Impact of strain waves traveling across a quantum dot on the optical response of the dot: Distinction between strain waves of different origin

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The influence of strain waves traveling across a semiconductor quantum dot on the optical spectra of the lowest quantum dot transition is analyzed. Pure dephasing interactions between electronic and phononic degrees of freedom in quantum dot systems are considered, which represent the most important type of electron-phonon interaction in strongly confined quantum dots. For the case of excitation by ultrafast laser pulses, a generating function formalism provides analytical results, which are exact within the framework of the model. Two situations are compared: (i) a phonon wave packet is generated by the optical excitation of a single-quantum dot near a surface, which after reflection at the surface reenters the quantum dot and (ii) a phonon wave packet is generated by the excitation of a nearby second dot and then travels across the quantum dot. Although the displacement fields passing the dot are almost identical in these two situations, we find that the real time responses as well as the corresponding spectra exhibit qualitative differences and thus allow for a discrimination of phonon wave packets from different origins.

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I. INTRODUCTION

Optically excited quantum dots (QDs) in the strong confinement limit with electronic levels coupled to phonons have been widely investigated.[1–7] At low temperatures and small dot sizes the dynamics of those systems are governed by non-Markovian processes, which are often referred to as pure dephasing. The pure dephasing coupling of the electronic degrees of freedom to phonons is known to be responsible for a number of characteristic dynamic features. For example, the excitation of a QD with ultrashort optical pulses is accompanied by the creation of an acoustic-phonon occupation that remains within the QD, i.e., the formation of a stable acoustic polaron and a phonon wave packet that propagates into the surrounding of the QD with a temporal envelope in the picosecond regime.[8] While the polaron formation is in principle a reversible process, the creation of an outgoing phonon wave entangles the dot with its environment and thus leads to an irreversible dynamics.[9] This is reflected by a loss of electronic coherence, which can be monitored by following the time evolution of the optical polarization of the QD.[10] The polarization dephases as long as the outgoing strain wave has a spatial overlap with the QD. Indeed, for a QD embedded in a bulk material, the polarization reaches a plateau once the phonon wave has left the dot.

Experimentally, the optical generation and detection of ultrafast strain waves has attracted much attention for many years.[11–16] In particular, a strong impact of picosecond phonon wave packets traveling across nanostructured semiconductors on the optical response of these structures has been observed in corresponding experiments performed on quantum wells.[17] Acoustic phonons emitted by photoexcited carriers in QDs have been measured by bolometric detection.[18,19] In principle also QDs cannot only be used for the generation of ultrashort phonon wave packets but also for their optical detection when these wave packets travel across the dot. This can be realized in different ways. One possibility is to place a dot near a surface. Then, the phonon wave that is emitted from the dot after a short-pulse laser excitation is reflected at the surface and thus can subsequently re-enter the QD. This situation has been previously analyzed and it has been found that the reentrance of the phonon wave in the QD has a pronounced effect on the polarization of the QD, which should be optically detectable.[20] Another possibility is to consider two nearby dots. In this case a simultaneous excitation of both dots will result in strain waves emerging from each of the dots. When the strain wave generated by one of the dots reaches the other dot, a corresponding effect on the optical polarization should be expected. In Fig. 1 both situations are sketched. Obviously, when the two dots in the second scenario are separated by twice the distance of the dot from the surface, then the two setups are connected, such as in electrostatics, where a charge in front of a conducting wall is related to its corresponding mirror charge construction. Thus, the lattice displacements in the left half space of Fig. 1 should be very similar. It is therefore tempting to expect that also the corresponding changes of the QD polarizations that are observed when the phonon wave travels across the dot should be similar. In this paper we will show, however, that the real time responses, as well as the corresponding spectra, are qualitatively different and thus allow for a discrimination of phonon wave packets originating either from the same dot or from another source.

