Resonant nature of phonon-induced damping of Rabi oscillations in quantum dots

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Optically controlled coherent dynamics of charge (excitonic) degrees of freedom in a semiconductor quantum dot under the influence of lattice dynamics (phonons) is discussed theoretically. We show that the dynamics of the lattice response in the strongly nonlinear regime is governed by a semiclassical resonance between the phonon modes and the optically driven dynamics. We stress on the importance of the stability of intermediate states for the truly coherent control.

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One of the most challenging proposals exploiting the atomiclike properties of quantum dots (QD's)[1,2] is to implement the quantum information processing schemes[3,4] using confined carrier states in these artificial systems. One of the proposed solutions is to use the charge degrees of freedom which may be efficiently controlled optically even on subpicosecond scale[5–8] which may look promising in view of the measured 1 ns lifetime of excitons in QD's.[9,10] The first steps towards this goal include experimental proofs of controlled coherent dynamics in these structures,[11] observation of Rabi oscillations,[12–16] demonstration of entanglement between states of interacting dots,[17] and performing a quantum logic gate based on a biexciton system in a QD.[18]

However, the coherent dynamics of quantum confined carrier states is very sensitive to interaction with the macroscopic number of degrees of freedom of the outside world. If the coherent dynamics involves an excited state of the confined exciton[11,13,14] the coherence time is limited by relaxation to the ground state (usually ~ 30 ps). The measured signal may further be affected by the particularities of the detection technique.[14] Another approach consists in measuring the dot occupation upon driving by pulses with constant length and varying amplitudes.[12,15,16] So far, it has always turned out that such pulse-area-dependent Rabi oscillations deviate from the ideal ones, the discrepancy being larger for stronger pulses. In principle, this might also be explained by experimental conditions or environmental perturbation: scattering by weakly localized excitons around an interface fluctuation QD (further confirmed by increasing decay for stronger pulses),[12] tunneling to leads in the photodiode structure (on ~ 10 ps time scale)[15], or dipole moment distribution in the QD ensemble.[9]

One might believe that all the perturbation comes from sources that may be removed or minimized by technology improvement and by optimizing the experimental conditions and hence produce no fundamental obstacle to arbitrarily perfect quantum control over the excitonic states. However, in every case the QD's are inherently coupled to the surrounding crystal lattice. The perturbing effect of lattice modes (phonons) has been observed experimentally as a fast (~ 1 ps) partial decay of optical polarization induced by an ultrafast laser pulse.[9] The theoretical analysis[19–21] shows that the experimentally observed effects may be quantitatively accounted for by invoking the carrier-phonon coupling. Indeed, under suitable excitation conditions in a high-quality sample the carrier density may be kept low enough to eliminate Coulomb scattering effects while the coupling to the electromagnetic modes is manifested by the exciton radiative decay on time scales many orders of magnitude longer than those relevant here. The theory shows that the coherence decay should be viewed as a trace of coherent lattice dynamics (due to lattice inertia), persisting even at zero temperature when the lattice is initially at its ground state and cannot perturb the stationary ground state of the carrier subsystem. Due to strong reservoir memory on time scales relevant for these processes, they cannot be fully understood within the Markovian approximations. In particular, the idea of a "decoherence time," with which the control dynamics competes, is misleading.[22]

The recent theoretical study[23] on optical Rabi flopping of excitons in QD's driven by finite-length optical pulses shows that exponential damping models fail to correctly describe the system kinetics. The appropriate description yields much less damping, especially for long pulses. It turns out that the lowest "quality" of the Rabi oscillation is obtained for pulse durations of a few picoseconds, while for longer durations the damping is again decreased.

In this paper we propose both qualitative and quantitative explanation of the mechanism leading to the phonon-induced damping reported in the theoretical and experimental studies. We show that the carrier-phonon interaction responsible for the damping of the oscillations has a resonant character. While in the linear limit the system response depends only on the spectral decomposition of the pulse, the situation is different when a strong pulse induces an oscillating charge distribution in the system. In a semiclassical picture, this would act as a driving force for the lattice dynamics. If the induced carrier dynamics is much faster than phonon oscillations the lattice has no time to react until the optical excitation is done. The subsequent dynamics will lead to exciton dressing, accompanied by emission of phonon packets, and will partly destroy coherence of superposition states[9,19–21] but cannot change the exciton occupation number. In the opposite limit, the carrier dynamics is slow enough for the lattice to follow adiabatically. The optical excitation may then be stopped at any stage without any lattice relaxation incurred, hence with no coherence loss. The intermediate case corresponds to modifying the charge distribution in the

QD with frequencies resonant with the lattice modes which leads to increased interaction with phonons and to decrease of the carrier coherence (see Ref. 24 for a simple, single-mode model).

