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 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 state transfer through avoided level crossing and is 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] [2] [3] [4] [5] [6] with good indistinguishability [7]. More recently it has been proposed and partially realized [2] [8] [9] [10] [11] that QDs in two-exciton states, called biexcitons, can be used to generate pairs of entangled photons, by 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]. 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 2-bit conditional quantum logic gate [21].

A number of works have already been done on the optical coherent control of the single-exciton states in, e.g., InAs/GaAs QDs [22] [23] [24] [25] [26] and CdSe/ZnSe QDs [6]. In the recent experiments of optical coherent control of exciton states, two approaches were used. The first one applies two optical beams each in resonance with the |g⟩→|X⟩ (ground state to single exciton) and |X⟩→|XX⟩ (single- to bi-exciton) transitions [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 [29] [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 bypassed. The utilization of level anti-crossing follows the idea of the STIRAP (stimulated Raman adiabatic passage) for adiabatic state transfer in a Λ-type 3-level system [32] [33]. But here since both 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. 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, e.g., may contaminate an entangled photon pair in quantum optics application. 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 give the waveform of the pulse used; 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; in Sec. IV we show that dephasing, modeled in the Lindblad formalism [34], only slightly reduce the efficiency.

II. MODEL AND MECHANISM

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

The Hamiltonian is written as

\[ H = (\omega + \delta) |X⟩⟨X| + 2\omega |XX⟩⟨XX| + [\Omega(t)] (|g⟩⟨X| + |X⟩⟨XX|) + H.c. \] (1)

where we have defined \( \omega \) such that \( 2\omega \) is the energy between ground state and biexciton, and \( \omega + \delta \) is the energy of the single exciton, with \( \delta \) equal to half the biexciton binding energy \( \Delta E \). \( \Omega(t) \) is the time-dependent coupling cause by a laser pulse. We write the Hamiltonian in a frequency-modulated rotating reference frame, with \( \Omega(t) = \Omega_r \exp{(i(\omega - \Delta)t - i\phi_s)} \) (where...
FIG. 1: (Color online) (a) Laser pulse amplitude and time-dependent frequency as defined in Eq. (3b). (b) The three eigenvalues of Hamiltonian in Eq. (2) under the pulse in Eq. (3b) (solid lines), and when \( \Omega_t = 0 \) (dashed lines). The eigenvector corresponding to each eigenvalue at the beginning and the end of the pulse is indicated. (c) The evolution of the system eigenvector corresponding to each eigenvalue at the beginning and end of the pulse is indicated. The parameters here are \( A = 0.6 \delta, \alpha = 0.06 \delta, \mu = 5, T = 80/\delta \).

\[ \Delta = \text{detuning} \], as

\[
H = \begin{pmatrix}
-\Delta - \phi_t & \Omega_t & 0 \\
\Omega_t & \delta & \Omega_t \\
0 & 0 & \Delta + \phi_t
\end{pmatrix}.
\] (2)

Here the basis is \( \{ e^{-i(\omega - \Delta)t} | g \rangle, | X \rangle, e^{i(\omega - \Delta)t} | XX \rangle \} \).

When \( \Omega_t = 0 \), the eigenvectors of \( H \) are the three basis states, with eigenvalues \(-\phi_t, \delta, \phi_t\). We envisage that when \( \phi_t \) sweeps from negative to positive, the eigenvector would change from \( | g \rangle \) to \( | XX \rangle \), following which the system would be excited, by-passing the intermediate state adiabatically, if a pulse, being sufficiently slow-varying, is applied to induce the level anti-crossing.

In the following, we choose the following specific functional forms for \( \Omega_t \) and \( \phi_t \),

\[
\Omega_t = A \text{sech}(\alpha t), \quad \phi_t = \mu \alpha \tanh(\alpha t),
\] (3a)

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 eigenvectors \( | g \rangle \) and \( | XX \rangle \) exchange with each other. To illustrate the level anti-crossing, the eigenvalues for the case of no interactions (\( \Omega_t = 0 \)) is also plotted in dashed lines. We see that the single-exciton state does not participate in the level crossing. Thus it can be inferred that the occupation of \( | X \rangle \) would be kept low because the third eigenvalue \( \delta \) is separated from the remaining two eigenvalues; the larger \( \delta \), the lower would be the occupation of \( | X \rangle \).

The square of components of eigenvectors for the middle eigenvalue 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 slow-varying.

