_{\text{cross}})$, where I_{co} (I_{cross}) is the intensity from the Zeeman exciton state co(cross)-polarized with the excitation polarization.

Figure 4(a) shows the fidelity as a function of storage time for left- and right-hand circular pump polarizations and for three excitation schemes: p-shell, phonon, and s-shell. Discrimination between the laser signal and the exciton emission when exciting into the s shell was achieved by taking advantage of the large Stark shift
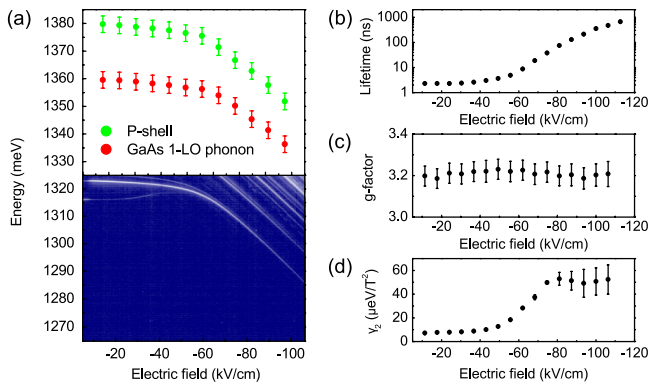

FIG. 3 (color online). (a) PL spectrum of the emission of the exciton used for exciton-spin storage as a function of the electric field along with the energies of the two resonances observed. (b) Exciton lifetime as a function of the electric field. (c),(d) g factor and diamagnetic coefficient as a function of the electric field. Data presented in (a) and (b) were taken at $B = 0T$. Each data point presented in (c) and (d) was obtained from measurements of the energy dependence of the Zeeman states with a magnetic field in the range $0T < B < 4T$.
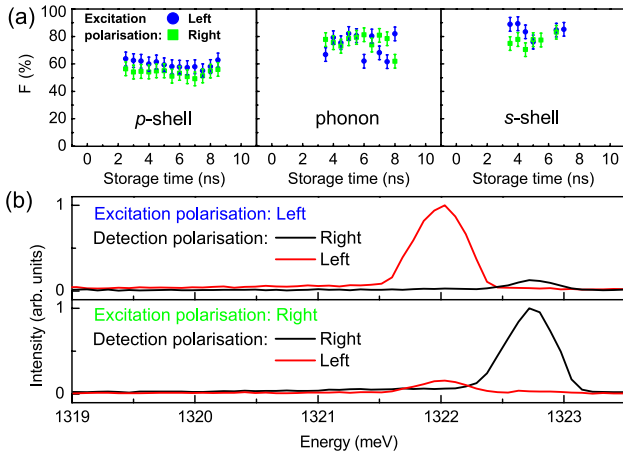
FIG. 4 (color online).   (a) Fidelity of the exciton-spin memory operation as a function of storage time for different excitation schemes and polarizations. The data were taken at $B = 4T$ (similar results were obtained for $2T < B < 4T$). (b) Typical spectra obtained after a storage time of 5 ns when exciting in the phonon resonance and at $B = 4T$.

between the energy of the indirect "write" state and the direct "readout" state. The storage time was limited only by the 12.5 ns period of the laser used for the experiment. Moreover, ringing in the electrical signal prevented measurements at some storage times, as a change in the electric field shifts the exciton energy out of the excitation or detection window through the Stark effect. This effect was particularly important in the case of s-shell excitation, where the resonance is the narrowest.