The above ideas are supported by the formal analysis in the following. First, we describe the theoretical method used; then the description is applied to a specific self-assembled InAs/GaAs quantum dot to get quantitative estimations; finally, we conclude with some summary and possible consequences of the results.

Following the earlier studies[19,20,25] successfully accounting for the observed carrier dynamics on picosecond time scales, we describe the system by the rotating wave approximation Hamiltonian (in the rotating frame)

$$H=H_{\mathrm{X}}+H_{\mathrm{ph}}+H_{\mathrm{int}}.$$

Here $H_{\mathrm{X}}=f(t)(a+a^{\dagger})$ describes the interaction of the ground-state exciton with the resonant laser pulse modulated by the real envelope function $f(t)$ ($a,a^{\dagger}$ are excitonic operators), $H_{\mathrm{ph}}=\Sigma_{\boldsymbol{k}}\omega_{\boldsymbol{k}}b_{\boldsymbol{k}}^{\dagger}b_{\boldsymbol{k}}$ is the lattice Hamiltonian ($b_{\boldsymbol{k}}^{\dagger},b_{\boldsymbol{k}}$ refer to the phonon mode $\boldsymbol{k}$ whose energy is $\omega_{\boldsymbol{k}}$), and

$$H_{\mathrm{int}}=a^{\dagger}a\sum_{\boldsymbol{k},s}(f_{\boldsymbol{k}}b_{\boldsymbol{k}}^{\dagger}+f_{\boldsymbol{k}}^{*}b_{\boldsymbol{k}})$$

is the interaction Hamiltonian ($f_{\boldsymbol{k}}$ are the coupling constants for the individual phonon modes). Due to momentum conservation, only long wavelength phonon modes (compared to dot size) are effectively coupled.[26,27] For the time scales relevant here, the damping effect is caused mostly by acoustic phonons.[23,25] In the calculations, we include deformation-potential coupling to longitudinal acoustical phonons, which describes the modification of the band structure due to lattice compression and is different for electrons and holes. Since no cancellation is involved, its effect depends only weakly on the exact wave-function geometry. We assume that transitions to higher states, including possible intervalley transitions, are well separated in energy, compared to the spectral characteristics of the dynamics considered here and may be neglected. Also biexciton transitions are assumed to be eliminated by a suitable polarization choice. Piezoelectric coupling to acoustic phonons has been shown to be negligible if the electron and hole function overlap[19] and is neglected here (but might contribute if the wave-function geometry is different). We have verified that the Fröhlich coupling to the longitudinal optical (LO) phonons is negligible, not only due to charge cancellation but mostly to large LO phonon frequencies, compared to the time scales involved.

Initially, the state of the system is described by the density matrix $\varrho=\varrho_0\otimes\varrho_1(T)$, where ϱ_0 is the state with no exciton and $\varrho_1(T)$ is the thermal equilibrium of lattice modes at the temperature T. The dynamics of the system is calculated by the perturbation expansion of the evolution operator in Born approximation. The lattice degrees of freedom are then traced out leading to the reduced density matrix for the carrier subsystem, which contains all the information accessible by optical methods. The details of the procedure may be found in Ref. 22. As a result, the reduced density matrix after time t may be written as $\varrho(t)=U(t)(\varrho_0+\varrho_1)U^{\dagger}(t)$, where $U(t)$ is the driven evolution for the unperturbed carrier system and ϱ_1 describes the perturbation to the carrier dynamics coming from phonons. The matrix elements of the latter, in the basis of empty dot ($|0\rangle$) and one exciton ($|1\rangle$) states, may be written as

$$\langle 0|\varrho_1|0\rangle=-\langle 1|\varrho_1|1\rangle=-\int_{-\infty}^{\infty}d\omega\frac{R(\omega)}{\omega^2}S_{00}(\omega), \quad (1a)$$

$$\langle 0|\varrho_1|1\rangle=\langle 1|\varrho_1|0\rangle^{*}=-\int_{-\infty}^{\infty}d\omega\frac{R(\omega)}{\omega^2}S_{01}(\omega). \quad (1b)$$

An additional component describing a unitary correction, e.g., light coupling renormalization and energy shifts, may be canceled by an appropriate modification of H_{X} (these effects may lead, e.g., to intensity dependence of the observed Rabi frequency[23]). The function $R(\omega)$ is the spectral density of the reservoir, fully characterizing the lattice properties at this order of perturbation treatment, defined as

$$R(\omega)=\sum_{\boldsymbol{k}}|f_{\boldsymbol{k}}|^2[(n_{\boldsymbol{k}}+1)\delta(\omega-\omega_{\boldsymbol{k}})+n_{\boldsymbol{k}}\delta(\omega+\omega_{\boldsymbol{k}})].$$

