Proposal for geometric generation of a biexciton in a quantum dot using a chirped pulse

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We propose to create a biexciton in a quantum dot by a coherent optical process using a frequency-sweeping (chirped) laser pulse. In contrast to the two-photon Rabi flop scheme, the present method uses the adiabatic state transfer through avoided level crossing. As a geometric control, the proposed process is robust against pulse area uncertainty, detuning, and dephasing. The speed of the adiabatic operation is constrained by the biexciton binding energy.

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I. INTRODUCTION

Semiconductor quantum dots (QDs) have manifold uses in quantum information and computation. They have been utilized to generate single photons[1–6] with good indistinguishability.[7] More recently it has been proposed and partially realized[2,8–11] that QDs in two-exciton states, called biexcitons, can be used to generate pairs of entangled photons by two-pathway cascade emission of photons.[12] The entanglement of photon pairs in this scheme was noted[13] to be imperfect because of the slight difference in energy between the two single-exciton levels,[14,15] which tells the which-pathway information of the cascade emission. However, considerable improvement has recently been made,[16,17] which suggests that the scheme should be of high experimental value in quantum optics, quantum computation,[18] and quantum cryptography[19,20] and can also be used to test foundations of quantum mechanics.[2] Biexciton is also of interest in itself because it serves as the physical basis for a two-bit conditional quantum logic gate.[21]

A number of works has already been done on the optical coherent control of the single-exciton states in, e.g., InAs/GaAs QDs (Refs. 22–26) and CdSe/ZnSe QDs.[6] In the recent experiments of optical coherent control of biexciton states, two approaches were used. The first one applies two optical beams each in resonance with the $|g\rangle \rightarrow |X\rangle$ (ground-state-to-single-exciton) and $|X\rangle \rightarrow |XX\rangle$ (single-exciton-to-biexciton) transitions.[21,27] However, it was noted[28] that a better approach is to apply degenerate pulses with frequency equal to half the biexciton energy, such that the spontaneously emitted photons have frequencies different from that of the excitation pulse. This has been followed by recent works.[28–30] Experiments have been done on both InAs/GaAs and CdSe/ZnSe QDs, and the phenomenon of two-photon Rabi oscillation is the prime indicator of successful control in these experiments.

In this paper, we propose to use a frequency-sweeping pulse[31] for a geometric generation of a biexciton in a quantum dot. The scheme is based on the adiabatic state transfer from the ground state to the final biexciton state via avoided energy-level crossing, in which the intermediate exciton is largely bypassed. The utilization of level anticrossing follows the idea of the stimulated Raman adiabatic passage (STIRAP) for adiabatic state transfer in a Λ-type three-level system.[32,33] But here since the exciton and biexciton transitions couple to the same optical pulse, independent control of the two transitions as required in the STIRAP is not feasible. Instead, the frequency sweeping[31] is proposed to realize the adiabatic state evolution, which has been considered for application in quantum information processing.[34,35] The geometric scheme bears the robustness against some uncertainty in the system parameters such as energy levels and dipole magnitude and in laser-pulse parameters such as amplitude, shape, and frequency, which is unavoidable in realistic experiments. Bypassing the intermediate single-exciton state minimizes the possibility of generating single-photon emission which may contaminate an entangled photon pair. Constrained by the biexciton binding energy, the adiabatic state transfer can be completed in picosecond timescales for a typical CdSe quantum dot, and thus the effect of the exciton dephasing can be largely avoided.

This paper is organized as follows: in Sec. II we formulate the problem and explain the basic idea; in Sec. III we demonstrate numerically the creation of a biexciton, which is robust against small uncertainty in all parameters characterizing the system and keeps the occupation of single-exciton state relatively low; and in Sec. IV we show that dephasing, modeled in the Lindblad formalism,[36] only slightly reduces the efficiency.

II. MODEL AND MECHANISM

The biexciton system can be modeled by a four-level system: the ground state $|g\rangle$, the biexciton state $|XX\rangle$, and two intermediate exciton states with different linear polarizations $|X\rangle$ and $|Y\rangle$.[14,15] Because the two pathways of excitation, $|g\rangle \rightarrow |X\rangle \rightarrow |XX\rangle$ and $|g\rangle \rightarrow |Y\rangle \rightarrow |XX\rangle$, are independent and can be implemented independently by applying different polarizations of lasers in experiments, we only consider $|g\rangle \rightarrow |X\rangle \rightarrow |XX\rangle$ for simplicity.

