Dephasing in the adiabatic rapid passage in quantum dots: the role of phonon-assisted biexciton generation

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We study the evolution of an exciton confined in a quantum dot adiabatically controlled by a frequency-swept (chirped) laser pulse in the presence of carrier-phonon coupling. We focus on the dynamics induced by a linearly polarized beam and analyze the decoherence due to phonon-assisted biexciton generation. We show that if the biexciton state is shifted down by a few meV, as is typically the case, the resulting decoherence is strong even at low temperatures. As a result, efficient state preparation is restricted to a small parameter area corresponding to low temperatures, positive chirps and moderate pulse areas.

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I. INTRODUCTION

The recently demonstrated1–2 high-fidelity preparation of a single exciton state in a self-assembled quantum dot (QD) by means of adiabatic evolution induced by a chirped laser pulse (referred to as adiabatic rapid passage, ARP) opens new possibilities of charge control in QDs. In contrast to the more traditional Rabi flops as well as to methods based on voltage control3–5, ARP is much less sensitive to the details of the driving field. In particular, in the ideal case, the ARP technique ensures the full inversion of occupation between the empty dot and exciton states as soon as the pulse intensity reaches the threshold for the adiabatic passage.

However, as QDs are embedded in a semiconductor crystal matrix, carrier-phonon interactions are usually found to considerably limit the fidelity of various optical control schemes. The phonon-induced dephasing process is inevitable in such systems and leads to loss of information. These mechanisms have been investigated theoretically6–9 and have been confirmed experimentally10,11. As we have recently shown11, in the case of the ARP the role of phonon coupling in the state preparation is essential. As the adiabatic evolution follows one of the spectral branches corresponding to dressed exciton states, carrier phonon coupling leads to transitions to the other branch. Depending on whether the upper or lower branch is followed (which, in turn, depends on the direction of the chirp), the transition is assisted by phonon emission or absorption, respectively. As a consequence, at low temperatures the phonon-induced decoherence leads to a strong asymmetry in the final occupation of the exciton state depending on the sign of the chirp11. While the analysis performed in Ref. 11 was based on a two-level model, corresponding to the excitation with a circularly polarized laser pulse, in actual experiments linearly polarized beams have been used. In this case, the selection rules allow the coupling to the biexciton state and phonon-assisted biexciton generation may occur even at low temperatures, as the biexciton state is typically shifted to lower energies.

In this paper, we study the evolution of the biexciton system driven by a linearly polarized chirped pulse in the regime of the ARP in the presence of the coupling to acoustic phonons. We show that including the biexciton state, which becomes relevant for linearly polarized laser pulses, indeed opens a new decoherence path related to phonon-assisted transitions to the biexciton state. As a result, the fidelity of exciton state preparation drops down considerably in particular in most of the parameter areas where almost ideal evolution was predicted by the two-level model. Thus, high-fidelity control becomes restricted to the range of moderate pulse areas for positive chirps and at low temperatures only, unless circular polarized pulses are used to suppress biexcitonic excitations.

The paper is organized as follows. In Sec. II we describe the system and define the model used in our study. Sec. III introduces our method of simulation of the open system dynamics. The results are presented in Sec. IV. Finally, Sec. V concludes the paper.

II. MODEL

The biexciton system in the QD, restricted to the lowest bright states, is modeled as a four-level system with |0⟩ representing the empty dot, |X⟩, |Y⟩ being the two exciton states with different linear polarizations and |B⟩ denoting the biexciton state. The corresponding Hamiltonian is

\[ H_X = E_0 (|X⟩⟨X| + |Y⟩⟨Y|) + (2E_0 + E_B)|B⟩⟨B|, \]

where \( E_0 \) is the exciton transition energy, \( E_B \) is the biexciton shift, and we neglect the fine structure splitting.
which is irrelevant on the picosecond time scales considered here.

This charge system is driven by a chirped Gaussian pulse with the original envelope

$$\Omega_0(t) = \frac{\Theta}{\sqrt{2\pi \tau_0}} \exp \left( -\frac{t^2}{2\tau_0^2} \right),$$

where \(\Theta\) is the original pulse area (before chirping) and \(\tau_0\) is the initial duration (pulse length). The central frequency \(\omega_0\) is chosen at resonance with the single exciton transition. We assume that the linear chirp is effected by passing this pulse through a Gaussian chirp filter with the chirp coefficient \(\alpha\). This results in the chirped pulse envelope

$$\Omega(t) = \frac{\Theta}{\sqrt{2\pi \tau_0 \tau}} \exp \left( -\frac{t^2}{2\tau^2} \right),$$

with the chirped pulse length \(\tau = (\alpha^2/\tau_0^2 + \tau_0^2)^{1/2}\), and the frequency chirp rate \(\alpha = \alpha/(\alpha^2 + \tau_0^2)\). The Hamiltonian describing the coupling to the chirped pulse in the rotating wave approximation is

$$H_{\text{int}} = \frac{\hbar \Omega(t)}{2} (|X\rangle\langle X| + |X\rangle\langle B|) e^{i\omega_0 t + i\alpha t^2/2} + \text{h.c.}$$