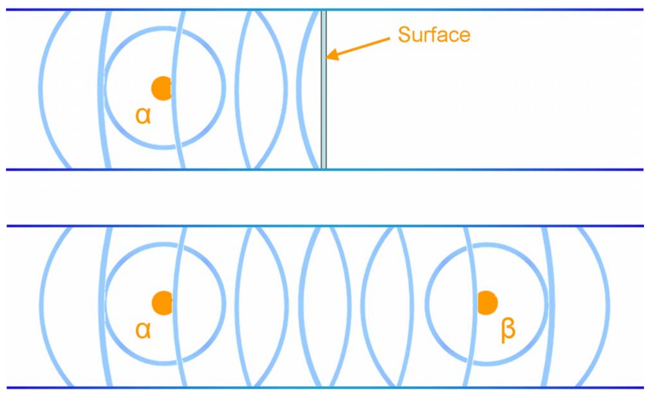

FIG. 1. (Color online) Schematic of the strain waves generated by a QD near a surface (upper part) and two-QDs (lower part).

II. THEORY

We consider QDs in the strong confinement limit modeled by an electronic two-level system coupled to phonons and to an external laser field. The standard carrier acoustic-phonon coupling Hamiltonian[2,8] is used and we concentrate on pure dephasing processes, which do not change the occupation of the electronic states as described by the independent boson model.[21,22] In recent experiments, pure dephasing has been revealed to be the dominant dephasing mechanism on short-time scales for optically excited QDs in the strong confinement limit.[23] For many typical QD systems, the deformation-potential coupling provides the largest contribution to pure dephasing.[2,24] We shall therefore concentrate on this mechanism. Furthermore, it was noted previously[24] that the contribution to pure dephasing resulting from couplings to higher excited states can be neglected under realistic conditions, even in dots which are larger than those studied here. This, as well as the convincing agreement of corresponding model studies with experiments,[23] justifies a restriction to only the lowest QD sublevels. If we assume excitations with circularly polarized light and a sufficiently small exchange splitting between the two bright dot excitons with lowest energy, then biexcitonic effects become irrelevant and the electronic system of the QD can be modeled as an effective two-level system.[10,25] The interaction with the classical light field is treated by the usual dipole coupling. We consider pulses that are shorter than the timescale of carrier-phonon interactions (for typical QDs shorter than about one hundred femtoseconds), which thus can well be modeled by δ functions. For this case exact analytical solutions have been derived by using a generating function approach both for an electronic two-level system[8] and for an electronic four-level system including biexcitons.[25] We will not repeat the general derivations but specify the two models that shall be considered here and indicate for each situation where the general technique and its application are described in detail.

A. Single-QD in a half space

We consider a single two-level QD in a half space at a distance d from the surface as sketched in the upper part of Fig. 1. The coordinates are taken such that the surface is in the xy plane and the dot is on the z axis located at $z=-d$. The electronic eigenstates are the ground states of the unexcited dot $|0\rangle$ and the exciton state $|\alpha\rangle$. For such a two-level system, the polarization after a δ-like pulse with a pulse area f and a phase φ reads[8]

$$\mathbf{P}(t) = \Theta(t)\frac{i\mathbf{M}_0^* e^{i\varphi}\sin f}{2}e^{-i\bar{\Omega}t}e^{\Xi(t)}, \quad (1)$$

with

$$\Xi(t) = -\sum_{\xi}|\gamma_\xi|^2[(1+2n_\xi)(1-\cos\omega_\xi t)+i\sin\omega_\xi t], \quad (2)$$

where ξ denotes the phonon mode index, ω_ξ is the frequency of a phonon in the mode ξ, n_ξ is the equilibrium phonon occupation at a given temperature T, and $\gamma_\xi := (g_\xi^e - g_\xi^h)/\omega_\xi$ is a dimensionless coupling strength with $g_\xi^{e/h}$ being the couplings to electrons or holes, respectively. The polaron shifted gap energy is given by $\bar{\Omega}=\Omega-\sum_\xi\omega_\xi|\gamma_\xi|^2$ with $\hbar\Omega$ being the bare exciton energy. Finally, $\mathbf{M}_0$ denotes the dipole matrix element.