The Hamiltonian is written as

$$H = (\omega + \delta)|X\rangle\langle X| + 2\omega|XX\rangle\langle XX| + [\Omega(t)(|X\rangle\langle g| + |XX\rangle\langle X|) + \text{H.c.}], \quad (1)$$

where ω is half the energy between the ground state and the biexciton, $\omega+\delta$ is the energy of the exciton state (2δ being the biexciton binding energy), and $\Omega(t)=\Omega_t \exp[-i(\omega-\Delta)t + i\phi(t)]$ is the time-dependent optical coupling caused by a laser pulse. We shall consider a chirped pulse with time-

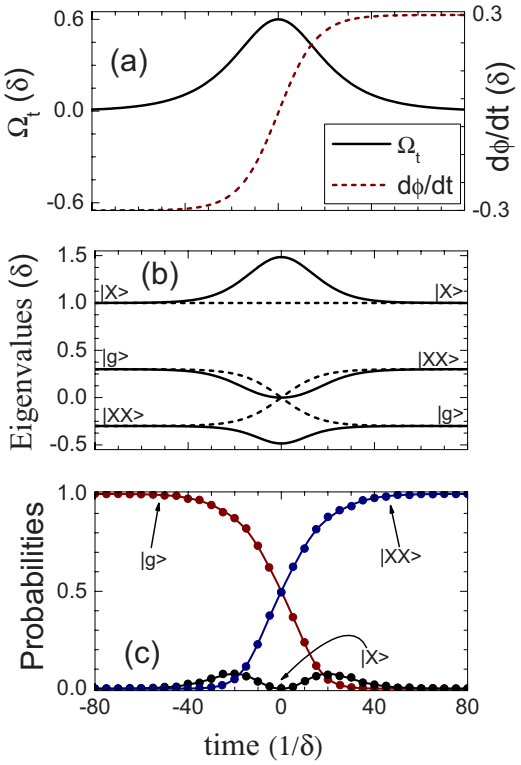

FIG. 1. (Color online) (a) Laser-pulse amplitude and time-dependent frequency as defined in Eq. (3); Ω_t and $\dot{\phi}(t)$ are in units of half the biexciton binding energy δ. (b) Time-dependent eigenvalues of Hamiltonian in Eq. (2) under the pulse in Eq. (3) (solid lines) and when $\Omega_t \to 0$ (dashed lines). The eigenvector corresponding to each eigenvalue at the beginning and the end of the pulse is indicated. (c) Occupations of different basis states during the evolution starting from the ground state $|g\rangle$ calculated by the adiabatic approximation (solid lines) and by the numerical integration of Schrödinger equation (dots). The parameters are such that $A=0.6\delta$, $\alpha=0.06\delta$, $\mu=5$, and $\Delta=0$.

dependent frequency $\omega-\Delta-\dot{\phi}(t)$ [see Fig. 1(a) for the envelope Ω_t and frequency sweeping $\dot{\phi}(t)$ of a chirped pulse]. Here we have assumed that the exciton and the biexciton transitions have the same dipole matrix element, which is a good approximation for quantum dots with strong confinement where the ground-state biexciton consists mostly of the ground-state excitons.[21] Actually, as will be discussed later, the geometrical generation of the biexciton is robust against parameter variation, so a small deviation of the biexciton dipole moment from the exciton one has negligible effect on the efficiency.

To consider the adiabatic state following under the driving of a frequency-sweeping (chirped) pulse, we write the Hamiltonian in a frequency-modulated rotating reference frame as

$$H(t) = \begin{pmatrix} -\Delta - \dot{\phi}(t) & \Omega_t & 0 \\ \Omega_t & \delta & \Omega_t \\ 0 & \Omega_t & \Delta + \dot{\phi}(t) \end{pmatrix}, \tag{2}$$

in the time-dependent basis

$$\{e^{-i\Delta t - i\phi(t)}|g\rangle, e^{-i\omega t}|X\rangle, e^{-i(2\omega-\Delta)t+i\phi(t)}|XX\rangle\}.$$