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

FIG. 2: (Color online) Final population of the biexciton state. Here \( \alpha = 0.6 \text{ meV} \approx 1 \text{ ps}^{-1} \) and \( T = 8 \text{ meV}^{-1} \approx 5 \text{ ps} \), which are determined from Fig. 1 with \( \delta = 10 \text{ meV} \), consistent with the binding energy of CdSe/ZnSe QDs. (a) Fixing \( \delta = 10 \text{ meV} \) and \( \Delta = 0 \), \( A \) and \( \mu \) (i.e. \( \mu \alpha \)) are varied. If \( A \) is too small, the process fails to be adiabatic. Alternatively, if \( \mu \alpha \) is too small, no anti-crossing phenomena could be observed. (b) Fixing \( A = 6 \text{ meV} \) and \( \mu \alpha = 3 \text{ meV} \), detuning \( \Delta \) and binding energy \( 2 \delta \) are varied. The result is acceptable for detuning within \( \pm 2 \text{ meV} \).
III. SIMULATIONS

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 an undesirable parameter in experiment because of dephasing. In the following we fix a duration of \( t \in [-T, T] \), and investigate the adiabaticity of the state transfer as well as the intermediate state population. We numerically solve the evolution \( \Psi(t) = a(t)|g\rangle + b(t)|X\rangle + c(t)|XX\rangle \), with initial conditions \( \Psi(0) = |g\rangle \). An example is given by the solid lines Fig. 2(c).

We see that the actual evolution follows adiabatic approximation closely. Note that with \( \delta \approx 10 \text{ meV} \) in the case of CdSe/ZnSe QDs [15, 35], the duration \( 2T \approx 10 \text{ ps} \), which is much shorter than the exciton dephasing time.

To investigate on the dependence on the parameters, we plot the final biexciton population \( |c(T)|^2 \) as a function of \( A \) (coupling magnitude) and \( \mu \alpha \) (frequency-sweeping amplitude) in Fig. 2(a), \( \delta \) (binding energy/2) and \( \Delta \) (detuning) in Fig. 2(b). These plots have some foreseeable 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. It is optimal for zero detuning, but some deviation of \( \Delta \) can be accepted. Uncertainty in \( \delta \) (as large as \( \pm 2 \text{ meV} \)) would not affect the efficacy of this process, either.

From the figures we remark that in contrast to the processes of Rabi oscillation, this process is largely independent of the pulse area (\( A \) and \( \alpha \)) and even the pulse shape. This is an experimentally crucial feature, as the control over pulse area is often inexact 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 relation between time duration \( T \sim \alpha^{-1} \) in Eq. (3b), and the single-exciton intermediate population \( \text{max} |b(t)|^2 \). In Fig. 3 we plot \( \text{max} |b(t)|^2 \) as a function of \( \alpha \), where for each \( \alpha \) we use large enough \( A \) such that the transferred population \( (|g\rangle \rightarrow |XX\rangle) \) is larger than 0.99 while max \( |b(t)|^2 \) is minimized. We see a near-linear correlation. Physically we understand that when we limit the process to complete in shorter interval, the process becomes simply a 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 pure state into mixed state. We thus consider the relaxation and dephasing of excitons and biexcitons, which may be caused by spontaneous emission and electron-phonon scattering [36, 37]. At low-temperatures, we consider just the spontaneous emission as the limiting factor of the quantum operation [38]. The spontaneous emission is modelled by an additional Lindblad term in the master equation for density matrix \( \rho \):

\[
\dot{\rho} = -i[H_0, \rho] + L(\rho),
\]

where the Lindblad super-operator \( L \) is defined by:

\[
L(\rho) = \sum_{ij} \frac{\gamma_{ij}}{2} \left[ 2\sigma_i^\dagger \sigma_j \rho \sigma_i \sigma_j^\dagger - \sigma_i \sigma_j^\dagger \sigma_j \sigma_i^\dagger \rho - \rho \sigma_i \sigma_j \sigma_j^\dagger \sigma_i^\dagger \right],
\]
with \( |i, j\rangle = \{|XX\rangle, |X\rangle\}, \) or \(|X\rangle, |g\rangle\), signifying the transition from \(i\) to \(j\).

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

V. CONCLUSION

We have demonstrated the process of biexciton creation which is robust against uncertainties in parameters of the system and the controlling pulse, and keeps the involvement of single exciton state low. Using a specific pulse shape, we showed the physical system adiabatically follows the eigenvector. By plotting the final biexciton population against various parameters, we have demonstrated the efficacy of the proposed process does not have sensitive dependence on the pulse area, pulse duration, level position, and detuning, which is the case for the two-photon Rabi flop scheme. The maximum occupation of single exciton state has an approximate linear relationship to the inverse duration of the process, which means that the single-exciton state could be kept low provided that sufficient time is given. Using the experimental dephasing rate for CdSe/ZnSe quantum dots, we showed that dephasing only causes a slight reduction in efficiency.