The calculation of the carrier-phonon couplings $g_\xi^{e/h}$ requires the knowledge of the electron/hole wave functions of the QD (Refs. 2 and 8) as well as the phonon modes of the structures.[20] For simplicity we assume Gaussian electron and hole wave functions valid for harmonic confinement potentials. The dots are modeled as ellipsoids with the shorter extension in the z direction. The phonon mode index ξ refers to the complete set of half-space phonon modes[20,26] comprising three different types of modes: bulklike modes, which represent oscillations that are undamped throughout the sample; surface modes with amplitudes that decay exponentially in the direction of the confinement; and mixed modes, which are composed of bulklike and surface components. Furthermore, the boundary conditions for a stress-free surface lead to a mixing of longitudinal and transverse oscillations.

B. Two-QDs in an infinite medium

The half-space results will be compared to a system with two strongly confined QDs at a distance $2d$ in an infinite medium as sketched in the lower part of Fig. 1. The dots are taken to be on the z axis, QD α is located at $z=-d$, and QD β at $z=d$. In such an infinite medium the phonon modes are simply given by plane waves and, since we restrict ourselves to deformation-potential interaction, only longitudinal-acoustic phonons have to be considered. Thus the mode index ξ can be identified with the phonon wave vector $\mathbf{q}$. Each QD is again modeled as a two-level system. The separation between the dots is supposed to be large enough to make tunneling effects negligible and we also neglect a possible Förster-type coupling via interband dipole moments. Such couplings would give rise to delocalized exciton states and, as a consequence, to real phonon-assisted transitions between these states.[27,28] In contrast, in our case, excitons localized in one of the dots remain the electronic eigenstates and after the optical excitation the dots interact only via the propagating phonon wave packets, which makes the system similar to the case of the single-QD in a half space. The electronic eigenbasis for this system consists of the four states $|0\rangle, |\alpha\rangle, |\beta\rangle, |B\rangle$, where $|0\rangle$ is the ground state, $|\alpha\rangle$ ($|\beta\rangle$) describes an exciton in dot α (β), and $|B\rangle$ denotes the two-exciton state, where both excitons are present. The corresponding energies are $\epsilon_0=0$, ϵ_α, ϵ_β, and $\epsilon_B=\epsilon_\alpha+\epsilon_\beta-\Delta_B\hbar$, where Δ_B is the biexciton shift accounting for electrostatic Coulomb interactions. This shift strongly depends on the dot geometry, on the sizes of the dots, and on other material parameters as well as on external static electric fields.[29] Thus, both situations, with or without a sizable value of Δ_B, may be realized. If not explicitly stated, we have assumed a vanishing biexciton shift in the calculations discussed below. The coupling of the exciton states to the phonons is described by the dimensionless coupling strengths γ_ξ^α and γ_ξ^β; the coupling strength of the biexciton state is then given by $\gamma_\xi^B=\gamma_\xi^\alpha+\gamma_\xi^\beta$.

The optical response of this two-dot system after ultrafast laser excitation can be obtained again by using the generating function approach. The solution is facilitated by noting that the present two-dot system can be formally mapped onto the four-level single-dot system with excitonic and biexcitonic excitations that has been treated in Ref. 25.[30] The two single-exciton states $|\alpha\rangle, |\beta\rangle$ (that in the present system represent excitons in two spatially separated dots) formally correspond to the two single-exciton eigenstates of the four-level single-dot system, while the states with zero or two excitons $|0\rangle$ and $|B\rangle$ can be mapped onto the ground and biexciton state. Thus, with a proper reinterpretation of the states and of the corresponding matrix elements we can immediately transfer the results for the four-level single-dot system to the present two-level two-dot system.