When Ω_t approaches zero, the eigenvectors of $H(t)$ are the three basis states, with time-dependent eigenvalues $\{-\Delta - \dot{\phi}(t), \delta, \Delta+\dot{\phi}(t)\}$ in the rotating reference frame. We envisage that when $\dot{\phi}(t)$ sweeps from negative to positive (or the opposite), the ground state and the biexciton state would cross at the degenerate point $[\Delta+\dot{\phi}(t)=0]$ [see dashed lines in Fig. 1(b)]. The degeneracy will be lifted if the optical coupling Ω_t is finite and the level crossing will be avoided [see solid lines in Fig. 1(b)]. For a slow-varying pulse, the state initially at $|g\rangle$, by adiabatic state transfer, would evolve to the biexciton state $|XX\rangle$ at the end, bypassing the intermediate exciton state which is separated in energy (in the rotating reference frame) rather far away from the ground state and the biexciton.

In the following, we choose the following specific functional forms for pulse envelope Ω_t and frequency sweeping $\dot{\phi}(t)$ (Ref. 31):

$$\Omega_t = A\ \mathrm{sech}(\alpha t), \tag{3a}$$

$$\dot{\phi}(t) = \mu\alpha\ \tanh(\alpha t), \tag{3b}$$

as shown in Fig. 1(a). Using these waveforms, the adiabatic eigenvalues of the Hamiltonian are computed and plotted in Fig. 1(b). As expected, the time-dependent eigenvector sweeps from $|g\rangle$ to $|XX\rangle$ as the pulse frequency is swept from above to below the two-photon resonance. We see that the exciton state $|X\rangle$ does not participate in the level crossing as its energy is far above the other two levels for the parameters used. Thus it can be inferred that the occupation of $|X\rangle$ would be kept low because the third eigenvalue δ is separated from the remaining two eigenvalues; the larger the biexciton binding energy 2δ, the lower would be the occupation of the intermediate exciton state $|X\rangle$.

The occupations of different basis eigenvectors in the evolution starting from the ground state are plotted in solid lines in Fig. 1(c). As this eigenvector changes from initially $|g\rangle$ to finally $|XX\rangle$, we expect this to be followed by the actual physical system if the pulse is sufficiently slowly varying.

It should be pointed out that, although we have chosen specific waveforms in Eq. (3) for the pulse shape, other choices are also possible provided that "anticrossing" similar to Fig. 1 can be produced. For instance, Gaussian shape for Ω_t and linear frequency sweep could also be used.[31] However, the waveforms in Eq. (3) show better adiabaticity and is used in the simulation.

III. NUMERICAL SIMULATION

The adiabaticity can be kept in two ways: by increasing the duration of process or the pulse amplitude A. The occupation of intermediate state can also be suppressed by increasing the duration. However, long duration is undesirable in experiment because of dephasing. In the following we fix the evolution duration of $t \in [-T, T]$ and investigate the adiabaticity of the state transfer as well as the intermediate-state