Thus, for one linear polarization, the coupling is \(|0\rangle \leftrightarrow |X\rangle \leftrightarrow |B\rangle\) and the other single-exciton state \(|Y\rangle\) is decoupled (it is not coupled by phonon transitions, either). Therefore, only a three-level model is needed.

The idea of the adiabatic passage is to drive the system state adiabatically along one of the spectral branches corresponding to the instantaneous eigenstates of the Hamiltonian \(H_0 = H_X + H_{\text{int}}\). These instantaneous eigenstates are shown in Fig. 1 for the biexciton shift \(E_B = -2.0\, \text{meV}\). The branch followed by the system state is plotted with a thicker line. In Fig. 1(a), we assume a positive chirp \((\alpha > 0)\), while in Fig. 1(b) a negative chirp is assumed. For pulse intensities above the ARP threshold, the central anticrossing at \(t = 0\) (between the vacuum and exciton states) is passed adiabatically along a single spectral branch. On the contrary, the two other anticrossings away from \(t = 0\) (excitonic-biexciton and vacuum biexciton) are met when the pulse is already very weak, hence they are very narrow (in the case shown here, the widths of both of these anticrossings are below 10 \(\mu\text{eV}\), unresolved in Fig. 1). At such narrow anticrossings, the sweep rate is very high compared to the distance between the two levels, which leads to a nearly purely non-adiabatic evolution: these anticrossings are almost completely crossed by the evolving system state, that is, the system nearly completely jumps to the other branch (as shown by color coding of the lines in Fig. 1). As a result, the charge configuration does not change here, as opposed to the central anticrossing.

For the parameter range corresponding to the experiments, excitation of optical phonon modes is excluded by the spectral properties of the system. In consequence, the coupling to acoustic phonons can be expected to dominate as a source of decoherence. The carrier-phonon interaction is described in terms of the usual independent boson Hamiltonian, \(H_{\text{int}} = S \otimes R\), with

$$S = |X\rangle\langle X| + |Y\rangle\langle Y| + 2|B\rangle\langle B|$$

and

$$R = \sum_k g_k b_k + \text{h.c.},$$

where \(b_k\) are phonon annihilation operators. We assume that the biexcitonic and excitonic wave functions can be factorized into products of single particle wave functions which are the same for both exciton and biexciton. This is well justified in the strong confinement limit where the wave functions are mostly determined by the confinement potentials. The coupling constants, accounting for the deformation potential interaction, can then be written as

$$g_k = \sqrt{\frac{\hbar k}{2\rho v c}} (D_{e\text{h}} F_e(k) - D_{h\text{h}} F_h(k)),$$

where \(v\) is the normalization volume, \(\rho\) is the crystal density, \(c\) is the speed of sound, \(D_{e\text{h}}\) is the deformation potential constant for an electron (hole) and

$$F_e(k) = \int d^3r |\psi_e(k)(r)|^2 e^{ik \cdot r},$$

are the form factors for electron (hole) wave function \(\psi_e(k)(r)\). Finally, the phonon reservoir is described by the Hamiltonian

$$H_{\text{ph}} = \sum_k \hbar \omega_k b_k^\dagger b_k,$$

where \(\omega_k = c k\) are the phonon frequencies.
The system is conveniently described in the non-uniformly rotating frame defined by the unitary transformation $e^{iA(t)}$, where

$$A(t) = \left( \frac{\omega_0 t}{2} + \frac{1}{2} a^2 t^2 \right) \left( |X\rangle\langle X| + |Y\rangle\langle Y| + 2|B\rangle\langle B| \right).$$

The resulting Hamiltonian is $H = H'_X + H'_{\text{las}} + H'_{\text{int}} + H'_{\text{ph}}$, with

$$H'_X = e^{iA(t)} H_X e^{-iA(t)} - h \dot{A}(t)$$

$$= \hbar \Delta(t) \left( |X\rangle\langle X| + |Y\rangle\langle Y| \right) + (2\hbar \Delta(t) + E_B) |B\rangle\langle B|,$$

where $\Delta(t) = -at$, and

$$H'_{\text{las}} = e^{iA(t)} H_{\text{las}} e^{-iA(t)}$$

$$= \frac{\hbar \Omega(t)}{2} \left( |0\rangle\langle X| + |X\rangle\langle B| \right) \text{ h.c.}$$