The QDs are excited by δ-like laser pulses with pulse areas f_α (f_β) and phases φ_α (φ_β) corresponding to the excitation of dot α (β). Applying the results of Ref. 25 to our present situation yields the following expression for the polarization of QD α:

$$\mathbf{P}_\alpha(t) = \Theta(t)\frac{i\mathbf{M}_\alpha^* e^{i\varphi_\alpha}\sin f_\alpha}{2} e^{-i\bar{\Omega}_\alpha t} e^{\Xi(t)}\left[\cos^2\frac{f_\beta}{2} + \sin^2\frac{f_\beta}{2} e^{i[\Delta_B t+\Phi(t)]}\right], \qquad (3)$$

where $\mathbf{M}_\alpha$ is the dipole matrix element of dot α, $\Xi(t)$ is the function defined in Eq. (2) with γ_ξ being replaced by γ_ξ^α, and $\Phi(t)$ is given by

$$\Phi(t) = -i\sum_\xi [(\gamma_\xi^{\beta*}\gamma_\xi^\alpha)(e^{-i\omega_\xi t}-1) - (\gamma_\xi^\beta\gamma_\xi^{\alpha*})(e^{i\omega_\xi t}-1)]. \qquad (4)$$

Except for the constant factor $\cos^2(f_\beta/2)$, the first part is equal to a single two-level QD [see Eq. (1)]. The second part involves the biexcitonic shift Δ_B as well as an additional time-dependent phase Φ that stems from the coupling of the dots via the phonons, as can be seen from the explicit appearance of the coupling matrix element of QD β.

In order to make the two-dot system as comparable as possible to the half-space problem with the single dot, for our calculations, we have assumed identical dots α and β, which agree in size and shape with the QD in the half-space case. This means that for the energies and dipole matrix elements, we have $\Omega_\alpha=\Omega_\beta=\Omega$ and $\mathbf{M}_\alpha=\mathbf{M}_\beta=\mathbf{M}_0$. The phonon coupling matrix elements are related by

$$\gamma_\xi^\alpha = \tilde{\gamma}_\xi e^{-idq_z}, \quad \gamma_\xi^\beta = \tilde{\gamma}_\xi e^{idq_z}, \qquad (5)$$

with q_z being the z component of the phonon wave vector and $\tilde{\gamma}_\xi$ is the coupling matrix element of one of the QDs if it were located at the origin of the coordinate system. In this case the time-dependent phase of Eq. (4) reads

$$\Phi(t) = -2\sum_\xi |\tilde{\gamma}_\xi|^2[\sin(\omega_\xi t + 2dq_z) - \sin(2dq_z)]. \qquad (6)$$

Finally, for identical dots excited simultaneously by the same pulse, one has $f_\alpha=f_\beta$ and $\varphi_\alpha=\varphi_\beta$.

Of course, in realistic structures it is more common that the dots exhibit some variation in their sizes and/or geometries and thus also in their exciton energies and phonon couplings. We have performed the calculations also for dots of different shapes and sizes and found, in all cases, the same qualitative features that will be discussed below. In order to emphasize the fact that the two-dot results are fundamentally different from the half-space results and that these differences are not only due to some practically hard-to-avoid asymmetries in the dot shapes, we shall concentrate on the case of identical dots in this paper. However, we want to stress that the qualitative behavior for the two-dot system does not change when realistic variations of the dot shapes are taken into account.

III. RESULTS

A. Polarization dynamics

The aim of this paper is to identify the differences in the optical response of QDs caused by strain waves of different origins traveling across the dots. All our calculations are done for ellipsoidal QDs with lateral and vertical sizes of $L_{x,y}=3$ nm and $L_z=1.5$ nm, respectively, the values referring to the full width at half maximum of the electron charge distribution.[31] For the single-QD in a half space, we consider the excitation by a single ultrashort laser pulse with pulse area $f=\frac{\pi}{2}$, which results in the strongest polarization signal. In order to analyze a situation most similar to the single-QD case, we consider two-QDs of the same size, which are excited simultaneously by a single pulse with $f_\alpha=f_\beta=\frac{\pi}{2}$. Figure 2(a) shows the results for the modulus squared of the optical polarization of dot α in a two-QD structure for different distances $2d=10$ and 20 nm. We obtain a shape familiar from studies of single-QD excitations,[2,32,33] consisting of an initial decay, which then recovers toward a constant long-time plateau value. The initial decay reflects the information loss due to the outgoing phonon wave packet, which propagates into the surrounding material with the longitudinal sound velocity c_l. The long-time value reflects the unbroadened zero-phonon line. With a distance $2d$ between the dots, the wave packet