[1] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science 290, 2282 (2000).
[2] O. Benson, C. Santori, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. 84, 2513 (2000).
[3] E. Moreau, I. Robert, J. M. Gérard, I. Abram, L. Manin, and V. Thierry-Mieg, Appl. Phys. Lett. 79, 2865 (2001).
[4] Z. Yuan, B. E. Kardynal, R. M. Stevenson, A. J. Shields, C. J. Lobo, K. Cooper, N. S. Beattie, D. A. Ritchie, and M. Pepper, Science 295, 102 (2002).
[5] C. Santori, M. Pelton, G. Solomon, Y. Dale, and Y. Yamamoto, Phys. Rev. Lett. 86, 1502 (2001).
[6] T. Flissikowski, A. Hundt, M. Lowisch, M. Rabe, and F. Henneberger, Phys. Rev. Lett. 86, 3172 (2001).
[7] C. Santori, D. Fattal, J. Vuckovic, G. Solomon, and Y. Yamamoto, Nature 419, 594 (2002).
[8] P. Michler, A. Imamoglu, M. Mason, P. Carson, G. Strouse, and S. Buratto, Nature 406, 968 (2000).
[9] C. Santori, D. Fattal, M. Pelton, G. Solomon, and Y. Yamamoto, Phys. Rev. B 66, 45308 (2002).
[10] S. M. Ulrich, S. Strauf, P. Michler, G. Bacher, and A. Forchel, Appl. Phys. Lett. 83, 1848 (2003).
[11] M. E. Reimer, M. Korkusinski, J. Lefebvre, J. Lapointe, P. J. Poole, G. C. Aers, D. Dalacu, W. R. McKinnon, S. Frederick, P. Hawrylak, et al., quant-ph/0706.1075 (2007).
[12] E. Moreau, I. Robert, L. Manin, V. Thierry-Mieg, J. M. Gérard, and I. Abram, Phys. Rev. Lett. 87, 183601 (2001).
[13] T. M. Stace, G. J. Milburn, and C. H. W. Barnes, Phys. Rev. B 67, 85317 (2003).
[14] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. 76, 3005 (1996).
[15] V. D. Kulakovskii, G. Bacher, R. Weigand, T. Kümmell, A. Forchel, E. Borovitskaya, K. Leonardi, and D. Hommel, Phys. Rev. Lett. 82, 1780 (1999).
[16] N. Akopian, N. H. Linder, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. 96, 130501 (2006).
[17] R. M. Stevenson, R. J. Young, P. Atkinson, K. Cooper, D. A. Ritchie, and A. J. Shields, Nature 439, 179 (2006).
[18] E. Knill, R. Laflamme, and G. J. Milburn, Nature 409, 46 (2001).
[19] A. K. Ekert, Phys. Rev. Lett. 67, 661 (1991).
[20] T. Jennewein, C. Simon, G. Weih, H. Weinfurter, and A. Zeilinger, Phys. Rev. Lett. 84, 4729 (2000).
[21] X. Li, Y. Wu, D. Steel, D. Gammon, T. H. Stievater, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Science 301, 809 (2003).
[22] N. H. Bonadeo, J. Erland, D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, Science 282, 1473 (1998).
[23] H. Htoon, T. Takagahara, D. Kulik, O. Baklenov, A. L. Holmes Jr, and C. K. Shih, Phys. Rev. Lett. 88, 87401 (2002).
[24] H. Kamada, H. Gotoh, J. Temmyo, T. Takagahara, and H. Ando, Phys. Rev. Lett. 87, 246401 (2001).
[25] T. H. Stievater, X. Li, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. 87, 133603 (2001).
[26] A. Zrenner, E. Beham, S. Stufler, F. Findeis, M. Bichler, and G. Abstreiter, Nature 418, 612 (2002).
[27] G. Chen, T. H. Stievater, E. T. Batteh, X. Li, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. 88, 117901 (2002).
[28] I. Akimov, J. Andrews, and F. Henneberger, Phys. Rev. Lett. 96, 67401 (2006).
[29] T. Flissikowski, A. Betke, I. A. Akimov, and F. Henneberger, Phys. Rev. Lett. 92, 227401 (2004).
[30] S. Stufler, P. Machnikowski, P. Ester, M. Bichler, V. M. Axt, T. Kuhn, and A. Zrenner, Phys. Rev. B 73, 125304 (2006).
[31] D. Goswami, Phys. Rep. 374, 385 (2003).
[32] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. 70, 1003 (1998).
[33] T. A. Laine and S. Stenholm, Phys. Rev. A 53, 2501 (1996).
[34] G. Lindblad, Commun. Math. Phys. 48, 119 (1976).
[35] B. Patton, W. Langbein, and U. Woggon, Phys. Rev. B 68, 125316 (2003).
[36] J. M. Villas-Boas, S. E. Ulloa, and A. O. Govorov, Phys. Rev. Lett. 94, 57404 (2005).
[37] J. Förstner, C. Weber, J. Danckwerts, and A. Knorr, Phys. Rev. Lett. 91, 127401 (2003).
[38] P. Palinginis, H. Wang, S. V. Goupalov, D. S. Citrin, M. Dobrowolska, and J. K. Furdyna, Phys. Rev. B 70, 073302 (2004).