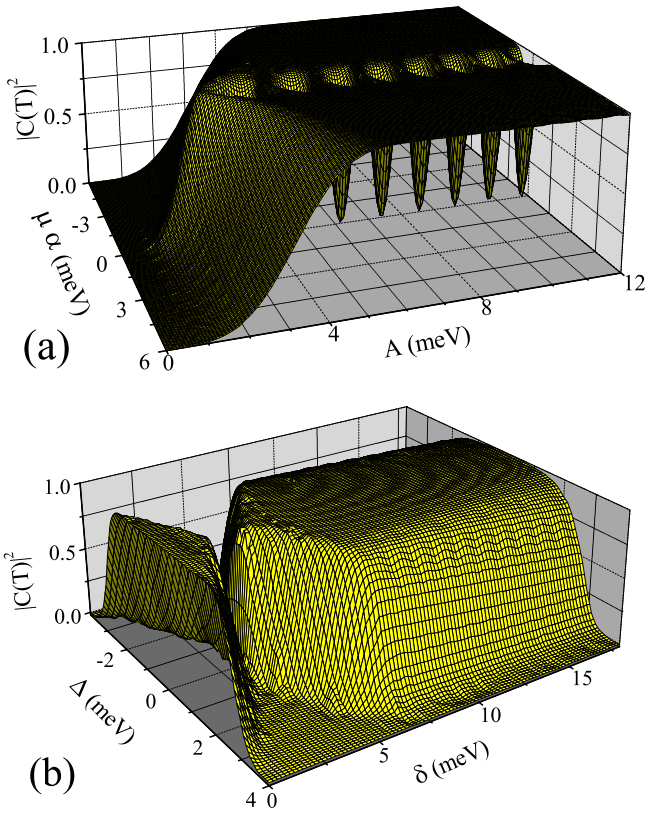

FIG. 2. (Color online) Final population of the biexciton state as a function of (a) the pulse amplitude A and the frequency-sweeping range $\mu\alpha$ for fixed biexciton binding energy $2\delta=20$ meV and pulse detuning $\Delta=0$ or (b) Δ and δ for fixed $A=6$ meV and $\mu\alpha=3$ meV. The inverse pulse duration is $\alpha=0.6$ meV≈ 1 ps^{-1} and total evolution time is $2T=160\delta^{-1}\approx 10$ ps.

population. We numerically solve the evolution $\Psi(t)=a(t)|g\rangle+b(t)|X\rangle+c(t)|XX\rangle$, with the initial condition $\Psi(0)=|g\rangle$. An example is shown as dots in Fig. 1(c). We see that the actual evolution follows the adiabatic approximation closely. Note that with $\delta\approx 10$ meV in the case of CdSe/ZnSe QDs,[15,37] the duration is $2T\approx 10$ ps, which is much shorter than the exciton dephasing time.

To investigate the robustness of the geometrical control, we show in Fig. 2 the dependence of the final biexciton population $|c(T)|^2$ on A (coupling magnitude), $\mu\alpha$ (frequency-sweeping amplitude), δ (biexciton binding energy divided by 2), and Δ (detuning). These plots have the foreseen characteristics. The case $\mu\alpha\approx 0$ corresponds to the usual case of two-photon Rabi oscillation, in which the population transfer depends sensitively on the pulse area A. This is demonstrated in the peaks and troughs along $\mu\alpha=0$ in Fig. 2(a), which are smoothed out as frequency sweeping is introduced. When the pulse amplitude A is too small ($\Omega_t\to 0$), the process fails to be adiabatic for the level splitting at the anticrossing point is small. As shown in Fig. 2(b), the state transfer is optimal for zero detuning, but a quite large detuning can be tolerated (the transfer is still almost perfect as long as Δ is well within the frequency-sweeping range $\mu\alpha$). The variation in the biexciton binding energy 2δ has also little effect on the efficacy of the state transfer as long as the intermediate exciton state is well above the

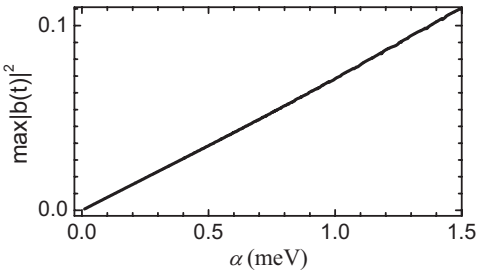

FIG. 3. The maximum intermediate exciton state population as a function of the inverse pulse duration. $\mu=2.4$ and $\delta=10$ meV are fixed, while A is adjusted for different pulse duration to minimize $\max|b(t)|^2$ while the final biexciton population is >0.99.

ground state and the biexciton state (in the rotating reference frame) plus the frequency-sweeping range [see Fig. 2(b)]. We remark that in contrast to the processes of Rabi oscillation, the adiabatic transfer is largely independent of the pulse area (A and α) and the pulse shape. This is an experimentally crucial feature, as the control over pulse area is often not exact under realistic conditions, which would make the transferred population lower than expected as in the ordinary two-photon Rabi flop scheme.

