Creation of entangled states in coupled quantum dots via adiabatic rapid passage

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Quantum state preparation through external control is fundamental to established methods in quantum information processing and in studies of dynamics. In this respect, excitons in semiconductor quantum dots are of particular interest, since their coupling to light allows them to be driven into a specified state using the coherent interaction with a tuned optical field, such as an external laser pulse. We propose a protocol, based on adiabatic rapid passage, for the creation of entangled states in an ensemble of pairwise coupled two-level systems, such as an ensemble of coupled quantum dots. We show by quantitative analysis using realistic parameters for semiconductor quantum dots that this method is feasible where other approaches are unavailable. Furthermore, this scheme can be generically transferred to some other physical systems, including circuit QED, nuclear and electron spins in solid-state environments, and photonic coupled cavities.

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I. INTRODUCTION

Whether one is interested in methods of quantum optics or computation, or more general many-body dynamics, it is important to be able to prepare interacting few-level systems in a range of initial states—from simple product states to complex entanglements. Excitons in low-dimensional semiconductors exemplify such a level structure. They are of interest both because their strong coupling to light provides communication with the external world and because interactions between excitons can be substantial. However, the lack of degeneracy due to the inhomogeneous broadening of the exciton levels becomes a challenge for manipulating them-either independently, for example, to invert a single transition, or collectively, for entangling a specified set. Controlled creation of a single exciton in a quantum dot has been demonstrated by using resonant laser pulses to induce Rabi flopping. 1-4 Although effective in certain cases, Rabi flopping has the disadvantage that the final state is sensitive to variations in dipole couplings and exciton energy. Such disorder becomes a serious challenge if one wishes to scale this approach beyond a single quantum dot, and similar limitations arise in many other systems.

More recently, the creation of a single exciton in a quantum dot has been demonstrated 5.6 using adiabatic rapid passage 7 (ARP). In this scheme the dot is driven by a chirped laser pulse, whose frequency is swept through the target one exciton state. This creates an adiabatic evolution from the initial ground state to this one exciton state, and the latter is populated with high probability. Unlike the Rabi approach, ARP is largely unaffected by the variations previously mentioned. This has prompted a variety of theoretical proposals for applications of ARP schemes, including the preparation of exciton populations in ensembles of quantum dots and the implementation of quantum operations 9-16 and forms of Bose-Einstein condensation. 8,17,18

The aim of this paper is to show how ARP may be generalized to create entangled states in a disordered system,

consisting of many two-level systems coupled together in pairs. Such an ensemble could be realized using coupled quantum dots,¹⁹ i.e., quantum dot molecules, with a possible coupling mechanism being resonant Förster energy transfer.^{11,20–22} Other implementations could be considered in systems such as coupled photonic cavities (with photons tunneling between neighboring cavities and strong intracavity nonlinearities²³), arrays of superconducting qubits (coupled by the exchange of virtual photons^{24,25}), and impurity states in semiconductors.²⁶

An important feature of our proposal is that it does not require precise engineering of individual quantum dots. In particular, we do not require that the dots in a particular pair have almost degenerate exciton levels. Instead, we consider an ensemble of pairs, a subset of which will obey this criterion. The excited states of this subset include spatially entangled states that could be identified spectrally. The robustness of ARP, then, allows a pulse to be constructed that transfers these pairs into their entangled excited states, without exciting the others. Thus, as shown in Fig. 3, the entanglement of formation per excitation could be very close to one, even within a strongly disordered ensemble. Our approach represents a significant simplification and improvement of the protocol required for the production of entangled states in realistic systems, since previous proposals have relied on coupling two states through a further excited level¹³ or have considered only a single and fine-tuned (degenerate) system. 9-11,27,28

The paper is organized as follows. In Sec. II we present the model we consider for a pair of coupled quantum dots driven by an external field and recall the mechanism of ARP. In Sec. III we discuss the application of ARP to generating an entangled state in a pair of (coupled) quantum dots. We examine the cases of both degenerate and nondegenerate dots and describe the pulse parameters required for the procedure. In Sec. IV we consider the extension to creating entanglement within a disordered ensemble of coupled dots. In Sec. V we briefly consider the effects of dephasing, and in Sec. VI we summarize our conclusions.