It depends on the material parameters and QD size via the coupling constants $f_{\boldsymbol{k}}$ [see, e.g., Ref. 21 for explicit formulas]. The functions $S_{00}(\omega),S_{01}(\omega)$ contain the complete information on the optically controlled carrier dynamics. If we assume that the final state is measured after a time long compared to the phonon oscillation periods, they may be written as

$$S_{00}(\omega)=\tfrac{1}{4}[\sin^2\alpha+|K_{\mathrm{s}}(\omega)|^2],$$

$$S_{01}(\omega)=\tfrac{1}{8}[\sin 2\alpha+2\,\mathrm{Re}\,K_{\mathrm{s}}(\omega)K_{\mathrm{c}}^{*}(\omega)],$$

where

$$K_{\mathrm{s}}(\omega)=\int_s^t d\tau e^{i\omega\tau}\frac{d}{d\tau}\sin 2F(\tau), \quad (2a)$$

$$K_{\mathrm{c}}(\omega)=\int_s^t d\tau e^{i\omega\tau}\frac{d}{d\tau}\cos 2F(\tau), \quad (2b)$$

where s,t are the initial and the final time of the evolution, $F(t)=\int_s^t d\tau f(\tau)$ is the rotation (on the Bloch sphere) performed on the exciton state up to time t, and α is the total angle of rotation. For a given α, the functions (2a,b) actually depend only on $\omega\tau_{\mathrm{p}}$, where τ_{p} is the pulse duration.

In a typical experiment[12,15,16] one measures the average dot occupation, $\langle n\rangle=\langle 1|\varrho(\infty)|1\rangle$, after a pulse of fixed length but variable amplitude. The simulation of such an experiment within the presented model is shown in Fig. 1 for Gaussian pulses [τ_{p} is the full width at half maximum of the pulse envelope $f(t)$]. On the grounds of earlier numerical calculations,[21] we have assumed the exciton ground state to be of nearly product character, with Gaussian electron and hole wave-functions, with in-plane localization widths of 4.9 nm and 4.0 nm, respectively, and with growth-direction width of 1 nm (this corresponds to a dot of roughly 20 nm

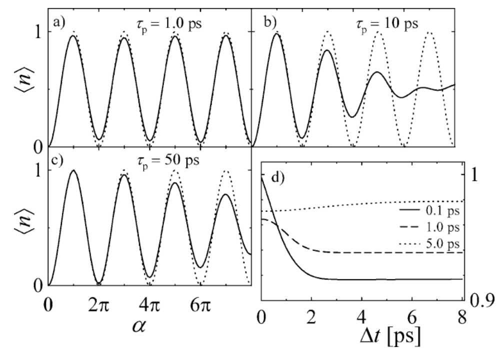

FIG. 1. (a–c) Pulse-area-dependent Rabi oscillations for various pulse durations τ_p as shown in the figure, for $T=10$ K (α is the rotation angle on the Bloch sphere). Dotted line shows unperturbed oscillations. (d) The final QD occupation after two $\pi/2$ pulses separated by time interval Δt for pulse durations as shown.

diameter and 4 nm height). The oscillations are almost perfect for very short pulses (~ 1 ps), then lose their quality for longer pulse durations (~ 10 ps). Although this might be expected from any simple decoherence model, the striking feature is that the effect dramatically grows for higher oscillations, despite the fact that the whole process has exactly constant duration. Even more surprising is the improvement of the quality of oscillations for long pulses (~ 50 nm) where, in addition, the first oscillation is nearly perfect.

The formulas (1a,b) quantify the idea of resonance between the induced dynamics and lattice modes: In Fig. 2 the phonon spectral density is compared to the nonlinear frequency characteristics of the optically controlled exciton dynamics for $\tau_p=1$ ps (the characteristics for other durations is easily obtained by scaling). According to Eqs. (1a,b), the overlap of these spectral characteristics with the phonon spectral density gives the perturbation of the coherent carrier dynamics.

For growing number of rotations n, the nonlinear pulse spectrum $S_{00}(\omega)$ develops a series of maxima of increasing strength [Figs. 2(a), 2(b), and 2(c)]. The position of the last and highest maximum corresponds approximately to $2\pi n/\tau_p$, in accordance with the semiclassical resonance concept. However, spectral components are also present at all the frequencies $2\pi n'/\tau_p$, $n'<n$, which is due to the turning on/off of the pulse. It is interesting to note that for high n, the low-frequency part of $S_{00}(\omega)$ does not evolve with n [Fig. 2(d)]. It is now clear that there are two ways of minimizing the overlap: either the pulse must be so short that all the maxima of $S_{00}(\omega)$ are pushed to the right into the exponentially vanishing tail of the reservoir spectral density $R(\omega)$, or the pulse must be very long, to "squeeze" the spectral function near $\omega=0$ and thus reduce its area. In the latter case, the maxima developing with growing number of oscillations will eventually overlap with $R(\omega)$ destroying the coherence.