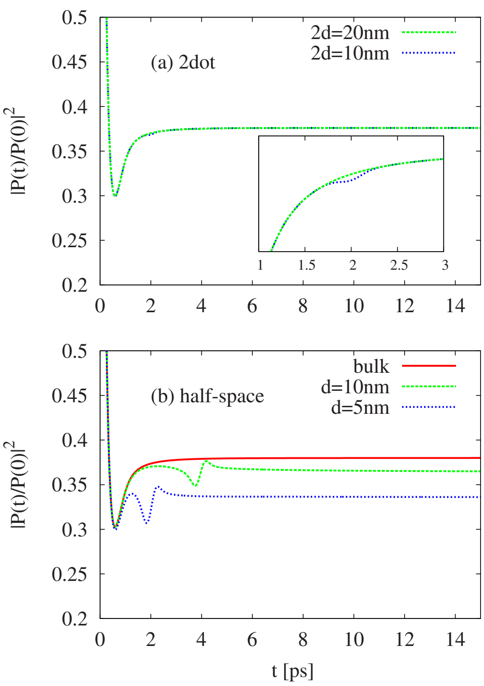

FIG. 2. (Color online) (a) Optical polarization of QD α in a model with two-QDs separated by a distance $2d$. (b) Optical polarization of a single-QD at a distance d from a surface. For comparison, the result for a single-QD in an infinite bulk system is also included. All calculations were made for a temperature $T=1$ K.

generated by dot β reaches dot α at time $t\approx 2d/c_l$, i.e., for $2d=10$ nm at $t\approx 2$ ps. As seen from Fig. 2(a), the passing strain wave has almost no impact on the modulus of the QD polarization. The general shape of the curve can hardly be distinguished from the corresponding single-QD result in an infinite crystal. The inset is a zoom into the region $t\approx 2$ ps, which reveals that a very small dip is actually present. It is interesting to notice that this small dip results from the interference between the first and the second part in Eq. (3), i.e., between the polarizations of QD α when either QD β is unexcited (first part) or QD β is excited (second part). If the two contributions can be measured separately, which is the case for a sufficiently large biexciton shift Δ_B, the modulus of each of the two polarizations exactly agrees with the case of a single-QD in an infinite medium as has been found previously in Ref. 34.

These findings are especially surprising when being compared to the corresponding half-space results. Figure 2(b) shows the optical polarization for a QD located at a distance d from a surface for two values of d as discussed previously in Ref. 20. For comparison the solid line shows the behavior of a QD in an infinite crystal without re-entering strain waves. We observe a pronounced dip that occurs roughly at a time $t=2d/c_l$ and thus corresponds to the time needed by the reflected wave to return to the dot. In addition, a slow decay is seen that begins at $t=d/c_l$, which has been discussed previously in Ref. 20.

One might think that the differences found here between the two-QD and the half-space results are caused by the remaining differences in the displacement fields. Indeed, the displacement fields are similar but not identical because of the stress-free boundary condition, which is the realistic boundary condition for a free surface. As noted before, this results in two major differences between half-space and bulk-phonon modes: the occurrence of exponentially decaying surface modes and the mixing between longitudinal and transverse oscillations. The displacement field of the half-space configuration can be made identical to the displacement in the two-QD model if the boundary conditions in the half-space geometry are modified, such that, they reproduce the symmetries of the two-QD model. Because of the mirror symmetry of this latter model with respect to the $z=0$ plane, the z component of the displacement field has to vanish in that plane. Together with the condition that there is no mode conversion, i.e., that the displacement field is purely longitudinal, this defines a set of phonon modes for an artificial half-space problem. Because of the longitudinal character, the displacement field $\mathbf{u}(\mathbf{r},t)$ can be derived from a potential function $\Psi(\mathbf{r},t)$ according to[35]

