Damping of Rabi oscillations in quantum dots
due to lattice dynamics

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Abstract. We show that the interaction between carriers confined in
a quantum dot and the surrounding lattice under external driving of
carrier dynamics has a dynamical, resonant character. The quality of Rabi
oscillations in such a system depends on the relation between nonlinear
spectral characteristics of the driven dynamics and the spectral density
of effectively coupled lattice modes (phonon frequencies and density of
states). For a large number of Rabi oscillations within a fixed time
(allowed by e.g. exciton recombination) the spectrum of the dynamics
extends towards high frequencies, coming into resonance with acoustical
and optical phonons. Thus, this resonant lattice response strongly restricts
the possibility of fully coherent control over the charge state in a quantum
dot.

Demonstration of Rabi oscillations in semiconductor quantum dots (QDs)
recently reported by a few research groups [1, 2, 3, 4, 5] is a major step towards
coherent control of the charge state in these systems. Such oscillations in a solid-
state environment are strongly perturbed by interaction with lattice modes. It
has been shown both experimentally [6] and theoretically [7, 8, 9] that coupling
to phonons reduces the coherence of the charge system on picosecond timescale.
This effect may be related to lattice relaxation following non-adiabatic (with
respect to lattice modes) optical excitation (phonon dressing effect) [10]. It has
been shown [11, 12] that this decoherence is a dynamical, non-Markovian effect
and may be minimized by slowing down the carrier excitation dynamics.

In this paper we describe the dynamics of an exciton in an InAs/GaAs QD
under coherent optical excitation in the presence of coupling to lattice modes.
In order to correctly account for the dynamical properties of the lattice, we solve
the non-Markovian Master equation for the reduced density matrix of the carrier
subsystem (in Born approximation, i.e. including one-phonon effects). The
exciton states in the QD are found numerically, including Coulomb interaction,
in a simple confinement model [10] and neglecting multi-exciton states. We
find the dot occupation (mean exciton number $\langle n \rangle$) upon exciting with a
Gaussian pulse of fixed duration and varying amplitude [13], parametrized by
the rotation angle $\alpha$ on the Bloch sphere. The proposed approach goes beyond
the phenomenological description used so far [11, 12] and yields insight into the
dynamical, resonant nature of the carrier–lattice interaction.

The resulting Rabi oscillations are shown on Fig. 1a-c (all calculations are
done for $T = 10$ K). Coupling to lattice modes reduces the amplitude of these
oscillations, the effect being small for short pulses ($\tau_p = 1$ ps) and increasing for longer pulses (10 ps), in qualitative agreement with the experiment [5]. However, for still longer pulses (50 ps), the quality of Rabi oscillation again improves, manifesting the adiabatic character of the carrier-lattice dynamics (transition through dressed states, as witnessed by their lower decoherence in a two-pulse experiment, Fig. 1d).

![Figure 1](image-url)

An explanation of this behavior may be obtained by invoking the lattice inertia, reflected by the natural timescales of phonon dynamics, i.e. by phonon frequencies. 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 [6, 7, 8, 10] 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 (see Fig. 1d). 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 (cf. also [14]).

More quantitatively, for growing number of rotations $m$, the nonlinear spectral characteristics of the optically induced dynamics develops a series of maxima of increasing strength. The position of the last and highest maximum corresponds approximately to $2\pi m/\tau_p$, in accordance with the resonance concept. However, spectral components are also present at all the frequencies $2\pi m'/\tau_p$, $m' < m$, which is due to the turning on/off of the pulse. The
interaction with lattice modes is strong when the frequencies of the induced
dynamics are in resonance with the natural lattice frequencies. An interesting
manifestation of this resonance effect may be observed for sub-picosecond
pulses, when the excitation of longitudinal optical (LO) phonons becomes
important (Fig. 2). Although the coupling between the ground state and the
LO modes is strongly reduced by charge cancellation, effects involving higher
(dark) exciton states may have considerable impact [10–15]. The effect depends
on the relative positions of the narrow LO phonon features and of the induced
dynamics frequencies and has a non-monotonous character (Fig. 2a).

In conclusion, we have shown that the quality of Rabi oscillations of
exciton occupation in a quantum dot deteriorates due to resonant excitation
of lattice modes (overlapping nonlinear pulse spectrum and phonon spectral
density). Coherence may be protected from this effect by slowing down the
dynamics which, however, increases the impact of a number of other decoherence
mechanisms. Hence, the feasibility of coherent control in such a system is
strongly limited.

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