### III. METHOD OF SIMULATION

To simplify the numerical simulation, we perform the unitary transformation defined by the operator

$$\mathcal{W} = |0\rangle\langle 0| + (|X\rangle\langle X| + |Y\rangle\langle Y|) W + |B\rangle\langle B| W^2,$$

where

$$W = \exp \left[ \sum_k \frac{g_k}{\omega_k} (b_k - b_k^\dagger) \right].$$

Upon this transformation and expanding to the leading order in the coupling constants, the Hamiltonian may be written as

$$\hat{H} = \mathcal{W} H \mathcal{W}^\dagger = H_1 + H_{\text{ph}} + V,$$

where $H_1 = H'_X + w H'_{\text{las}}$ and

$$V = \frac{\hbar \Omega(t)}{2} \left( |0\rangle\langle X| + |X\rangle\langle B| \right) W^\dagger + \text{h.c.}$$

Here

$$w = 1 - \frac{1}{2} \sum_k \left| \frac{g_k}{\omega_k} \right|^2$$

accounts for the phonon-induced renormalization of the pulse amplitude in the slow driving limit. We assume that the central frequency of the driving pulse is corrected for the phonon-induced energy shifts. The evolution of the reduced density matrix of the charge subsystem is then found by numerically solving the lowest-order time-convolutionless (TCL) evolution equation

$$\dot{\rho}(t) = -\int_0^t d\tau \text{Tr}_{\text{ph}} \left[ V(t), [V(\tau), \rho(t) \otimes \rho_{\text{ph}}] \right],$$

where $V(t)$ is the interaction Hamiltonian $V$ in the interaction picture with respect to $H_1 + H_{\text{ph}}$, $\rho_{\text{ph}}$ is the phonon density matrix at the thermal equilibrium, and $\text{Tr}_{\text{ph}}$ denotes the partial trace with respect to phonon states.

![FIG. 2:](image1)

**FIG. 2:** (Color online) The final occupation of the single-exciton state (color coded) as a function of the original pulse area $\Theta$ and the chirp $\alpha$ in the absence of coupling to phonons: (a) for $E_B = -1.0$ meV (b) for $E_B = -2.0$ meV.

![FIG. 3:](image2)

**FIG. 3:** (Color online) The final occupation of the single-exciton state (color coded) as a function of the original pulse area $\Theta$ and the chirp $\alpha$ with phonon effects included: (a,c,e) for $E_B = -2.0$ meV (b,d,f) for $E_B = -8.0$ meV.

### IV. RESULTS

With the simulation method outlined in Sec. III, we study the exciton dynamics focusing on the possibility of phonon-assisted transitions to the biexciton state, when the system is excited with a linearly polarized laser pulse. In our simulations, we assume $\rho = 5360$ kg/m$^3$, $c = 5110$ m/s, $D_e = 7$ eV and $D_h = -3.5$ eV. The electron wave functions are assumed to be Gaussians with the extension $l_e = 4.5$ nm in the $xy$ plane and $l_{e,z} = 1$ nm along the growth direction. In the case of holes, we take $l_h = 0.87l_e$ and $l_{h,z} = 0.87l_{e,z}$ respectively. The pulse length (before chirping) is taken to be $\tau_0 = 2$ ps.

First, we need to assess the degree of perturbation to the adiabatic evolution which stems from the additional level crossings with the biexciton (see Fig. 1) and is not
related to phonons. To this end, in Fig. 2, we show the single-exciton occupation without carrier-phonon coupling for $E_B = -1.0$ meV and $E_B = -2.0$ meV. The results show a reduction of the final occupation of the single exciton state at higher pulse areas for a very small biexciton shift ($E_B = -1.0$ meV) and positive chirps. We have found that under these conditions, a biexciton occupation on the order of 0.1 occurs (not shown). On the other hand, for the still small value of $E_B = -2.0$ meV, the evolution is nearly unperturbed by the presence of the biexciton state. Thus we conclude that for $|E_B| \gtrsim 2$ meV no direct population of the biexciton occurs with the pulses studied here, as the driving of the exciton to biexciton transition is too off-resonant.

In the next step, we include phonons and consider two cases: a) the biexciton state is shifted by a typical energy ($E_B = -2.0$ meV) below the single-exciton state, b) for comparison, the biexciton is shifted by a very large (and rarely realized) energy ($E_B = -8.0$ meV). In the first case, one expects that phonon-induced transitions to the biexciton state are efficient. In the second case, due to the large energy separation between the states (beyond the cut-off frequency of the carrier-phonon coupling), the biexciton generation is expected to be negligible and the system can effectively be treated as a two-level system. The simulated evolution of the single exciton occupation for both cases is shown in Fig. 3. The left panels (Fig. 3(a,c,e)) contain results for $E_B = -2.0$ meV, and the right panels (Fig. 3(b,d,f)) for $E_B = -8.0$ meV. The final occupation is calculated as a function of the pulse area $\Theta$ and the chirp rate $\alpha$ at three temperatures ($T = 1, 20, 40$ K).