$$\mathbf{u}(\mathbf{r},t) = -\,\mathrm{grad}\,\Psi(\mathbf{r},t). \tag{7}$$

The function $\Psi(\mathbf{r},t)$ has to satisfy the wave equation and the mirror symmetry with respect to $z=0$. These conditions are fulfilled for

$$\Psi(\mathbf{r},t) = N\cos(q_z z)e^{i(q_x x+q_y y-c_l q t)}, \tag{8}$$

where N is a normalization constant and $q^2=q_x^2+q_y^2+q_z^2$. It is easy to verify that the displacement field, resulting from a short-pulse excitation of a QD coupled to these cos-phonon modes, is indeed in the half space of the QD identical to the corresponding result in the two-QD model. Figure 3 compares the modulus of the optical polarization obtained for a QD in the half space, with distance $d=10$ nm, by using the two different types of phonon modes. We notice two qualitative differences. First, it is seen that the structure resulting from the reflected wave packet has the opposite sign when comparing cos modes with the modes corresponding to the stress-free boundary. This reflects the fact that the relative volume change, which enters the deformation-potential interaction, exhibits a phase jump upon reflection in the case of a stress-free boundary while no phase jump is present in the case of cos modes. Second, there is no additional decay in the calculations with cos modes. As demon-

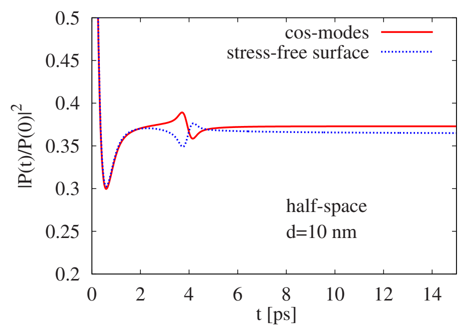

FIG. 3. (Color online) Optical polarization induced by an ultrashort laser pulse at $T=1$ K for a single-QD with distance $d=10$ nm from the surface, which is either coupled to phonon modes corresponding to a stress-free surface or to cos-phonon modes.

strated previously,[20] the additional decay is caused by the excitation of exponentially decaying surface phonons, which are not present in the calculations based on cos modes. However, it turns out that the occurrence of the additional decay and the phase jump are the only features that are due to the differences in the phonon mode structure. In particular, even though the displacement field in the cos-mode calculation is identical to the displacement field in the two-QD model, in the first case we find a pronounced structure in the time-dependent polarization amplitude, while in the second case there is practically no effect visible in $|\mathbf{P}(t)|$ that would indicate that the strain wave is traveling across the QD.

Further insight into the differences of the real time responses of the two-QD system and the QD with a surface can be obtained by analyzing the frequency shifts of the excitonic line during the passage of the wave packets. These instantaneous energy shifts are proportional to the time derivative of the phases of the polarizations $\mathbf{P}(t)$ and $\mathbf{P}_\alpha$ in Eqs. (1) and (3). For quantum well structures under the influence of strain waves, such time-dependent shifts have recently been measured in Ref. 17.

Figure 4(a) shows the time derivative of the phase of the optical polarization for the two-QD system for two different dot distances. Clearly, the excitonic line is shifted twice. The first change describes the polaron formation, which takes place within one picosecond after the optical excitation. After the completion of the polaron formation, the polaron shifted exciton line is observed. The second phase shift occurs roughly when the strain wave generated by QD β travels across QD α. The amplitudes of the shifts are decaying with increasing dot distances due to the decrease of the phonon amplitude for larger distances. The phase shift obtained for a single-QD in a half space coupled to cos-phonon modes is practically identical to the two-QD result as seen from Fig. 4(b). However, when the surface is modeled by stress-free boundary conditions [Fig. 4(c)], the phase shift produced by the reflected strain wave traveling across the QD has the opposite sign and exhibits a slight asymmetry. This change of sign is due to the phase jump of the volume change upon reflection at a stress-free boundary, while the slight asymmetry is again a signature of the surface phonons. It turns out that, in contrast to the polarization amplitude $|\mathbf{P}(t)|$, the instantaneous energy shift of the exciton line is a direct measure for the relative volume change,[20] which implies that identical displacement fields yield identical energy shifts no matter whether the displacement results from reflection or from the excitation of another QD.

