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Gigahertz bandwidth electrical control over a dark exciton-based memory bit in a single quantum dot

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An optical write-store-read process is demonstrated in a single InGaAs quantum dot within a charge-tunable device. A single dark exciton is created by nongeminate optical excitation allowing a dark exciton-based memory bit to be stored for over ~1 μs. Read-out is performed with a gigahertz bandwidth electrical pulse, forcing an electron spin-flip followed by recombination as a bright neutral exciton, or by charging with an additional electron followed by a recombination as a negative trion. These processes have been used to determine accurately the dark exciton spin-flip lifetime as it varies with static electric field. © 2009 American Institute of Physics.

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Spin is a natural physical quantity for storing and manipulating information in the solid state as a localized spin couples only weakly to the phonons.1,2 In semiconductor quantum dots, very large spin relaxation times have been demonstrated.3,4 Spin read-out has been achieved with a spin-to-charge conversion scheme.5 However, in the case of self-assembled quantum dots, the spin can be created and read-out by exploiting the optical selection rules.6–8 The selection rules provide a robust link between exciton spin and photon polarization. This is an attractive feature for spin manipulation as radiative recombination is typically a much faster process than spin relaxation. In addition, the use of semiconductor materials allows sophisticated optoelectronic devices to be used. However, spin read-out remains a challenging problem. We present here a proof-of-principle experiment showing how a dark exciton can be converted into a bright exciton within its lifetime, yielding a single photon.

At the heart of our experiment is a device which allows gigahertz control over the electron charge trapped in a single quantum dot. Our wafer structure gives us absolute control over the number of electrons in the dot using an applied bias,11 and a single hole is provided by laser excitation allowing Coulomb blockade at He temperatures. Two devices were constructed using low dot density wafer material (<10 dots μm−2). The premise of the design (Fig. 1) was to produce high optical collection efficiency devices with low resistance and capacitance properties such that the dots respond to an applied voltage on a nanosecond timescale. This was achieved by paying attention to four factors. First, the “parallel plate” capacitance formed by the Schottky gate and the back contact was miniaturized by reducing the Schottky gate area to less than 700 μm2. Second, stray capacitances were minimized by removing unneeded areas of the back contact with a ~400 nm deep wet etch. Third, low resistance macroscopic contacts connect the device to a high speed coaxial cable using low resistance silver conductive paint; a 360 nm thick layer of NiCr connects the Schottky gate to the coax inner, and annealed layers of AuGe/Ni/AuGe (60/10/60 nm, respectively) form an Ohmic contact between the back contact and the coax outer. Lastly, the connections to the coax were placed more than 1 mm away from the Schottky gate, the 400 nm deep wet etch, the Ohmic contact, and the metallic top surface (a 5 nm thin semitransparent NiCr Schottky gate).

The dots were grown with a lateral density gradient on top of a 25 nm layer of GaAs (tunneling barrier), beneath which there is a heavily n-doped layer of GaAs (back contact, n=4×1018 cm−3). A 10 nm GaAs capping layer is deposited on top of the dots, followed by a superlattice consisting of 24 AlAs/GaAs pairs to prevent hole tunneling to the metallized back contact (blue).

![FIG. 1. (Color online) Device 1 (a) schematic (not to scale) and (b) top view optical microscope image (with scaling). Shown are the 5 nm semitransparent Schottky gate, the 400 nm deep wet etch, the Ohmic contact, and the 360 nm thick NiCr electrical contact strip. (a) includes a representation of the AlAs/GaAs superlattice (red), the dot layer (black), and the n-doped back contact (blue).](http://scitation.aip.org/termsconditions. Downloaded to IP: 131.152.109.100 On: Fri, 16 May 2014 15:22:14)
away from the optically active area, allowing a superhemi-
spherical solid immersion lens (n=2.15) to be placed cen-
trally over the Schottky gate in order to boost the photolu-
minescence (PL) collection efficiency by a factor of ~5.15 In
device 1 the Ohmic contact is placed 10 μm from the Schottky
gate, minimizing the resistance of the device. In
device 1 the Ohmic contact, etch, and contact metal dimen-
sions form a coplanar waveguide impedance-matched to the
50 Ω high-speed cabling. In device 2, the Ohmic contact is
placed 3 mm from the Schottky gate.

