Proposed Robust and High-Fidelity Preparation of Excitons and Biexcitons in Semiconductor Quantum Dots Making Active Use of Phonons

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It is demonstrated how the exciton and the biexciton state of a quantum dot can be prepared with high fidelity on a picosecond time scale by driving the dot with a strong laser pulse that is tuned above the exciton resonance for exciton preparation and in resonance with the exciton transition for biexciton preparation. The proposed protocols make use of the phonon-induced relaxation towards photon dressed states in optically driven quantum dots and combine the simplicity of traditional Rabi oscillation schemes with the robustness of adiabatic rapid passage schemes. Our protocols allow for an on-demand, fast, and almost perfect state preparation even at strong carrier-phonon interaction where other schemes fail. In fact, the performance of the presented protocols is shown to be better the stronger the carrier-phonon interaction is.

Realizing a high-quality, robust, on-demand, and fast exciton or biexciton preparation in semiconductor quantum dots (QDs) is of great importance for many promising QD-based devices such as single [1,2] or entangled [3–6] photon sources that are crucial for various applications in the field of quantum information processing [7,8] as well as for tests of fundamental aspects of quantum mechanics [9]. While a perfect initiation of the QD in both states can in principle be realized via Rabi oscillations [10,11], these simple schemes suffer from a high sensitivity on the dipole moments and the pulse intensity. A preparation that is robust against fluctuations in the coupling strengths can be achieved using protocols with frequency swept pulses that rely on adiabatic rapid passage (ARP) [12–14]. However, the degree of exciton inversion realized in ARP-based experiments [13,14] stayed considerably below the ideal case and most recent theoretical works gave compelling evidence that this reduction can be attributed to acoustic phonon coupling [15,16] that is also known to strongly limit the fidelity of traditional Rabi oscillation schemes [17–19].

In this Letter, we shall present protocols that combine the simplicity of Rabi oscillation schemes with the robustness of ARP schemes and give the discussion of phonon influences a completely different perspective by demonstrating how one can highly benefit from the otherwise undesired carrier-phonon coupling. To be specific, we propose protocols allowing for a high-fidelity and robust phonon-assisted preparation of the exciton as well as the biexciton state in strongly confined QDs that rely on exciting the system with an off-resonant intense pulse. Our schemes make active use of the acoustic phonon coupling by exploiting the characteristics of the stationary nonequilibrium state towards which the QD is driven due to the system-environment interaction and are shown to perform better the stronger the carrier phonon coupling is.

Let us first concentrate on preparing the single exciton state. To this end, we assume a circularly polarized excitation, which allows us to model the laser-driven QD as an electronic two-level system that consists of the ground state $|G\rangle$ without electron-hole pairs and the single exciton state $|X\rangle$ described by the Hamiltonian

$$H_{\mathrm{QD},L} = \hbar\omega_X|X\rangle\langle X| - \left[\frac{\hbar f(t)}{2} e^{-i\omega_L t}|X\rangle\langle G| + \mathrm{H.c.}\right], \quad (1)$$

where ω_L is the laser frequency and $f(t)$ denotes the instantaneous Rabi frequency that for a Gaussian pulse is given by $f(t) = \alpha/(\sqrt{2\pi}\tau_0)\exp(-t^2/2\tau_0^2)$, where α is the pulse area and τ_0 defines the pulse length. The pure dephasing carrier-phonon coupling is modeled by the Hamiltonian

$$H_{\mathrm{QD,ph}} = \sum_{\mathbf{q}}\hbar\omega_{\mathbf{q}}b_{\mathbf{q}}^{\dagger}b_{\mathbf{q}} + \sum_{\mathbf{q},\nu}\hbar n_{\nu}(g_{\mathbf{q}}b_{\mathbf{q}} + g_{\mathbf{q}}^*b_{\mathbf{q}}^{\dagger})|\nu\rangle\langle\nu|,$$

$$(2)$$

where ν labels the electronic states, n_{ν} counts the excitons present in the state $|\nu\rangle$, $b_{\mathbf{q}}^{\dagger}$ creates a longitudinal acoustic (LA) bulk phonon with wave vector $\mathbf{q}$ and energy $\hbar\omega_{\mathbf{q}}$, and $g_{\mathbf{q}}$ denote the exciton-phonon coupling constants. We concentrate on the deformation potential coupling to LA phonons that is dominant for GaAs QDs [19,20], we choose a radial electron confinement length of 3 nm and use the same coupling parameters as in Ref. [21] that have been shown to nicely reproduce experimental results [21]. To calculate the coupled carrier-phonon dynamics, we shall apply a numerically exact real-time path-integral approach that allows us to study the system evolution without invoking any approximations to the model given above. Details of this method can be found in Ref. [22].