Let us start the discussion for the case of the large biexciton shift $E_B = -8.0$ meV and low temperature $T = 1$ K shown in Fig. 3(b). As expected, the results from the two-level model are retrieved and a strong asymmetry in the final occupation with respect to the chirp is seen. In the two-level model, for negative chirp the evolution follows the upper branch and phonon emission leads to damping, while for positive chirps the lower branch is followed and since phonon absorption is almost absent at 1 K a full exciton occupation is reached. In contrast, in Fig. 3(a), for the small biexciton shift $E_B = -2.0$ meV, the influence of the biexciton state can be clearly seen. In particular, for positive chirps a decoherence at high pulse areas is visible due to the opening of the new decoherence channel consisting in transitions to the biexciton state (phonon-assisted biexciton generation). As can be seen in Fig. 1 for a negative biexciton shift (positive biexciton binding energy), this process corresponds to phonon emission no matter which adiabatic branch is followed, hence it is effective irrespective of the sign of the chirp even at low temperatures. Also for negative chirps the damping has become more effective. Thus, only for moderate pulse areas around $\Theta = 2\pi$ a high-fidelity preparation of the single exciton state is possible.

When considering higher temperatures, as seen in Fig. 3(c-f), phonon absorption becomes more likely and now also for positive chirps decoherence due to transitions between the main branches is stronger. Hence, considerable phonon-induced perturbation is seen even in the case of strongly detuned biexciton (Fig. 3(d,f)). In consequence, the additional effect of the biexciton shift is relatively weak.

For a more quantitative analysis we show the final occupation as a function of pulse area for different chirps $\alpha = -20.0, 20$ ps$^2$ in Fig. 4(a-d) for $E_B = -2.0$ meV (Fig. 4(a,b)) and for $E_B = -8.0$ meV (Fig. 4(c,d)). The
most remarkable difference for the two biexciton shifts is again seen for positive chirp $\alpha = 20 \, \text{ps}^2$ at $T = 1 \, \text{K}$. While in Fig. 4(a), for the small biexciton shift, a strong damping of the exciton occupation is found, in Fig. 4(c), for the large biexciton shift, an exciton occupation of one is reached for all pulse areas above the adiabatic threshold. Also the decoherence for the negative chirp $\alpha = -20 \, \text{ps}^2$ and damping of the Rabi oscillations at zero chirp are stronger due to the phonon-assisted biexciton generation. For higher temperature at $T = 40 \, \text{K}$ in Fig. 4(b,d) the difference between small and large biexciton shift is less pronounced.

The strong decoherence for the small biexciton shift $E_B = -2.0 \, \text{meV}$ is accompanied by an occupation of the biexciton state as shown in Fig. 4(e,f) confirming our previous statements.

Since in some experiments a photocurrent detection scheme is used, in which the detection signal is proportional to the total number of photocreated excitons, we have calculated also the total number of excitons in the system, $N_{\text{total}} = N_X + 2N_B$, where $N_X$ and $N_B$ are the occupation of the single- and biexciton state respectively. The results are shown in Fig. 5 for $E_B = -2 \, \text{meV}$ at $T = 1 \, \text{K}$ (Fig. 5(a)) and $T = 20 \, \text{K}$ (Fig. 5(b)). One can see that at a certain level of phonon-assisted biexciton generation the result exceeds 1, which has no sense if interpreted in terms of the two-level model. Moreover, it is clear that even below this limit, the biexciton contribution leads to an overestimated occupation of the single exciton state.

V. CONCLUSIONS

In summary, we have studied the open system evolution of a QD adiabatically controlled by a chirped linearly polarized laser pulse. We have shown that the presence of a biexciton state, which becomes optically coupled for linearly polarized laser pulses, opens new decoherence paths related to phonon-assisted biexciton generation. If the biexciton state is shifted down by 1 or 2 meV it becomes accessible via phonon emission processes and the resulting decoherence is strong even at low temperatures. This reduces the achieved occupation of the single exciton state in the parameters range where the chirped pulse control was very efficient in a model without the biexciton state. Since the phonon-assisted biexciton generation increases with the pulse area moderate pulse areas are optimal for the state preparation. On the other hand, if the biexciton shift is larger, the two-level evolution is essentially recovered.

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