High-speed coaxial cables (<100 ps, 10:90 voltage re-
response time) allow a voltage to propagate from an Agilent
81133A pulse pattern generator (PPG) (60 ps and 10:90 rise
time) to the sample at low temperature with subnanosecond rise time. The PL is spectrally dispersed by a blazed grating
spectrometer. There are two options for detecting the PL.
The first is to record the spectra using a liquid nitrogen-cooled
charge-coupled device camera (~50 μeV spectral resolu-
tion) positioned at one exit aperture of the spectrometer.
The second option utilizes a second exit aperture of the spec-
trometer allowing a small energy range (~0.5 meV band-
width) to be coupled into a multimode fiber which leads to a
Si-based single photon avalanche detector (SPAD) (~400 ps
with full width at half maximum jitter), allowing time corre-
lated single photon counting (TCSPC).

Under 830 nm continuous wave (cw) laser illumination,
varying the gate voltage (Vg) and detecting the PL gives a
clear picture of the dot charging1 [Fig. 2(a)]. At low values
of Vg (e.g., Vg=V1), the conduction energy levels of the dot
are above the Fermi level (defined by the back contact) and
an electron in the dot will tunnel out through the tunneling
barrier within ~10 ps.16 A single hole (h) however can be
confined in the dot in this bias regime. Hole tunneling is
prohibited by the thin capping layer and the blocking barrier.
At Vg<V2<Vg in the presence of a hole, a single electron
tunnels into the dot and the Coulomb blockade prevents fur-
ther electron tunneling, so a neutral exciton (X0) is formed.
In a previous work,13 a nongenerate exciton creation reaction
occurs in 50% probability of creating either bright or dark X0. At
0.35 V<Vg<0.56 V, a second electron can overcome the
Coulomb repulsion from the first electron and tunnel into the
dot, creating a negative trion (X−).

The 10:90 voltage response time of each device was
measured using TCSPC of a single dot X0 PL. Under 830 nm
cw illumination, the PPG was connected to the Schottky gate
and set to output a 10 MHz square wave with a low voltage
of 10 mV below the h→X0 charging point and a high voltage
of 10 mV above the X0→X1 charging point. The dot is only
possible of forming X0 while Vg experienced by the dot is
within the X0 plateau, which only occurs at the changing
edges of the square wave. The time during which PL is re-
corded at each edge of the square wave can then be consid-
ered the Vg rise/fall time experienced by the dot, and repre-
sents a measure of the voltage response time of the device.
Device 1 (2) has a voltage response time of ~1.6 ns
(~4 ns). The larger response time for device 2 relative to
device 1 arises from the larger RC time constant, as expected
from the fabrication.

A single dot in device 2 was excited by 60 ps optical
pulses from an 830 nm laser source [Fig. 2(b)]. TCSPC was
performed on PL from X0 at 1.3133 eV under two separate
applied voltages. The first was a static gate voltage set at Vg.
The second was a time varying voltage as shown in Fig. 2(c).
In the first case [Fig. 2(d), black] a biexponential decay is ob-
erved,13 the fast (1.1 ns) decay arises from recombination
of the bright exciton; the slow, (6.6 ns) decay arises from the
dark exciton which is incapable of radiative recombination
without first experiencing a change in spin from L=2 to L
=1. In this device, the exciton changes its spin via an elec-
tron spin-flip, the electron exchanging its spin with the back
contact (cotunneling) in order to become bright.13 Two sig-
ificant changes are observed when compared with the TC-
SPC for the second case [Fig. 2(d), red]. First, between t
=8 ns and t=16 ns the PL is suppressed. When the voltage
changes to V1 the electron tunnels out of the dot and the PL
is quenched. The reappearance of the PL once the voltage is
restored to V2 shows that the hole is stored throughout.
Second, peaks can be seen at 7 and 17 ns. At both edges of the
X0 plateau are voltage regions in which cotunneling is faster
than the radiative decay. Within these regions tunneling be-
 tween the dot and the back contact randomizes the electron
spin of the dark exciton, allowing radiative recombination as
a bright neutral exciton. As the voltage is changed from V2 to
V1 the dark-bright spin-flip rate increases, causing the peaks.

The peaks at t=7 ns and t=17 ns in Fig. 2(d) suggest that
dark excitons can be deterministically forced to become
bright by using GHz voltage pulses to control the spin-flip
lifetime. We confirm this with a write-store-read process with
the single dark excitons shown in Fig. 3(c). Two static voltage
biases are defined in Fig. 2(a). The first is in the center of
the X0 plateau (V3) and defines the “write” voltage. The sec-
ond bias point is in either of the two cotunneling dominated
regions (V2 or V4) and functions as the “read” voltage. A 60
ps pulse from an 830 nm laser [Fig. 3(a)] nonresonantly cre-
ates an X0 in the dot at time t=0 and Vg=V3 [Fig. 3(b)]. The
TCSPC of X0 [Fig. 3(c)] shows a peak at t=0 which is due to
the fast (~600 ps) radiative recombination of the bright
exciton. A secondary slow decay is observed corresponding
to the long (~800 ns) spin-flip lifetime of the dark exciton at
this voltage. At seven storage values (Δt) the voltage was
changed to V2 or V4 [Fig. 3(b)], and a PL peak was recorded.
Randomization of the electron spin occurs at V2 and V4 caus-
ing any dark excitons to become bright, allowing recombi-

