Phonon-Assisted Damping of Rabi Oscillations in Semiconductor Quantum Dots

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Electron-phonon interaction is a major source of optical dephasing in semiconductor quantum dots. Within a density matrix theory the electron-phonon interaction is considered up to the second order of a correlation expansion, allowing the calculation of the quantum kinetic dephasing dynamics of optically induced nonlinearities in GaAs quantum dots for arbitrary pulse strengths and shapes. We find Rabi oscillations renormalized and a damping that depends on the input pulse strength, a behavior not known from exponential dephasing mechanisms.

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Coherence and decoherence are central research topics in atomic [1,2] and solid state physics [3,4]. Especially the generation of a coherent superposition of quantum mechanical states using ultrashort laser pulses and the subsequent decoherence due to interaction with the environment is a fundamental method to study open quantum mechanical systems. In this respect, semiconductor quantum dots [5,6], as so-called artificial atoms, have gained increasing interest. They provide design capabilities and the prospect of future applications in quantum information processing and novel laser devices [7–14]. While there are many similarities between quantum dots and atomic systems, such as the typical level structure which results from three-dimensional confinement of electrons, there are also important differences: While atoms are mostly treated as isolated systems, only influenced by the radiation field [1,2], semiconductor quantum dots cannot be separated from their surrounding solid state matrix. Consequently, coupling of electrons to phonons plays a major role for those systems; in particular, it provides a dephasing mechanism for optically induced coherence on times scales (few picoseconds [11,15]) much shorter than for radiative interaction (several hundred picoseconds [2,6]).

Many dephasing processes are covered by a general theory [2] which is based on Markovian, i.e., strictly energy conserving, coupling of electrons to a so-called bath, which acts as an energy reservoir and is assumed to be unaffected by the coupling. Such scattering processes of electrons with quasiparticles from the bath lead to simultaneous energy and phase relaxation (optical dephasing). As a consequence, the dephasing time (T_2) is given by twice the energy relaxation time (T_1). While photons (radiation damping) and optical phonons are typical examples which can bridge the (sub-)band gap in semiconductors of higher dimensionality, acoustic phonons are mainly found in intraband scattering. In quantum dots, strictly energy conserving processes are suppressed due to the discrete energy level structure. Nevertheless, quantum kinetic processes [3], relying on energy-time uncertainty, lead to so-called pure dephasing of the quantum dot polarization, not assisted by energy relaxation [15,16]. In the linear optical regime and for two-level systems, the pure dephasing processes can be theoretically investigated by applying the independent Boson model (BM) [17] to the electron-phonon interaction. According to recent experiments [9,11,12] as well as evaluations of the BM [15,16,18], linear spectra exhibit some common signatures: a Lorentzian zero-phonon line surrounded by broad acoustic phonon sidebands. While there has been rapid progress concerning the experimental study of nonlinear coherent optical processes in quantum dots [10,12,13,19], there is to the best of our knowledge no theoretical work available to predict the dephasing dynamics induced by acoustic phonons during excitation with nonlinear optical pulses of arbitrary shape and strength, necessary for describing effects such as damping of Rabi oscillations (for related work on LO phonons see Refs. [20–23], excitation with δ pulses is considered in Ref. [24]). The microscopic understanding of these dephasing mechanisms is of central importance for nonlinear applications in quantum computing and quantum dot lasers.

In this Letter we provide a theoretical description of the damping of optically induced Rabi oscillations in semiconductor quantum dots. As the major dephasing mechanism we consider, in correspondence with recent experiments [11], the coupling of the electronic coherence (optical polarization) to acoustic phonons. The equations of motion are derived within a correlation expansion for the density matrix and all electron-phonon correlations including up to two-phonon contributions are systematically taken into account. In the range of linear optics, the quality of the truncation procedure can be tested by comparing the numerical results to the (to all orders) exactly solvable BM [15,16,18]. We obtain satisfactory agreement between both calculations for the considered GaAs quantum dots [6]. Having this independent test of our theory, we study damping of Rabi oscillations for nonlinear optical excitation with pulses of varying strength and duration.