II. MODEL

To simplify the notation we consider a limit in which there are only two relevant states, a ground state and a one exciton state, for each dot. The generalization to more complex level structures, such as those arising from the exciton spin, is straightforward. We represent the states of each dot in terms of a Bloch vector or pseudospin, with spin up (down) corresponding to the one (no) exciton state, and make the rotating-wave approximation. The relevant Hamiltonian for two coupled dots, in a frame rotating with the time-dependent frequency $\omega(t)$ of the laser pulse, is then ($\hbar=1$)

$$\hat{H} = \sum_{i=1,2} \left[\frac{\tilde{\varepsilon}_i(t)}{2} \hat{\sigma}_i^z + g(t)(\hat{\sigma}_i^+ + \hat{\sigma}_i^-) \right] - j_T(\hat{\sigma}_1^+ \hat{\sigma}_2^- + \hat{\sigma}_2^+ \hat{\sigma}_1^-).$$
 (1)

Here, $\tilde{\varepsilon}_i(t) = \varepsilon_i - \omega(t)$, ε_i is the exciton energy on dot i = 1, 2, g(t) is the time-dependent amplitude of the driving field used to perform ARP, and σ_i are the Pauli operators describing the state of dot i. j_T is a coupling between the two dot states, arising, for example, from Förster transfer. ^{11,20–22} We have decomposed the driving field, F(t), into amplitude and frequency as

$$F(t) = g(t)\exp\left[i\int^{t} \omega(t')dt'\right]. \tag{2}$$

For definiteness, we consider excitation with a chirped Gaussian:

$$F(t) = g_0 e^{-t^2/2\tau^2} e^{i \int_0^t \omega(t') dt'}, \ \omega(t) = \omega_0 + \alpha t,$$
 (3)

where g_0 defines the pulse amplitude, τ its duration, ω_0 its central frequency, and α the chirp. We take the pulse duration to define units of time and energy and use dimensionless parameters $\alpha \tau^2$, $g_0 \tau$, $j_T \tau$, and $\delta \tau$ ($\delta = \varepsilon_1 - \varepsilon_2$ being the detuning between the uncoupled exciton levels).

In general, ARP schemes are generalizations of the Landau-Zener problem^{29,30} to a time-dependent and controllable mixing g(t) between the levels. Consider first noninteracting dots $(j_T = 0)$: if the driving frequency sweeps through the transition frequency of a dot, $\tilde{\epsilon}_i(t)$ passes through zero, and the lowest energy state of Eq. (1) changes from the zero to the one exciton state. In the Landau-Zener problem, there is a constant mixing g_0 between these levels that generates an avoided crossing. Provided the resulting gap is large compared with the rate of change of the energy levels, the evolution is adiabatic, and a system in the initial ground state will be driven into the excited state with a probability close to one, P_{inv} = $1 - \exp(-g_0^2/\alpha)$. In the case of ARP, the mixing depends on time and must be significant during the period when the detuning $\tilde{\varepsilon}_i(t)$ is small if the evolution is to be adiabatic. This introduces the requirement $\alpha \tau^2 \gg 1$. However, the time dependence of the amplitude g(t) also allows ARP to be made spectrally selective. For an uncoupled pair, for example, one can choose to excite one dot or the other by having the pulse on $[g(t) \neq 0]$ only while the frequency sweeps through the desired level. The available spectral discrimination is thus of order \hbar/τ .

III. ENTANGLEMENT OF TWO COUPLED TWO-LEVEL SYSTEMS

The spectral discrimination of ARP could be used to create entangled states in the ideal case of a pair of identical two-level systems. $^{9-11}$ In this case $\delta=0$, and in the absence of the driving field Eq. (1) is diagonal in the singlet-triplet basis $\{|S\rangle,|T_-\rangle,|T_0\rangle,|T_+\rangle\}$, with $|S\rangle=(1/\sqrt{2})(|\downarrow\uparrow\rangle-|\uparrow\downarrow\rangle)$, $|T_-\rangle=|\downarrow\downarrow\rangle,|T_0\rangle=(1/\sqrt{2})(|\downarrow\uparrow\rangle+|\uparrow\downarrow\rangle),|T_+\rangle=|\uparrow\uparrow\rangle$. The intermediate states $|T_0\rangle$ and $|S\rangle$ between the ground state and the fully occupied state $|T_+\rangle$ are spatially entangled if the coupling dominates over the detuning, $\Delta\varepsilon=\varepsilon_1-\varepsilon_2\lesssim j_T$.