FIG. 2. (Color online) (a) Contour plot showing the PL from a dot in device 1 as a function of the applied bias. Read-out noise on the detector (dot PL > 150 counts) is shown as white (dark blue). The labeled parts are the bias point in which only a hole is stored in the dot (V1), the X0 plateau extent (V2 to V3), and the X1− read-out bias (Vc). The write voltage V1 can be varied over the X0 plateau. (b) A time-resolved measurement of the nonresonant laser excitation, the duration of the pulse being determined by the timing jitter of the single photon detector. (c) An oscilloscope measurement of the voltage pulse applied to the sample. (d) TCSPC of X0 recorded from a dot in device 2 following laser excitation at t=0 (b) while a voltage pulse (c) is applied (red), alongside a second plot using a static voltage bias V2 (black).

nation within $\sim 1$ ns. This method has been successful with measured $\Delta t$ of over 1.5 $\mu$s.

A second write-store-read method again involves creating $X^0$ in the dot nonresonantly with a 60 ps pulse of 830 nm laser light at $t=0$ and $V_g=V_3$ [Fig. 3(a)]. In this case however, the read-out process involves converting a dark $X^0$ into an $X^{1-}$. The dark exciton is stored for seven different $\Delta t$ and the PPG is used to apply a voltage pulse from $V_3$ to $V_2$ [Fig. 3(b)] causing the appearance of an $X^{1-}$ PL peak [Fig. 3(d)]. At $t=\Delta t$ the change in voltage causes a second electron to tunnel into the dot from the back contact, forming an $X^{1-}$. Unlike $X^0$, $X^{1-}$ has no fine structure and is able to recombine radiatively in $\sim 700$ ps, giving a distinct read-out signal.

We can fit an exponential decay [Figs. 3(c) and 3(d), red lines] to the peaks in PL signal versus decay time. The fit gives a value for the electron spin-flip time $\tau$ of a dark exciton at voltage $V_3$. The advantage of the pulsed voltage experiment is that all the signal at $t>\Delta t$ in the dc experiment is bundled into one small time window, enhancing the accuracy with which $\tau$ can be determined. By varying $V_g$ across the bias extent of the neutral exciton we show in Fig. 4 that the $\tau$ values obtained closely follow the secondary lifetime of $X^0$ taken with static voltage bias. The dependence of $\tau$ on $V_3$ can be understood by calculating the cotunneling rate from the Anderson Hamiltonian.\textsuperscript{13} The model is parameterized with the electron tunneling time ($\tau_e$), the $X^0$ energy splitting between dark and bright states ($\delta_{HP}$), the temperature ($T$), and the radiative ($\gamma_r$) and nonradiative ($\gamma_n$) decay rates. The model provides a good fit as shown in Fig. 4. In particular, a nonradiative decay rate is not required ($\gamma_n=0 \text{ ns}^{-1}$) and there is a 50:50 relative intensity between the primary and secondary decay in the center of the $X^0$ extent. This confirms that the $X^0$ secondary lifetime is limited by cotunneling even at the plateau center, in contrast to Ref. 13, where there is a nonradiative decay rate. This is due to the increased capping layer thickness of the wafer in Ref. 13; hole tunneling from the dot into the capping layer provides a nonradiative decay path for the neutral exciton.\textsuperscript{14} In this particular device, dark exciton spin relaxation is always dominated by cotunneling. However, by increasing the tunneling barrier, cotunneling can be suppressed by many orders of magnitude,\textsuperscript{15} and it will be difficult to measure the dark exciton spin relaxation via a decay curve. Instead, spin read-out with a gigahertz bandwidth pulse is much more suitable.

In conclusion we have demonstrated that gigahertz bandwidth voltage pulses can be applied over a charge-tunable single quantum dot device to manipulate the charge of an exciton deterministically within its recombination lifetime. Using this method, single dark excitons have been used as bit memory elements. Optical read-out is carried out by converting the dark $X^0$ into a bright $X^0$ (emission at $X^0$ wavelength) or into an $X^{1-}$ (emission at $X^{1-}$ wavelength). This technique offers great potential for a deterministic single photon source in which individual photons are generated with a gate voltage pulse, and for hole spin read-out.