The phonon coupling strongly affects the QD dynamics. Most important for our present discussion, it pushes the laser driven electronic system towards a stationary

nonequilibrium state. We demonstrated in Ref. [23] that, for a two-level system with weak carrier-phonon coupling as realized in GaAs QDs and a constant optical driving with $f(t) = $ const, this stationary state can be well approximated by a thermal distribution over the eigenstates of $H_{QD,L}$, often referred to as photon dressed states, leading to a stationary exciton occupation of

$$C_X(t = \infty) = \frac{1}{2}\left[1 + \frac{\Delta}{\hbar\Omega} \tanh\left(\frac{\hbar\Omega}{2k_BT}\right)\right], \quad (3)$$

where $\Omega = \sqrt{f^2 + (\Delta/\hbar)^2}$ and $\Delta = \hbar(\omega_L - \omega_X)$ is the detuning between the laser and the QD transition. Obviously, Eq. (3) predicts a stationary inversion for $\Delta/(\hbar\Omega) \approx 1$ and $\hbar\Omega/2kT \gg 1$. Indeed, the exact stationary exciton occupation as calculated by using the path-integral approach and shown in Fig. 1(a) as a function of Δ and T is almost perfectly described by Eq. (3): while for redshifted excitation with $\Delta < 0$ the stationary occupation is below $1/2$, an almost perfect stationary inversion is found at low T for a blueshifted laser, similar to what was previously predicted for a two-level system consisting of two tunneling-coupled QDs within a Born-Markov approximation [24]. However, for the system considered here, several comments are in order. First, the time needed to reach the stationary state strongly depends on the excitation conditions and can be up to several hundreds of picoseconds at small f, low T, and large Δ. On these time scales other relaxation processes like radiative decay, that are not included in our model, gain in importance and reduce the accessible degree of inversion [25]. Further, cw excitations as assumed in Fig. 1 and Eq. (3) are of no use when an on-demand inversion is the target. Instead, pulsed excitations are needed to complete a preparation at a given time. Therefore, to evaluate whether the stationary inversion as shown in Fig. 1(a) has relevance for state
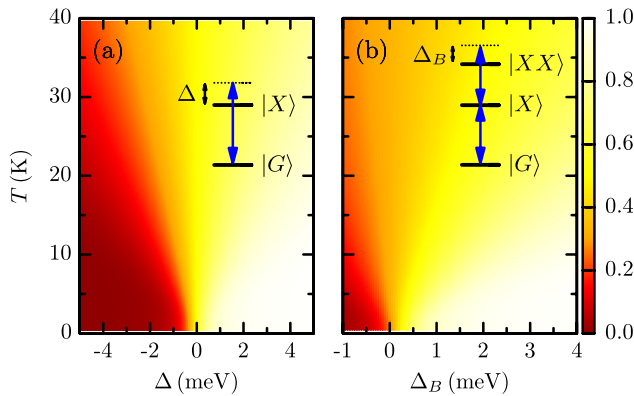
preparation, several questions have to be answered. For example, which degree of inversion can be reached for off-resonant pulses of finite length? What determines the optimal detuning and how sensitive is the achieved inversion against variations in the pulse intensity? In this Letter, we will demonstrate that by applying an off-resonant intense pulse at low temperatures it is not only possible to achieve some inversion (as achieved in recent experiments with moderate pulse intensities [26]) but an almost perfect state preparation on a time scale of several picoseconds.

