Detecting Strain Wave Propagation Through Quantum Dots by Pump-Probe Spectroscopy: A Theoretical Analysis

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Abstract. The influence of strain waves traveling across a quantum dot structure on its optical response is studied for two different situations: First, a strain wave is created by the optical excitation of a single quantum dot near a surface which, after reflection at the surface, reenters the dot; second, a phonon wave packet is emitted by the excitation of a nearby second dot and then travels across the quantum dot. Pump-probe type excitations are simulated for quantum dots in the strong confinement limit. We show that the optical signals allow us to monitor crossing strain waves for both structures in the real-time response as well as in the corresponding pump-probe spectra. In the time-derivative of the phase of the polarization a distinct trace reflects the instantaneous shifts of the transition energy during the passage while in the spectra pronounced oscillations reveal the passage of the strain waves.

1. Introduction

The interaction of electrons and acoustic phonons in optically excited quantum dots (QDs) has been widely investigated [1–5]. At low temperatures and small dot sizes the dynamics of those systems are governed by non-Markovian processes often referred to as pure dephasing. In this case the ultra-short excitation of a QD gives rise to the generation of an acoustic-phonon occupation that remains within the QD, i.e., the formation of a stable acoustic polaron, and a strain wave that travels into the surrounding of the dot with a temporal envelope in the picosecond regime [6]. While the polaron formation is in principle a reversible process, the outgoing strain wave carries away information and thus causes a loss of electronic coherence [7].

In previous studies we have analyzed the impact of strain waves traveling across a QD on the optical response of the dot for two different set-ups: First, a single QD near a surface where the optically generated wave packets are reflected at the surface and subsequently can reenter the dot. Second, two nearby QDs where the excitation of both dots creates strain waves that travel afterwards through the neighboring dot. We have found that the corresponding optical polarizations resulting from a single laser pulse excitation are noticeably affected by such strain waves [8]. In this contribution we simulate two-pulse excitations of pump-probe type for the above mentioned two set-ups. We show that pump-probe type excitations allow us to monitor strain waves traveling across a QD for both structures in the real-time response as well as in the corresponding spectra.
2. Theoretical background

We consider QDs in the strong confinement limit excited by circularly polarized light assuming a vanishing or sufficiently weak exchange splitting between the two bright excitons. In this case biexcitonic effects are negligible and the QDs can be modeled as effective two-level systems (the ground state of the unexcited dot and the exciton state). The standard carrier acoustic-phonon coupling via the deformation potential is used and we focus on pure dephasing processes, which do not change the occupation of the electronic states as described by the independent boson model[6]. For the optical excitation we take pulses shorter than the characteristic timescale of carrier-phonon interactions, which thus can be modeled by δ-pulses. For this situation the optical response of both set-ups can by obtained by an approach based on generating functions that provides exact analytical results for any sequence of ultra fast pulses [9]. Its application to a single QD near surface is described in Ref. [10] while the two-QD case can be found in Ref. [8]. Here we will just specify the excitation scenarios; for details we refer the reader to the above mentioned papers.

For the single QD (SQD) located at a distance \( d \) from a surface we assume that the pump pulse with pulse area \( f_1 = \pi \) arrives at \( t_1 = -\tau \). This pulse inverts the QD occupation without inducing a polarization [9]. In addition, a completely coherent phonon wave packet is emitted [6]. The probe pulse is supposed to have a pulse area \( f_2 = \pi/2 \). It excites the dot at \( t_2 = 0 \) and creates a polarization of the form \( P^{SQD}_{PP}(t) = |P_0(t)|e^{-i\bar{\Omega}t + i\Phi_{SQD}} \). Here, \( |P_0(t)| \) denotes the modulus of the optical polarization after a single pulse excitation [2], however calculated with the coupling to half-space phonon modes [10]; \( \bar{\Omega} \) is the polaron-shifted exciton frequency. Hence, the impact of the strain wave generated by the pump pulse on the polarization induced by the probe pulse is only given by a shift in the phase \( \Phi_{SQD} \) while the amplitude is unaffected.

For the two QD (TQD) system we assume QDs of different sizes separated by a distance \( 2d \), thus allowing for a selective excitation of each QD due to different exciton energies. Each QD is excited by one pulse: the pump pulse with pulse area \( f_1 = \pi \) arrives at \( t_1 = -\tau \) and excites one of the QDs which subsequently emits a phonon wave packet without inducing a polarization. The probe pulse with pulse area \( f_2 = \pi/2 \) excites the neighboring dot at \( t_2 = 0 \). The probe pulse induces a polarization \( P^{TQD}_{PP}(t) = |P_0(t)|e^{-i\bar{\Omega}t + i\Phi_{TQD}} \), where \( |P_0(t)| \) again coincides with the modulus of the optical polarization after a single pulse excitation, here however calculated with the usual full-space (plane wave) phonon modes. Again, the information on the wave packet generated by the other dot enters only in the phase \( \Phi_{TQD} \). We assume that the two dots are arranged in an infinite medium, the separation between both being large enough to make tunneling effects negligible. Förster-type couplings via interband dipole moments are negligible due to the different exciton energies.