Our model system is described by the Hamiltonian $H = H_0 + H^{\rm int}_{\rm el\text{-}ph} + H^{\rm int}_{\rm el\text{-}e.m.}$. The free kinetics of Bloch electrons and phonons reads as follows:

$$H_0 = \sum_i \epsilon_i a_i^+ a_i + \sum_{\mu q} \hbar\omega_q b_q^+ b_q, \qquad (1)$$

where we introduced the electronic state energy ϵ_i of electrons in level i, the dispersion ω_q of phonons with wave number q, and the electron (phonon) annihilation a_i (b_q) and creation a_i^+ (b_q^+) operators. The electron-phonon coupling is given by

$$H^{\rm int}_{\rm el\text{-}ph} = \sum_{i,i,q} g^q_{ii} a_i^+ a_i (b_q + b^+_{-q}), \qquad (2)$$

with coupling elements g^q_{ij} [25]. Finally, the dipole interaction with the classically treated optical field read as

$$H^{\rm int}_{\rm el\text{-}e.m.} = -\sum_{i,j} M_{ij} E(t) a_i^+ a_j. \qquad (3)$$

The electromagnetic field $E(t)$ acts through the quantum dot dipole transition element M_{ij}. In this work, Gaussian pulses with $\tilde{E}(t) = E_0 \exp[-(t/\tau)^2]$ are used.

For small quantum dots, dominated by the confinement energy, excitonic effects can be neglected [16]. For simplicity and to compare our results to the BM, we restrict our calculations to a two-level model ($i, j = v, c$) and a single electron-hole pair. An extension to a multilevel system is straightforward. We consider a deformation potential coupling via longitudinal acoustic (LA) bulk phonons described within bath approximation and assume a thermal Bose distribution. Test calculations showed that the influence of both optical phonons and excitation-generated hot and coherent acoustic phonons on the polarization and occupation dynamics are negligible for the considered parameters and quantum dot densities (for strong coupling compare Ref. [23]). Therefore they are not considered in the following. Phonon-phonon interaction, even if it may strongly affect the phonon system at higher temperatures [16,18], is still not understood microscopically and thus not discussed here.

Within a second order correlation expansion, quantum kinetic equations of motions for the coherences (polarization), the occupation probabilities, and phonon-assisted quantities can be derived. The full set of equations is given in Ref. [26]. It can be evaluated using standard numerical techniques and from this the macroscopic polarization $P(t) = n_D M_{cv} \langle a_v^+ a_c \rangle$ + c.c. of a homogenous ensemble of quantum dots (with density n_D) can be obtained. For linear excitation the spectral absorption function $\alpha(\omega) \propto {\rm Im}[P(\omega)/E(\omega)]$ can then be calculated using Fourier transformation. In the linear regime the exactly solvable BM which embraces the whole hierarchy of electron-phonon interactions provides an analytical result [15,16,18,26]. Figure 1 shows a comparison of the exact absorption spectrum (solid curve) with numerically obtained results, where correlations with one (dashed curve) and up to two phonons (dotted curve) are included. The parameters are chosen for GaAs dots with parabolic confinement potential [25]. In all cases, a narrow Lorentzian zero-phonon line (ZPL), which is broadened by a radiative damping of 500 ps [2], can be seen at the band gap energy reduced by the small polaron self-energy Δ. The ZPL is surrounded by broad sidebands which occur due to the dispersion of the involved acoustic phonons. Note that the ZPL peak is truncated here and would have a maximum value of about 100 on the given scale. Comparison of the exact model with the correlation expansion shows clear deviation in the vicinity of the ZPL, if only one-phonon correlations are considered. Inclusion of two-phonon processes smooths the structure toward the exact result by allowing a broader range of wave number combinations for lower energies. All in all, adequate agreement is obtained if correlations of electrons with up to two phonons are included. It is expected that higher correlations introduce only minor corrections because (i) the weak coupling in the considered GaAs system leads to convergence already in second order (in the linear and nonlinear regime), (ii) there is good agreement with the exact linear results, (iii) we get results for excitation with very short pulses which are in agreement with analytical calculations for δ-pulse excitation [24].