It is also of interest to investigate on the relationship between the pulse duration $\sim\alpha^{-1}$ [see Eq. (3)] and the intermediate exciton state population $\max|b(t)|^2$. In Fig. 3, we plot $\max|b(t)|^2$ as a function of α, where for each α we choose the pulse amplitude A to minimize $\max|b(t)|^2$ while the state transfer efficiency is guaranteed to be greater than 0.99. We see a near-linear correlation. This can be understood since the process completed in a short interval becomes simply a

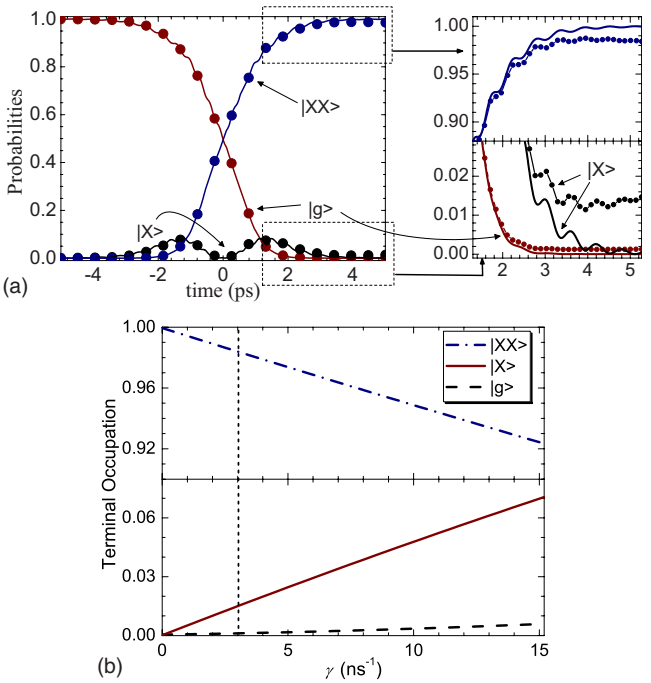

FIG. 4. (Color online) (a) State populations in the case of zero dephasing (solid lines) and finite dephasing (dots). (b) Final-state populations as functions of the dephasing rate γ_{ij}. The vertical dotted line indicates the case of (a) for $\gamma_{ij}=3.0$ ns^{-1}.

transfer via real excitation of the intermediate state ($|g\rangle \rightarrow |X\rangle \rightarrow |XX\rangle$).

IV. EFFECTS OF DEPHASING

The analysis so far is less realistic in that we have neglected the effect of dephasing present in QDs, which generally drives a pure state into a mixed state. We thus consider the relaxation and dephasing of excitons and biexcitons, which may be caused by spontaneous emission and electron-phonon scattering.[38,39] At low temperatures, we consider just the spontaneous emission as the limiting factor of the quantum operation.[40] The spontaneous emission is modeled by an additional Lindblad term[36] in the master equation for density matrix ρ as follows:

$$\partial_t \rho = -i[H_0, \rho] + L(\rho), \tag{4}$$

where the Lindblad superoperator L is defined by

$$L(\rho) = \sum_{ij} \frac{\gamma_{ij}}{2} [2\sigma_{ij}^{\dagger} \rho \sigma_{ij} - \sigma_{ij} \sigma_{ij}^{\dagger} \rho - \rho \sigma_{ij} \sigma_{ij}^{\dagger}], \tag{5}$$

with $\sigma_{ij} \equiv |i\rangle\langle j|$ and $\{i,j\} = \{|XX\rangle, |X\rangle\}$ or $\{|X\rangle, |g\rangle\}$, signifying the transition from i to j.

In the case of CdSe/ZnSe QDs, with the suppression of electron-phonon scattering at low temperatures, the dephasing rate was determined to be $\gamma_{ij} \approx 3.0$ ns^{-1}.[6] Together with $\delta = 10$ meV and the pulse shape of Eq. (3), the evolution of different state populations are plotted in Fig. 4. It shows only a slight reduction in the final population of $|XX\rangle$, while those of $|X\rangle$ and $|g\rangle$ increase.

V. CONCLUSION

We have studied the geometrical creation of biexcitons in quantum dots via adiabatic state following under the driving of a chirped pulse and demonstrated the robustness of the process against uncertainties in parameters of the system and the controlling pulse such as the pulse area, the pulse duration, the detuning, and the energy levels. The occupation of the intermediate exciton state is largely avoided during the adiabatic state transfer. Using the dephasing rate for CdSe/ZnSe quantum dots, we showed that dephasing only causes a slight reduction in efficiency. The geometrical control by a chirped pulse may be extended to apply to implementation of control gates of, e.g., superconducting qubits.[35]

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