

# Supplemental material for “Complete control of a matter qubit using a single picosecond laser pulse”

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## SAMPLE DESCRIPTION

The sample used in this work was grown by molecular-beam epitaxy on a (001) oriented GaAs substrate. One layer of strain-induced  $\text{In}_x\text{Ga}_{1-x}\text{As}$  quantum dots (QDs) was deposited in the center of a one-wavelength microcavity formed by two unequal stacks of alternating quarter-wavelength layers of AlAs and GaAs, respectively. The height and composition of the QDs were controlled by partially covering the InAs QDs with a 3 nm layer of GaAs and subsequent growth interruption. To improve photon collection efficiency, the microcavity was designed to have a cavity mode which matches the QD emission due to ground-state e-h pair recombinations. During the growth of the QD layer the sample was not rotated, resulting in a gradient in the density of the formed QDs. The estimated QD density in the sample areas that were measured is  $10^8\text{cm}^{-2}$ ; however, the density of QDs that emit in resonance with the microcavity mode is more than two orders of magnitude lower [1]. Thus, single QDs separated by few tens of micrometers were easily located by scanning the sample surface during micro-photoluminescence (PL) measurements. Strong antibunching in intensity autocorrelation measurements was then used to verify that the isolated QDs are single ones and that they form single-photon sources.

## EXPERIMENTAL SETUP

The experimental setup that we used for the optical measurements is described in Fig. 1. The sample was placed inside a sealed metal tube immersed in liquid helium, maintaining a temperature of 4.2 K. 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. In the measurements described in Figs. 3-5 of the Letter we used two dye lasers (Styryl 13), synchronously pumped by the same frequency-doubled Nd:YVO<sub>4</sub> (Spectra Physics-Vanguard<sup>TM</sup>) laser for generating the resonantly tuned optical pulses, as described in the figure. The repetition rate of the setup was 76 MHz, corresponding to a pulse separation of about 13 nsec. The lasers' emission energy could have been continuously tuned using coordinated rotations of two plate birefringent fil-

ters and a thin etalon. The temporal width of the dye lasers' pulses was about 9 psec and their spectral width about 100  $\mu\text{eV}$ . A third, Ti:Sapphire pulsed laser (Spectra Physics, Tsunami<sup>TM</sup>) was locked to the clock of the Vanguard<sup>TM</sup> laser. Its pulse duration was about 2 psec and spectral width of about 500  $\mu\text{eV}$ . The delay between the pulses was controlled by 2 retroreflectors on translation stages. The polarizations of the pulses were independently adjusted using 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 PL was spectrally analyzed by a 1-meter monochromator and detected by either a silicon avalanche photodetector (using lock-in detection) or by a cooled charged coupled array detector.

## ROTATION BY A DETUNED PULSE TO A NON-DEGENERATE BIEXCITONIC TRANSITION.

A coherent exciton spin state may be described as a vector on the Bloch sphere:

$$|X(\theta, \phi)\rangle = \cos\left(\frac{\theta}{2}\right)|H\rangle + ie^{i\phi}\sin\left(\frac{\theta}{2}\right)|V\rangle \\ = \alpha(\theta)|H\rangle + \beta(\theta, \phi)|V\rangle. \quad (1)$$

Here  $|H\rangle$  and  $|V\rangle$  are the two exciton's eigenstates  $1/\sqrt{2}[(1e^1)_{-1/2}(1h^1)_{3/2} + (1e^1)_{1/2}(1h^1)_{-3/2}]$  and  $1/\sqrt{2}[(1e^1)_{-1/2}(1h^1)_{3/2} - (1e^1)_{1/2}(1h^1)_{-3/2}]$  respectively. Here the number denotes the energetic order of the confined single carrier level, the letter stands for the carrier type (e-for electron and h for heavy hole), the superscript (either 1 or 2) for the level's occupation and the subscript for the spin projection of same charge carriers [2]. These non-degenerate eigenstates form the poles of the Bloch sphere (Fig. 2 of the Letter).

Such an exciton is photogenerated by a short optical pulse (whose bandwidth is larger than the energy difference between the two eigen-energies) resonantly tuned to an excitonic transition provided that the pulse polarization is given by:

$$\vec{P}_X(\theta, \phi) = \cos\left(\frac{\theta}{2}\right)\hat{H} + ie^{i\phi}\sin\left(\frac{\theta}{2}\right)\hat{V}, \quad (2)$$

where  $\hat{H}$  and  $\hat{V}$  represent linear polarizations parallel to the major and minor axis of the QD, respectively [2, 3].

FIG. 1. Schematic description of the experimental setup. The delay between the three pulses is controlled by 2 computer controlled motorized retroreflectors. (P)BS stands for (polarizing) beam splitter, VBS for a variable beam splitter, BC for beam combiner, and LCVR for a liquid crystal variable retarder.

This exciton is excited by a second pulse which is resonantly tuned to a *non-degenerate* biexciton resonance. Here the resonance is  $(1e^2)(1h^14h^1)_{T_0}$ , in which the two electrons form a spin singlet in the ground state and the holes, one in the ground s-like level and one in the  $d_{HH}$ -like 4<sup>th</sup> level, form a total spin projection zero triplet [2]. Maximal absorption occurs if the polarization of the second pulse is crossed-polarized with the exciton spin (and therefore with the polarization of the first pulse, if both pulses are simultaneous),

$$\vec{P}_{XX}(\pi - \theta, \pi + \phi) = \sin\left(\frac{\theta}{2}\right)\hat{H} - ie^{i\phi}\cos\left(\frac{\theta}{2}\right)\hat{V}, \quad (3)$$

as described in Refs. 2 and 3. The cross-polarized exciton state:

$$\begin{aligned} |\bar{X}(\theta, \phi)\rangle &= |X(\pi - \theta, \pi + \phi)\rangle = \\ &= \sin\left(\frac{\theta}{2}\right)|H\rangle - ie^{i\phi}\cos\left(\frac{\theta}{2}\right)|V\rangle = \\ &= \alpha(\theta)|H\rangle + \beta(\theta, \phi)|V\rangle. \end{aligned} \quad (4)$$

is completely unaffected by resonant pulses with this polarization, but maximally coupled to "cross polarized" pulses with polarization described by Eq. (2).