The inset of Fig. 1 shows the corresponding time-resolved polarization decay of the two-phonon model for linear excitation. After a rapid initial dephasing (corresponding to the broad spectral sidebands) which leaves

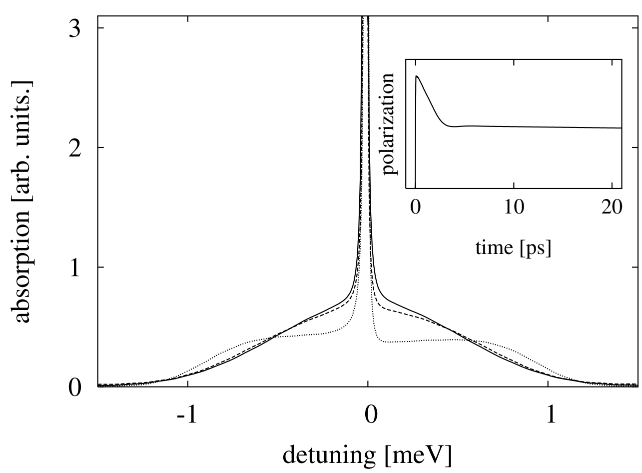

FIG. 1. Linear absorption spectrum of a homogeneous quantum dot ensemble ($T = 77$ K). Compared are calculations which include scattering events of electrons with one (dotted curve), up to two (dashed curve), and an arbitrary number (solid curve) of acoustic phonons. The differences in the broad phonon sidebands, which surround the narrow ZPL (truncated), show that inclusion of two-phonon correlations is important. The inset displays the temporal polarization dynamics for linear excitation with a Gaussian $\tau = 25$ fs pulse.

a certain amount of coherence, the system exhibits a slow exponential decay. The fast initial dephasing can be explained as destructive interference effect among the optically excited phonon-assisted virtual states, which are oscillating within a range of frequencies allowed by the phonon dispersion and multiple phonon scattering. The slow exponential decay on long time scales is given by radiative damping of 500 ps and leads to the spectrally narrow ZPL.

Next, the optical dephasing under nonlinear excitation conditions will be discussed. Rabi flopping, which is a fundamental effect in coherent nonlinear light-matter interaction and the key to coherent control in quantum computation, corresponds to optically induced coherent transfer of density from the ground state to the excited state and vice versa during excitation and thereby leads to oscillations of the upper state occupation [1,2]. Figure 2 shows the upper state occupation dynamics for band gap resonant excitation with a 2 ps Gaussian pulse for different pulse areas (from Fig. 1 a typical decay time scale of the polarization of about 4 ps can be extracted). Weak excitation (0.2π, dashed curve) leads to only a small transfer of electrons into the upper state. While in an undamped system (inset) the upper state occupation would oscillate from 0 to 1 and then back to 0 for a 2π pulse (solid curve), the considered system does not reach full inversion and is not left in the ground state due to the polarization damping by electron-phonon scattering. For a 4π pulse (dotted curve) the system exhibits clear but damped Rabi flopping.

Next we focus on the character of the phonon-induced Rabi oscillation damping in quantum dots for different pulse durations. In Fig. 3(a) we compare the upper state occupation dynamics of three systems with different dephasing mechanisms: (1) the discussed quantum dot system which is dominated by electron-phonon induced pure dephasing and does not exhibit energy relaxation (solid curves), (2) an artificial two-level model system with exponential pure dephasing of $T_2 = 4$ ps and no energy relaxation ($T_1 = 0$, dashed curves), and (3) an undamped two-level system. Figure 3(a) shows that for short pulses ($\tau = 0.5$ ps) the Rabi oscillations are mainly unaffected by the damping processes. For $\tau = 5$ ps which is in the order of the decay times, the two damped systems exhibit similarly reduced but still clear Rabi oscillations. The pulse with a duration of $\tau = 20$ ps, being much longer than the typical decay time, reveals the fundamental differences between the systems: With exponential pure dephasing there still is half-inversion but almost no oscillations occur. In contrast to this, the considered phonon-damped quantum dot system does not show such a complete suppression of Rabi oscillations. This is due to the dephasing induced by the quantum kinetic electron-phonon coupling, which acts only on relatively short time scales, as already seen in the partial suppression of the coherence in the inset of Fig. 1. Additionally, the occupation dynamics does not show the full two flops that one would expect for excitation with a 4π pulse. Instead, the system is only driven into a partially inverted state, which would be expected for a 3π excitation.

While part of these results may be expected from the linear spectra, the dependence of Rabi flopping on the pulse intensity shows a qualitative unexpected behavior. Figure 3(b), shows the upper state occupation after pulse excitation as a function of the input pulse area. This information is accessible in ultrashort optical experiments [12,13,19,27]. As before, the Rabi oscillations are almost unaltered for an excitation with a short 500 fs pulse. For $\tau = 5$ ps the exponential pure dephasing leads to a uniform suppression of the area-dependent oscillation amplitude and converges toward the incoherent limit, i.e., complete suppression of the oscillations, for longer pulses. In contrast to this, for phonon-induced damping the first maximum is still very pronounced and only the subsequent amplitudes are reduced. This specific behavior may contribute to the understanding of an overshoot during the first Rabi flop seen in recent experiments, for instance Ref. [27]. Additionally, both damping mechanisms exhibit a strong (up to 1π) renormalization of the pulse areas at which maxima and minima occur.