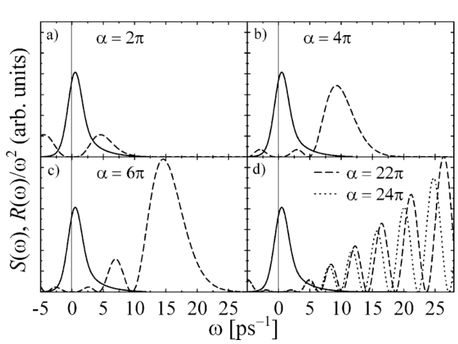

FIG. 2. Phonon spectral density $R(\omega)/\omega^2$ (solid) for deformation-potential coupling to LA phonons at $T=10$ K and the nonlinear pulse spectrum $S_{00}(\omega)$ (dashed and dotted lines) for $\tau_p=1$ ps, for rotation angles α as shown.

Although it might seem that speeding up the process is the preferred solution, it is clear that this works only because no high-frequency features are included into the present model. In reality, speeding up the dynamics is limited, e.g., by the presence of excited states and nonadiabatically enhanced LO phonon coupling.[28] Moreover, it turns out that the resulting dynamics, even within this model, is actually not fully coherent. It has been shown[20] that superposition of states created by an ultrashort $\pi/2$ pulse becomes corrupted, preventing a second pulse (after some delay time Δt) from generating the final state of $\langle n\rangle=1$ with unit efficiency. In order to prove the fully coherent character of carrier dynamics it is necessary to demonstrate the stability of the intermediate state in a two-pulse experiment. The simulations of such an experiment are shown in Fig. 1(d). A short pulse ($\tau_p=0.1$ ps) creates a superposition of bare states (surrounded by nondistorted lattice) which then decohere due to dressing processes.[19,27] As a result, the exciton cannot be created by the second pulse with unit probability.[20] For a longer pulse ($\tau_p=1.0$ ps), the lattice partly manages to follow the evolution of charge distribution during the optical operation and the destructive effect is smaller. Finally, if the carrier dynamics is slow compared to the lattice response times ($\tau_p\sim 10.0$ ps), the lattice distortion follows adiabatically the changes in the charge distribution and the superposition created by the first pulse is an eigenstate of the interacting carrier-lattice system, hence does not undergo any decoherence and the final effect is the same for any delay time (its quality limited by decoherence effects during pulsing). In fact, splitting the π pulse into two corresponds to slowing down the carrier dynamics which, in the absence of decoherence during delay time, improves the quality of the final state, as seen in Fig. 1(d).

The above analysis shows that damping of pulse-area-dependent Rabi oscillations due to interaction with lattice modes is a fundamental effect of non-Markovian character: it is due to a semiclassical resonance between optically induced confined charge dynamics and lattice modes. The destructive effect may be minimized both by speeding up and slowing down the dynamics. However, in the former case, the system passes through unstable (decohering) states. Moreover, fast operation on a real system induces many undesirable effects: transitions to higher states, biexciton generation, or resonant LO phonon dynamics. On the other hand, for slow operation, the number of "good" oscillations is limited. Thus, it is impossible to perform an arbitrary number of fully coherent Rabi oscillations on an exciton confined in a quantum dot.

By increasing the pulse duration as $\tau_{\rm p} \sim \alpha^2$, the phonon effect on the exciton dynamics may be kept constant. However, in this case the achievable number of oscillations is strongly restricted by the exciton lifetime and other (thermally activated) processes. Eliminating the radiative losses, e.g., by using stimulated Raman adiabatic passage instead of a simple optical excitation[29] seems to be a promising direction from this point of view.

The model presented above accounts for the decrease of the quality of Rabi oscillations in the short duration range observed in the experiment.[16] It predicts, however, that this trend is reversed for longer pulse durations. The quantitative value of 96% for the first maximum of the oscillations with $\tau_{\rm p}=1$ ps agrees very well with the experimental result,[15] although the following extrema are much worse in reality than predicted here. This suggests an increased lattice response at higher frequencies which may be due to more complicated wave-function geometry[30] or electric-field-induced charge separation leading to strong piezoelectric effects.[19] Nevertheless, as far as it may be inferred from the experiment,[15] the decrease of the oscillation quality seems to saturate after one full Rabi rotation, as predicted by the model calculations.

In conclusion, we have studied the carrier-lattice dynamics for optically induced Rabi oscillations of exciton occupation in a quantum dot. We have shown that the lattice response is resonantly driven by a combination of the (linear) pulse spectrum and the Rabi frequency. This semiclassical interpretation allows one to qualitatively predict the damping effect based on the general knowledge of the spectral properties of the lattice modes and leads to the conclusion that the resonant lattice response precludes performing a large number of fully coherent oscillations in this system.

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