Shown in Fig. 2 is the exciton occupation C_X after a Gaussian pulse of $\tau = 2\sqrt{2\ln2}\tau_0 = 15$ ps FWHM as a function of the pulse area α for different detunings Δ at $T = 4$ K. While for $\Delta = 0$ the occupation performs damped Rabi oscillations around a mean value of $1/2$, the results do already considerably change for a slightly blueshifted excitation with $\Delta = 0.2$ meV: not only does the oscillation amplitude decrease (as it is well known for off-resonant driving [27]), but at high pulse areas, where the phonon damping is stronger [17,18] and the system is efficiently pushed towards its stationary state, significantly higher occupations are reached with C_X taking values of roughly 0.9. Even higher occupations are realized for larger Δ. For $\Delta = 1.0$ meV, which turns out to be the optimal choice for the detuning, an almost ideal and robust exciton preparation is realized at high pulse areas. We would like to stress that an almost equally high inversion is found for a rather wide range of detunings, as shown in Fig. 3(a), where C_X is plotted as a function of Δ for a fixed pulse area of $\alpha = 20\pi$. Thus, the phonon-assisted state preparation as realized by exciting the system off-resonantly by a single intense pulse is not only of high fidelity but is also robust against variations in the pulse intensity or the detuning provided that the pulse is strong enough.

It should be noted that, although the stationary occupation rises monotonically with rising detuning, cf. Fig. 1, for
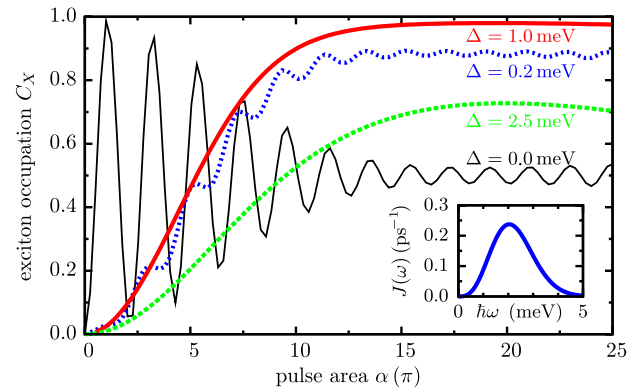

FIG. 1 (color online). Stationary (a) exciton and (b) biexciton occupation that is reached under constant optical driving with $f = 1.0$ ps^{-1} as a function of (a) the temperature and the detuning Δ between the laser frequency and the QD transition frequency and (b) the temperature and the biexciton binding energy Δ_B. Inset: Sketch of the excitation schemes (see text).

FIG. 2 (color online). Final exciton occupation C_X after a Gaussian pulse of 15 ps FWHM at $T = 4$ K as a function of the applied pulse area α for different detunings Δ as indicated. Inset: Phonon spectral density for a spherical GaAS QD with a radial electron confinement length of 3 nm.
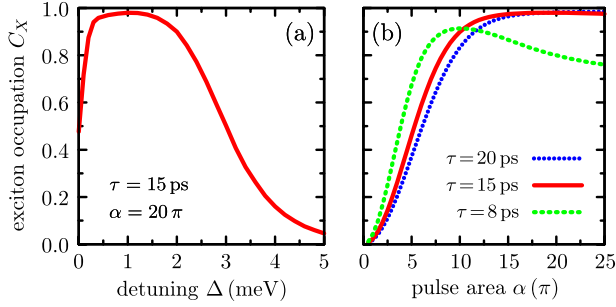
FIG. 3 (color online). Final exciton occupation C_X after a Gaussian pulse with pulse length τ (FWHM) at $T = 4$ K. (a) C_X as a function of Δ for $\tau = 15$ ps and $\alpha = 20\pi$. (b) C_X as a function of α for $\Delta = 1$ meV and different τ as indicated.

large Δ, the degree of inversion achieved for a finite pulse no longer increases with Δ, but eventually decreases again as illustrated in Figs. 2 and 3(a). To understand this non-monotonic dependence of the protocol efficiency on Δ, two things must be borne in mind: first, the phonon coupling, that drives the system towards the stationary state, exhibits a spectral cutoff, i.e., the phonon spectral density $J(\omega) = \sum_{\mathbf{q}} |g_{\mathbf{q}}|^2 \delta(\omega - \omega_{\mathbf{q}})$, that is shown in the inset of Fig. 2, vanishes for large ω, and second, the strength of the phonon-induced relaxation can be approximately described by $J(\Omega)$ [19,28]. As soon as the Rabi frequency Ω exceeds the frequency $\omega_{\max}$, where $J(\omega)$ is maximal, the phonon coupling becomes less efficient and the inversion that is reached for finite pulses is reduced. Further, as $\Omega = \sqrt{f^2 + (\Delta/\hbar)^2}$, the optimal detuning is less than $\hbar\omega_{\max}$, as is clearly seen from Figs. 2 and 3(a).