For a single pair of identical dots ($\varepsilon_1 = \varepsilon_2 = \varepsilon_0$), the use of ARP to create this entangled state is illustrated in Fig. 1, which shows the time evolution of the eigenvalues of the Hamiltonian [Eq. (1)] when the levels are coupled by a linearly chirped Gaussian pump ($g_0\tau = 5$, colored continuous lines) and uncoupled ($g_0\tau = 0$, thin dashed lines). By chirping through the level crossing (A) between the states $|T_-\rangle$ and $|T_0\rangle$ (at $\omega_0 = \varepsilon_0 - j_T$) with a pulse duration such that coupling switches off before the $|T_0\rangle \rightarrow |T_+\rangle$ crossing (B), the system will be adiabatically driven and left in the entangled triplet state $|T_0\rangle$.

We now consider the parameters required for this procedure for a single pair and examine both the cases of degenerate $(\delta=0)$ and spectrally detuned $(\delta\neq0)$ dots. To evaluate the entanglement produced, we use the *entanglement of formation* (EOF), which is a widely accepted measure of the entanglement for bipartite states. For a pure state $|\Psi\rangle$ the EOF $E=E(|\Psi\rangle)$ is given by 31

$$E(|\Psi\rangle) = S(\rho),\tag{4}$$

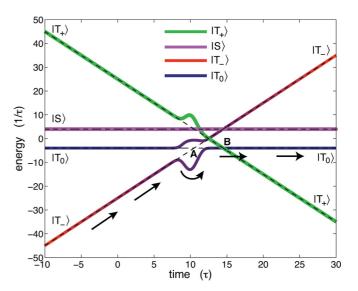


FIG. 1. (Color online) Time-dependent energy levels of a coupled system of two identical dots. The continuous colored lines indicate the energy levels with a Gaussian driving pulse $(g_0\tau=5)$ of duration τ , while the thin dashed lines are the undriven levels $(g_0=0)$. The chirp and interdot couplings are $\alpha\tau^2=2$ and $j_T\tau=4$. The central frequency of the ARP pulse is ε_0-j_T , resonant with the transition between the ground state $|T_-\rangle$ and the entangled state $|T_0\rangle$ (see point A).

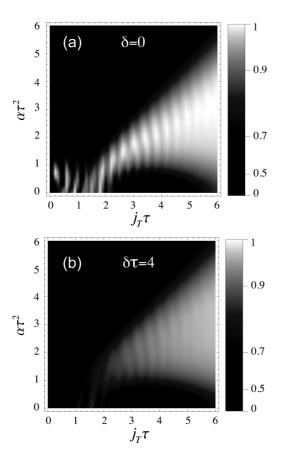


FIG. 2. Entanglement of formation (cubic scale) in two coupled dots as a function of the dimensionless linear chirp, $\alpha \tau^2$, and coupling, $j_T \tau$. The remaining parameters are the same as described in the legend of Fig. 1. In each case, the frequency of the pulse at its peak, ω_0 , is tuned to coincide with that of the triplet state $|T_0\rangle$: $2\omega_0 = (\varepsilon_1 + \varepsilon_2) - \sqrt{(\varepsilon_1 - \varepsilon_2)^2 + 4j_T^2}$. In (a) the two dots have degenerate energy levels, $\varepsilon_1 = \varepsilon_2 = \varepsilon_0$ and $\omega_0 = \varepsilon_0 - j_T$, while in (b) the dots are spectrally detuned by $\delta \tau = 4$. The white region (EOF = 1) shows the range of values of chirp and exchange coupling where the pair is driven into the entangled state $|T_0\rangle$ with high probability.

where $S(\rho) = -\mathrm{Tr}(\rho^{\mathrm{red}}\log_2\rho^{\mathrm{red}})$ is the von Neumann entropy and $\rho^{\mathrm{red}} = \mathrm{Tr}^{\mathrm{red}}|\Psi\rangle\langle\Psi|$ is the reduced density matrix obtained by tracing the whole system density matrix $|\Psi\rangle\langle\Psi|$ over one of the two subsystems of which the pure state $|\Psi\rangle$ consists. In this case, the two subsystems are the two paired dots. The EOF ranges from zero (for a product state) to $\log_2 N$ for a maximally entangled state of two N-state particles. Hence, the EOF of the triplet state $|T_0\rangle = (1/\sqrt{2})(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ (which is a maximally entangled Bell state) is equal to one.