All in all, the presented phonon-induced pure dephasing exhibits novel features such as reduced damping and renormalization of Rabi oscillations in GaAs quantum dots. For pulse durations on the time scale below the phonon-induced dephasing, a pure single exponential approximation can be applied. This is not possible for longer pulses, where the full quantum kinetic dynamics must be included. This applies as well for the description of the transition from weak to very large pulse areas [Fig. 3(b)]. For higher temperatures and materials with stronger electron-phonon coupling, inclusion

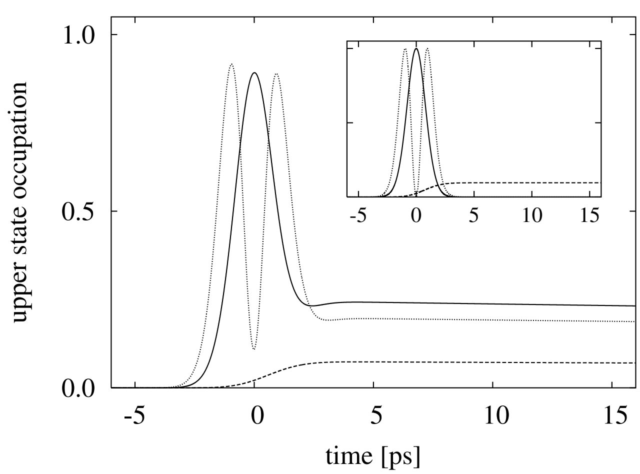

FIG. 2. Upper state occupation dynamics for increasing pulse area (strength): 0.2π (dashed curve), 2π (solid curve), 4π (dotted curve). Rabi flopping (for 2π single, 4π double cycle) occurs but is damped due to electron-phonon interaction. Inset: two-level system without damping.

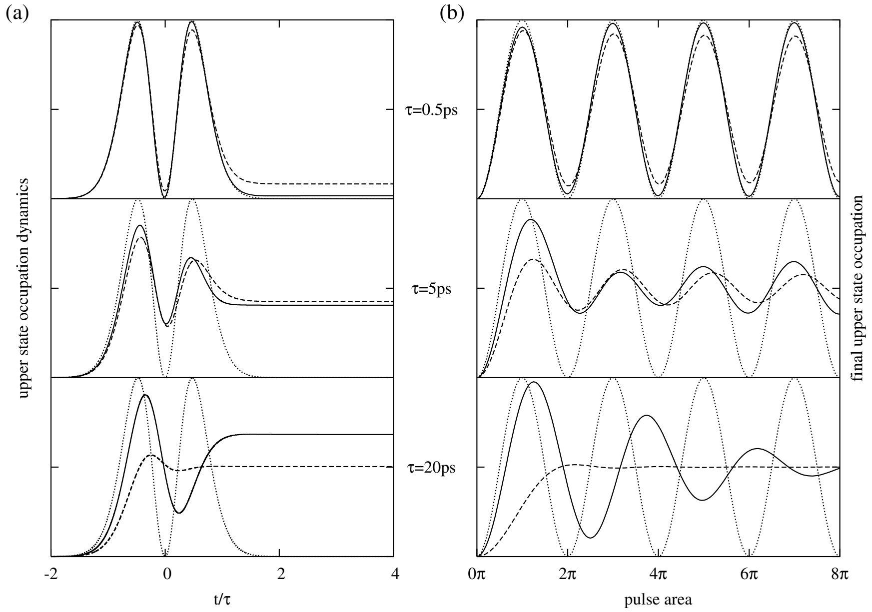

FIG. 3. Comparison of the upper state occupation dynamics for increasing pulse lengths, in systems damped by acoustic phonons (solid curves), an exponential pure dephasing of $T_2 = 4$ ps (dashed curves), and an undamped two-level system (dotted curves). (a) Occupation dynamics for excitation with a 4π pulse. (b) Occupation after pulse excitation as a function of the input pulse area. Scale of the vertical axes is each from 0 to 1. See text for details.

of the complete phonon hierarchy and a microscopic theory for the ZPL width would be desirable.

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