Obviously, the proposed scheme becomes less efficient when the pulse is too short and the system is not driven close enough to its stationary state. This is illustrated in Fig. 3(b). While pulses longer than those considered so far do not affect the efficiency, for a pulse length below 10 ps, the achieved inversion drops and the protocol is less robust with respect to variations in the pulse intensity. However, we would like to stress that the pulse length of 10–15 ps that is needed to guarantee a stable phonon-assisted preparation via off-resonant driving is very much the same as the one that was needed in recent ARP-based experiments to ensure an adiabatic evolution [13,14].

Unique to the present protocol is that its performance becomes better when the strength of the carrier-phonon coupling is increased, thus allowing for an almost ideal state preparation in the regime of strong system-environment interaction, where traditional Rabi oscillation or ARP schemes are known to fail [15,17–19]. Shown in Fig. 4 is the exciton occupation after a 15 ps lasting pulse (FWHM) as a function of Δ and α for the carrier-phonon coupling of GaAs as studied so far [Fig. 4(a)] and for a situation, where we have increased $|g_{\mathbf{q}}|^2$ by a factor of 3 by hand [Fig. 4(b)] in order to roughly simulate the coupling strength of materials like GaN [29]. Clearly, the efficiency
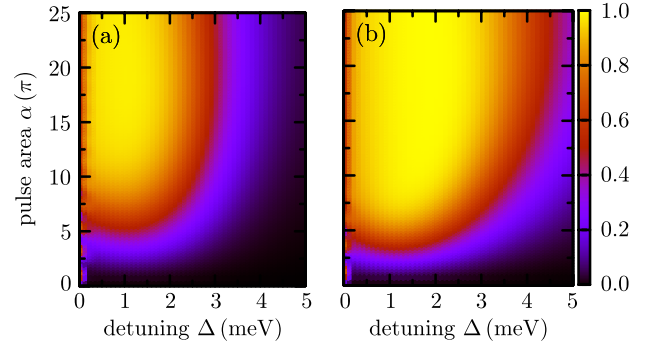

FIG. 4 (color online). Final exciton occupation C_X after a Gaussian pulse of 15 ps FWHM at $T = 4$ K as a function of the detuning Δ and the pulse area α for (a) GaAs parameters and (b) a phonon coupling where $|g_{\mathbf{q}}|^2$ has been increased by a factor of 3.

increases with rising coupling: the maximal inversion is even closer to 1 and the range of pulse intensities and detunings for which high occupations are reached extends considerably. We stress that carrying out calculations for strong phonon couplings is a challenge and that even highly elaborate approximate methods such as a fourth-order correlation expansion are known to break down in this regime [23]. Here, the performance of reliable simulations is possible by using a numerically exact path-integral approach [22] that accounts fully for all non-Markovian effects and arbitrary multiphonon processes.

In light of recent progress in realizing QD-based entangled photon sources [6,30], the task of preparing the biexciton state $|XX\rangle$ has also become of topical interest. Whether or not the ideas so far developed for the preparation of the single exciton state can be transferred to this task is not obvious, because now we have to deal with more electronic levels, and thus a perfect preparation of the biexciton requires that not only the ground state but also the single exciton state is completely depopulated. Furthermore, there is not only a single resonance. In particular, the biexciton state can be optically excited either by a two-photon process [11] that is resonant when $2\hbar\omega = 2\hbar\omega_X - \Delta_B$, where Δ_B is the biexciton binding energy, or by a sequential process where first the single exciton is excited and then a transition from the exciton to the biexciton is induced. The resonance for the latter process at $\hbar\omega = \hbar\omega_X - \Delta_B$ is known to be a dephasing-induced resonance, which in $\chi^{(3)}$ signals shows up only due to the coupling of the electronic system to a bath [31].