Figure 2 shows the EOF calculated as a function of the dimensionless parameters $\alpha \tau^2$ and $j_T \tau$ in the resonant case $\varepsilon_1 = \varepsilon_2$ [$\delta = 0$, Fig. 2(a)], and when the two dots are detuned by $\delta \tau = 4$ [Fig. 2(b)]. In the ideal resonant case, the region where the EOF is one is seen to extend over a significant range of realistic values of chirp and coupling. In the presence of a significant detuning [see Fig. 2(b)] this region slightly shrinks and darkens, as arbitrarily detuned quantum systems cannot be spatially entangled, resulting in an EOF always smaller than one. Smaller values of the detuning yield very

little differences compared to the ideal resonant case shown in Fig. 2(a), indicating the robustness of the adiabatic protocol even in the presence of inhomogeneities.

The ARP parameters we have used are fully compatible with a linearly chirped pulse similar to that applied to invert a single quantum dot: we have considered a transform-limited pulse width of 2 ps and a chirped temporal width $\tau = 4.5$ ps. Thus, for $\alpha \tau^2 \approx 2$ we require a dimensionless coupling $j_T \tau \gtrsim$ 4 [see Fig. 2] for the ARP transition to the entangled state to occur, which corresponds to a value of Förster coupling of 0.6 meV. Previous studies have estimated an upper limit of 10 meV for Förster coupling in semiconductor quantum dots.²² Thus, the scheme could be implemented using two stacked (vertical) quantum dots at a distance of few nanometers, coupled by Förster energy transfer, but without single-particle tunneling. Such conditions can be achieved in InAs/GaAs coupled quantum dots. 32,33 The scheme would also apply to tunnel-coupled dots, ¹⁹ provided target entangled states could be identified.³⁴

IV. APPLICATION TO INHOMOGENEOUS ENSEMBLES

We now show how this adiabatic protocol can be generalized to create entanglement in ensembles of coupled quantum dots, in which there are significant fluctuations in the dot energy. In this case, the entanglement pulse can be spectrally tuned to address a specific pair of dots; several entangled pairs could be created within an ensemble by superposing such pulses.

It is important to recognize that the requirement of exact degeneracy of the uncoupled transition is relaxed up to the magnitude of the coupling energy. This affords a route to practical realizations of the scheme, as the coupling energy and level splitting can be traded to optimize the probability of producing an appropriate double-dot structure. In the following, we consider an ensemble of such systems, modeled here by an average coupling strength and having an inhomogeneous distribution of energies. In each coupled pair the energy levels are not degenerate, and only pairs with detunings smaller than the interdot coupling strength can be entangled. Hence, the desired entangled states, which arise from strongly coupled dots, must be identified spectrally prior to ARP manipulation. We suggest that this could be done by exploiting two-dimensional nonlinear spectroscopy,^{35–38} in which a strongly coupled pair gives an off-diagonal peak in the two-dimensional four-wave mixing spectrum; such pairs are rare and would be isolated in a sufficiently dilute ensemble. Then, spectrally selected components of a broad-band pulse, each close to resonance with a particular chosen pair, can be chirped in the same linear optical process, such as a grating-based delay stage.

We test this scheme simulating an ensemble consisting of 30 pairs of dots with energies taken from a Gaussian distribution of standard deviation 10 meV, and coupled by an average coupling strength $j_T = 2$ meV. In one typical realization taken as an example, three couples can be entangled as their energy levels are detuned by an amount smaller than $j_T/2$. Figure 3 shows the EOF and the total excitation in a typical realization calculated as a function of the strength of the applied chirped Gaussian pulse. We recall that the properties of the EOF as a natural measure of the entanglement include that the

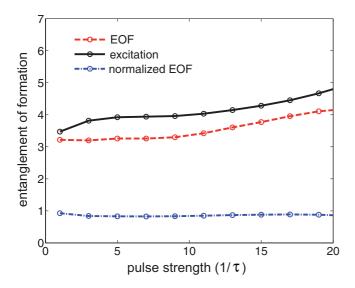


FIG. 3. (Color online) Predicted entanglement generated when an ensemble of 30 coupled pairs of dots is driven by an ARP pulse. The pulse is constructed to drive the three strongly coupled pairs in the ensemble into their entangled state (see text). The red dashed curve shows the total entanglement of formation, the black solid curve the total excitation, and the blue dot-dashed curve the entanglement normalized by the total excitation. The horizontal axis corresponds to the peak amplitude of the driving field, g_0 , in units of the inverse pulse duration $1/\tau$.