3. Results

Let us first discuss the time-derivative \( \dot{\Phi}(t) \) of the phase of the optical polarization. This time-derivative can be interpreted as the instantaneous energy shift of the exciton caused by the coupling to the phonons. Indeed, in the case of quantum well such shifts of the exciton line resulting from the passage of a strain wave have already been observed experimentally [11]. Figure 1(a) shows \( \Phi_{TQD}(t) \) for the TQD system with dots separated by \( 2d = 20 \) nm as a function of the real time \( t \) and the delay time \( \tau \). Two regions where \( \dot{\Phi}_{TQD}(t) \) exhibits a pronounced time-dependence can be observed. For all delay times strong time-dependencies occur during the first picosecond after the optical excitation. They describe the formation of the polaron created by the probe pulse and are therefore independent of the pump pulse. In addition a clear signal is visible for values \( t + \tau \approx 4 \) ps. Indeed, here \( t + \tau = 2d/c_L \) with the longitudinal sound velocity \( c_L \); therefore this signal appears when the strain wave created by the pump pulse in the other dot passes the probed dot. For \( \tau > 2d/c_L \) the wave packet has passed the probed dot before its excitation and does not affect its polarization.
Figure 1. Time derivative of the phase of the optical polarization induced by the probe pulse as a function of real time \( t \) and delay time \( \tau \) at \( T = 1 \) K (a) for a two-QD system (distance \( 2d = 20 \) nm); (b) for a single QD near a surface (\( d = 10 \) nm).

Let us now analyze \( \Phi_{SQD}(t) \) for a SQD located at a distance \( d = 10 \) nm from a surface [Fig. 1(b)]. Here three structures are visible. As in the TQD case, we find for all delay times a pronounced time-dependence during the first picosecond but with opposite sign. This is because the probe pulse induces a polarization in an initially inverted QD; \( \Phi(t) \) thus reflects the polaron destruction which goes along with carrier transitions from the exciton state to the ground state. For values \( t + \tau > 2d/c_l \) also in the half-space case a pronounced structure is visible, which can be traced back to the reflected strain wave generated by the pump pulse. It is therefore absent for \( \tau > 2d/c_l \). Finally, at times \( t = 2d/c_l \), and hence independent of the delay time, the wave packet generated by the probe pulse and reflected from the surface is recorded. The probe pulse is weaker then the pump pulse and as a consequence it emits a wave packet with a smaller amplitude resulting in a smaller signal in \( \Phi_{SQD}(t) \).

The influence of passing strain waves can also be monitored in the absorption spectra of the probe pulse. It is calculated as the imaginary part of the Fourier transform of \( P(t) \). To model the long time decay of the polarization caused, e.g., by radiative decay or other Markovian decay channels, the polarization has been multiplied by a weak exponential decay \( e^{-\gamma t} \) with \( 1/\gamma = 500 \) ps, thus accounting phenomenologically for the finite width of the zero-phonon line (ZPL). All curves are calculated for \( T = 1 \) K.

Figure 2(a) shows the spectra of the TQD system for \( 2d = 20 \) nm and different delay times. For \( \tau > 2d/c_l \) the strain wave has already passed the probed dot before the latter is excited. Thus the shape of the spectra agrees with the absorption spectrum of a single QD in an infinite bulk medium [2]: a narrow ZPL superimposed on a broad acoustic phonon background which, due to the low temperature, is highly asymmetric. For \( \tau < 2d/c_l \) spectral oscillations appear in the phonon background. The oscillation period corresponds to the time between the excitation by the probe pulse and the arrival time of strain wave generated by the pump pulse.

The optical spectra in the SQD case with \( d = 10 \) nm and for different delay times are presented in Fig. 2(b). The overall shape resembles that of the TQD case but inverted with respect to the origin, since transitions from the excited state to the ground state are induced. For all delay times we observe on the low energy side (below the ZPL) spectral oscillations in the background spectra with a period corresponding to the arrival time of the wave packet generated by the
Figure 2. Probe absorption spectra for different delay times $\tau$ at $T = 1$ K (a) for a two-QD system with $2d = 20$ nm; (b) for a single QD near a surface with $d = 10$ nm.

probe pulse. For $\tau < 2d/c_1$ the strain wave generated by the pump pulse gives rise to additional oscillations with a period corresponding to the arrival time of the pump-generated strain wave. Like in the TQD case these oscillations are present both below and above the ZPL leading to a complicated shape with two frequencies below the ZPL.

4. Conclusions

We have simulated pump-probe excitations in strongly confined QDs and analyzed the influence of strain waves passing the QDs. Two situations were considered: a single QD near a surface, where the strain wave is generated by the QD itself and travels again across the dot after being reflected at the surface; and a two-QD system in an infinite medium, where the strain wave is generated by a nearby QD and then passes across the dot. In both set-ups the strain fields noticeably affect the polarization. In the phase of the polarization a distinct trace reflects the instantaneous shifts of the transition energy during the passage of the wave packet while in the spectra pronounced oscillations occur.

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