To couple the biexciton we switch the polarization of the exciting pulse from circular to linear. Then, the light-matter Hamiltonian reads as [32]

$$H_{\mathrm{QD},L} = \hbar\omega_X|X\rangle\langle X| + (2\hbar\omega_X - \Delta_B)|XX\rangle\langle XX|$$
$$- \left[\frac{\hbar f(t)}{2} e^{-i\omega_L t}(|X\rangle\langle G| + |XX\rangle\langle X|) + \mathrm{H.c.}\right]. \quad (4)$$

In general, Δ_B can be positive as well as negative, but for self-assembled GaAs QDs it typically takes values ranging from 1 to 3 meV [19,33,34]. The carrier-phonon coupling is the same as in Eq. (2), with the only difference that now the sum over $|\nu\rangle$ runs over three electronic states, $|G\rangle$, $|X\rangle$, and $|XX\rangle$.

In the remainder of this Letter we shall demonstrate that an almost perfect phonon-assisted biexciton preparation is possible by choosing an excitation that is resonant to the ground state to exciton transition provided that the exciton to biexciton transition is redshifted, which typically is the case. This situation is schematically sketched in the inset of Fig. 1(b).

Again, it turns out to be instructive to look first at the stationary state, that for this excitation is reached due to the acoustic phonon coupling. Figure 1(b) shows the stationary biexciton occupation as calculated within the path-integral approach for constant driving as a function of temperature and biexciton binding energy: very high values of C_{XX} are found for low T and positive Δ_B, indicating that a phonon-mediated preparation of the biexciton should be possible for the typical case where the biexciton binding energy shifts the biexciton down in energy. This supposition is indeed confirmed by the results shown in Fig. 5, where the biexciton occupation that is reached after a Gaussian pulse with 15 ps FWHM is plotted as a function of the pulse area and Δ_B at $T = 4$ K: an almost perfect biexciton preparation is realized for a wide range of biexciton binding energies which is robust against variations in the pulse intensity provided that the pulse is strong enough. It should be noted that an occupation probability for $|XX\rangle$ near 1 implies that, due to the action of the phonons, the exciton state as well as the ground state are practically unoccupied although the ground state to exciton transition is resonantly driven. As discussed before in detail for the case of the
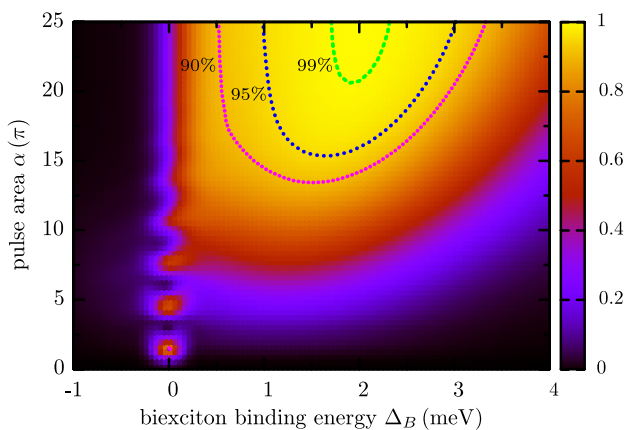
single exciton preparation, the performance of the scheme becomes worse when the pulse is chosen too short, but improves with rising strength of the carrier-phonon coupling (not shown).

In summary, we have presented schemes allowing for an on-demand, high-fidelity, and robust preparation of the single exciton as well as the biexciton state in semiconductor QDs that are based on off-resonant excitations with strong optical pulses and make active use of the carrier-phonon coupling. In particular, we predict that the biexciton can be prepared by resonantly driving the ground state to exciton transition, implying that the exciton state will be unoccupied despite its resonant excitation. The proposed protocols allow for fast state preparations on the time scale of 10 ps. We expect our findings to inspire future experimental research as the regime of off-resonant driving at high intensities is so far almost unexplored and we believe that the proposed phonon-assisted state preparation schemes can pave the way to more efficient sources for single or entangled photons. Importantly, the presented protocols not only combine the simplicity of traditional Rabi oscillation schemes (without the need of realizing chirped laser pulses) with the robustness of ARP-based schemes, but they perform better the stronger the carrier phonon coupling is, thus allowing for an ideal state preparation even in situations with strong system-environment interaction that are usually thought of as making control protocols impossible.

FIG. 5 (color online). Final biexciton occupation C_{XX} after a Gaussian pulse with a FWHM of 15 ps at $T = 4$ K as a function of the biexciton binding energy Δ_B and the pulse area α. The laser frequency is chosen in resonance with the ground state to exciton transition. The contour lines display where certain values of C_{XX} are reached.

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