entanglement of independent systems, such as pairs of coupled dots in an ensemble, is additive. The excitation induced in the system after the application of an external pulse has been evaluated as $X = \text{Tr}(\rho \hat{X})$, with the excitation operator $\hat{X} =$ $(\sigma_1^z + \sigma_2^z)/2 + 1$. We have used $\tau = 15$ ps and $\alpha = 0.01$ ps⁻² $(\alpha \tau^2 = 2.25)$ yielding a small value of $\alpha \tau$ —which determines the energy range spanned by each component of the ARP pulse—while keeping the whole process in the adiabatic regime $\alpha \tau^2 > 1$, so that few states apart from the entangled ones will be excited. The total entanglement (dashed red curve) always deviates from the ideal value of three, i.e., the total number of triplet states, due to the unavoidable excitation of other states that are not entangled, but yield a small amount of entropy. Furthermore, as the intensity of the pulse increases, other states such as the two-exciton state $|\uparrow\uparrow\rangle$ can be excited, thus explaining the behavior of the excitation (continuous black) curve. However, the EOF normalized with respect to the total excitation (dashed-dotted blue curve), which is an effective measure of the total entanglement produced in the system, is close to the ideal value of one (≈ 0.85 for all the different pump strengths considered). In order to validate these results we have simulated 50 different realizations of Gaussian-distributed coupled dots and found an average value for the normalized EOF to be approximately 0.8 for several realistic values of pulse strengths.

V. DEPHASING

We now consider the extent to which dephasing would affect our results. Typical experimental lifetimes and dephasing times for quantum dot excitons are several hundreds of picoseconds,³⁹ while the control pulses we consider have

durations of several picoseconds. We expect the probability that the process fails due to dephasing to be of the order of the ratio of these quantities, 10^{-2} . While shorter pulses could reduce the failure probability, their reduced spectral selectivity will eventually lead to the excitation of other states. Thus, within a model of a constant dephasing rate, the failure probability is limited by the ratio of the dephasing rate to the coupling (which is $\sim 10^{-3}$, for a coupling of 5 meV and a dephasing time of 100 ps).

In addition, however, we note that driving a quantum dot with a laser changes the apparent dephasing rate, as recently discussed for both Rabi flopping 40,41 and ARP 42,43 experiments on single dots. Such excitation-controlled dephasing arises because the field dresses the exciton states, and phonons can induce transitions between these dressed states. In our case, such dephasing will arise if a phonon is absorbed near to point A in Fig. 1, causing a transition out of the desired adiabatic state. The significance of this effect may be estimated using the absorption rate for a single dot [Eq. (8) in Ref. 42]:

$$\gamma_a = 2\left(\frac{g(t)}{\Lambda}\right)^2 \pi J(\Lambda) n(\Lambda),$$
(5)

where $\Lambda = \sqrt{\tilde{\epsilon}(t)^2 + 4g(t)^2}$ is the dressed-state splitting, and $n(\Lambda)$ is the Bose function. $J(\omega) \approx (0.03 \, \mathrm{ps^{-2}}) \omega^3 e^{-\omega^2/\omega_c^2}$ is the phonon spectral density, with cut-off frequency $\omega_c \approx 2 \, \mathrm{ps^{-1}}$. This damping rate, γ_a , varies with time through its dependence on the driving field. The minimum of the corresponding lifetime, $1/\gamma_a$, is approximately 300 ps at a temperature of 4 K, again leading to an estimate $\sim 10^{-2}$ for the probability of failure with picosecond pulses. As in the ARP protocol for a single dot, 42,43 the process will be less reliable for negative chirp rates, where phonon emission will also occur, as well as at higher temperatures.

VI. CONCLUSIONS

In summary, we have demonstrated that ARP could be used for the generation of entangled states in strongly inhomogeneous systems, such as ensembles of coupled quantum dots. Our calculations, based on realistic values for Förster coupling and dot distribution, provide a feasible route for the realization of entanglement in solid-state systems, which is more practical than other approaches due to the flexible and robust nature of ARP. In particular, the flexibility of ARP relaxes the demand of homogeneity for the dots, to the extent that entangled pairs could be created within relatively small ensembles without tuning of individual dots (Fig. 3). In principle, the same methodology can apply to radiatively coupled Josephson junctions,⁴⁴ to coupled electron spins in semiconductors⁴⁵ (using ESR), and to coupled nuclear spins (via NMR). The latter arena is the genesis of the ARP technique, though we are not aware of it being explicitly used in the form proposed here.

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