When a control,  $2\pi$ -pulse with  $\vec{P}_{XX}$  polarization is applied to an arbitrary coherent excitonic state such as  $|X(\theta', \phi')\rangle$

$$\begin{aligned} |X(\theta', \phi')\rangle &= \cos\left(\frac{\theta'}{2}\right)|H\rangle + ie^{i\phi'}\sin\left(\frac{\theta'}{2}\right)|V\rangle = \\ &= \alpha(\theta')|H\rangle + \beta(\theta', \phi')|V\rangle. \end{aligned} \quad (5)$$

which can be conveniently expressed also in terms of the coherent states  $|X(\theta, \phi)\rangle$  and  $|\bar{X}(\theta, \phi)\rangle$  as:

$$|X(\theta', \phi')\rangle = \alpha(\theta^p)|X(\theta, \phi)\rangle + \beta(\theta^p, \phi^p)|\bar{X}(\theta, \phi)\rangle \quad (6)$$

In Eq. (6) spherical symmetry considerations are used and the angles  $\theta^p$  and  $\phi^p$  are measured relative to the pulse's polarization direction.

Since the  $\vec{P}_{XX}(\theta, \phi)$  polarized  $2\pi$  pulse couples only to the  $|\bar{X}(\theta, \phi)\rangle$  part of the exciton wavefunction, this part acquires a geometrical phase of  $\delta$  (defined by Eq. (1) in the Letter) relative to the  $|X(\theta, \phi)\rangle$  part of the exciton. Therefore after the control  $2\pi$ -pulse ends, the new exciton state is given by:

$$\begin{aligned} |X(\theta'', \phi'')\rangle &= \\ &= \alpha(\theta^p)|X(\theta, \phi)\rangle + e^{-i\delta}\beta(\theta^p, \phi^p)|\bar{X}(\theta, \phi)\rangle = \\ &= \alpha(\theta^p)|X(\theta, \phi)\rangle + \beta(\theta^p, \phi^p - \delta)|\bar{X}(\theta, \phi)\rangle. \end{aligned} \quad (7)$$

Thus, the geometrical description of the control pulse action is a clockwise rotation by an angle  $\delta$  about an axis connecting the states  $|X(\theta, \phi)\rangle$  and  $|\bar{X}(\theta, \phi)\rangle$ , parallel to the polarization direction of the control pulse.

### DESCRIPTION OF THE ACTION OF THE POLARIZED $2\pi$ CONTROL PULSE AS A UNIVERSAL ROTATION

A unit vector in the polarization direction of an exciton  $|X(\theta, \phi)\rangle$ , which is coupled to a polarized  $2\pi$  control pulse is given by:

$$\hat{n} = (n_x, n_y, n_z) = (\cos\phi\sin\theta, \sin\phi\sin\theta, \cos\theta), \quad (8)$$

where the Cartesian axes are chosen such that:  $\hat{x} \equiv |R\rangle = 1/\sqrt{2}(|H\rangle + i|V\rangle)$ ,  $\hat{y} \equiv |\bar{D}\rangle = 1/\sqrt{2}(|H\rangle - i|V\rangle)$ ,  $\hat{z} \equiv |H\rangle$ . As discussed above, the effect of the control pulse on the exciton wavefunction can be simply viewed as a rotation about  $\hat{n}$  by the angle  $-\delta$ . Such a rotation in the eigenstates base is described by the operator:

$$R_{\hat{n}}(\delta) = \exp\left(i\vec{\sigma} \cdot \hat{n} \frac{\delta}{2}\right), \quad (9)$$

where  $\vec{\sigma} \equiv (\sigma_x, \sigma_y, \sigma_z)$  is the Pauli matrix vector. When this operator is applied to an exciton state such as  $|X(\theta', \phi')\rangle$  (Eq. (7)), the freedom in choosing  $\theta, \phi$  and  $\delta$  constructs a universal rotation of the exciton spin polarization.

### ESTIMATION OF THE PHASE SHIFT AND VISIBILITY OF THE CONTROL PULSE ACTION.

Each one of the measurements displayed in Figs. 2-3 of the Letter, was fitted to a functional of the form:

$$C \cdot e^{-\frac{t}{\tau}} \left[ 1 - V \cdot \cos\left(\frac{2\pi}{T} \cdot \Delta t + \Delta\phi\right) \right], \quad (10)$$

where  $\tau$ ,  $T$  and  $C$  are the exciton lifetime, its precession period and an overall normalization factor. The parameters  $\tau$  and  $T$  are accurately evaluated independently. The parameters  $V$  and  $\Delta\phi$  are the visibility and the phase shift of the signal, respectively. These are extracted from the fits of Eq. (10) to each one of the traces in the three pulse experiments presented in Figs. 3-5, for  $\Delta t > T$ . Here  $\Delta\phi$  is measured relative to  $\phi = 0$  in the two-pulse experiments (lowest trace in Figs. 3-5). The visibility of the control pulse is then normalized by the visibility

of the two-pulse experiments (without the control pulse), which is slightly less than one [3].

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