The fidelity does not degrade over the 12.5 ns repetition period of the experiment, suggesting that the spin relaxation time is substantially longer than the storage time. Average fidelities over time and over both pump polarizations are $57 \pm 5\%$ for p-shell excitation, $75 \pm 7\%$ for phonon excitation, and $81 \pm 6\%$ for s-shell excitation. No spin memory effect was observed when exciting in the continuum states, which we attribute to spin relaxation as the charge carriers thermalize down to the exciton ground state. The fidelity is usually found to be slightly higher when exciting with left-hand circularly polarized light (particularly when using p-shell excitation), which is attributed to thermal relaxation from the right-polarized [high-energy; see Fig. 4(b)] Zeeman eigenstate down to the left-polarized (low-energy) Zeeman eigenstate. Figure 4(b) illustrates the fidelity of the stored emission obtained under phonon excitation. Comparable results were obtained when measuring the fidelity of the write state (without storage operation). As a consequence, the storage operation is found to have no observable effect on the fidelity, and the limitation arises from the write operation, which may be improved by using wave plates calibrated at the QDM energy.

In conclusion, we have successfully stored single photons up to 1 μs and exciton spins with a fidelity above 80% in a single QDM. No decay in the fidelity was apparent

within the period of the excitation laser of 12.5 ns. Increasing the barrier thickness of the QDM, which would lead to longer lifetimes in the indirect state, and using a laser with a tunable period, we anticipate that spin storage in a single QDM is achievable for times in excess of 1 ms. Moreover, it is possible to extend the presented exciton-spin memory to a quantum memory and store a superposition of the spin eigenstates if these can be made to be near degenerate. The energy separation of the eigenstates is limited by the exciton fine structure splitting, and studies of single QDs have shown that it can be reduced to a level where the precession of a superposition state can be recorded with standard time-resolved spectroscopy [22]. This offers new possibilities to study and improve the coherence of an exciton-spin state, which is expected to be limited by hyperfine interaction between the electron spin and the nuclear spins [23]. Eventually, several QDM quantum memories could be combined on a single chip. Recent advances in the field of solid-state cavity quantum electrodynamics make it likely that such a system would be scalable [24].

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 [1] S. Tanzilli *et al.*, Nature (London) **437**, 116 (2005).
 [2] A. J. Ramsay *et al.*, Phys. Rev. Lett. **100**, 197401 (2008).
 [3] A. Boyer de la Giroday *et al.*, Phys. Rev. B **82**, 241301 (2010).
 [4] A. Zrenner *et al.*, Nature (London) **418**, 612 (2002).
 [5] S. Michaelis de Vasconcellos *et al.*, Nat. Photon. **4**, 545 (2010).
 [6] M. Kroutvar *et al.*, Nature (London) **432**, 81 (2004).
 [7] R. J. Young *et al.*, New J. Phys. **9**, 365 (2007).
 [8] D. Heiss *et al.*, Phys. Rev. B **77**, 235442 (2008).
 [9] T. Lundstrom *et al.*, Science **286**, 2312 (1999).
[10] H. J. Krenner *et al.*, Nano Lett. **8**, 1750 (2008).
[11] A. A. High *et al.*, Nano Lett. **9**, 2094 (2009).
[12] R. M. Kraus *et al.*, Phys. Rev. Lett. **98**, 017401 (2007).
[13] C. Rocke *et al.*, Phys. Rev. Lett. **78**, 4099 (1997).
[14] S. Zimmermann *et al.*, Science **283**, 1292 (1999).
[15] A. G. Winbow *et al.*, Nano Lett. **7**, 1349 (2007).
[16] E. A. Stinaff *et al.*, Science **311**, 636 (2006).
[17] M. F. Doty *et al.*, Phys. Rev. Lett. **102**, 047401 (2009).
[18] M. Scheibner *et al.*, Phys. Rev. Lett. **99**, 197402 (2007).
[19] D. Strauch and B. Dorner, J. Phys. Condens. Matter **2**, 1457 (1990).
[20] S. N. Walck and T. L. Reinecke, Phys. Rev. B **57**, 9088 (1998).
[21] M. F. Doty *et al.*, Phys. Rev. Lett. **97**, 197202 (2006).
[22] R. M. Stevenson *et al.*, Phys. Rev. Lett. **101**, 170501 (2008).
[23] R. Hanson *et al.*, Rev. Mod. Phys. **79**, 1217 (2007).
[24] J. L. O'Brien *et al.*, Nat. Photon. **3**, 687 (2009).