B. QD spectra

The qualitatively different impacts of strain waves of different origin on the optical response of a QD can also be monitored in the frequency domain. To this end we have calculated QD spectra, which have been obtained as the imaginary part of the Fourier transform of Eq. (1) for the half space and Eq. (3) for the two-QD model, respectively. Since we do not include a further decay mechanism, e.g., radiative decay, the long-time value of the polarization is nonzero, leading to unbroadened zero-phonon lines (ZPLs), which are

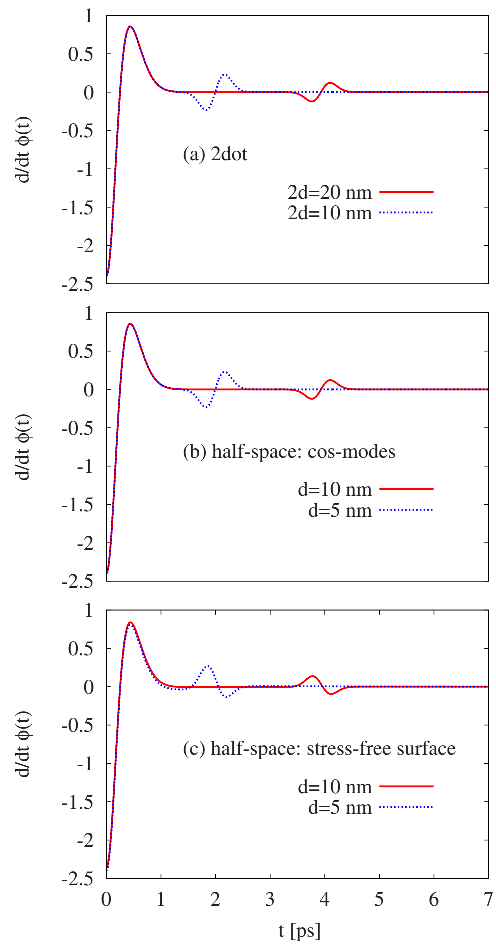

FIG. 4. (Color online) Time derivative of the phase of the optical polarization induced by an ultrashort laser pulse (a) of dot α in a two-QD system for two different dot distances $2d$; (b) of a single-QD in a half space for two distances d from the surface with cos-phonon modes; and (c) same as (b) but with phonon modes corresponding to a stress-free surface.

superimposed on a phonon-induced background.[1,2,10,33] For numerical calculations of the spectra, we have eliminated the ZPLs by subtracting the longtime value of the polarization for each transition and we plot only the remaining background spectra.[10,33] The zero of the energy axis in Figs. 5(a)–5(f) denotes the position of the ZPL. Figure 5 compares the background spectra for a two-QD model [Figs. 5(a) and 5(b)] with a QD in a half space using either cos-phonon modes [Figs. 5(c) and 5(d)] or phonon modes corresponding to a stress-free surface [Figs. 5(e) and 5(f)]. In all cases, the strain waves that travel across the dot lead to pronounced oscillations superimposed on the background spectra. The amplitudes of these oscillations depend on the distance of the QD from the surface or from the second QD and decrease with increasing distance. The frequency difference $\Delta\omega$ between two maxima of the spectral oscillations corresponds to the time the wave packet requires to travel from the QD to the surface and back again or, in the two-dot case, from the second to the first dot. For the QD near a stress-free surface, this has been previously noted in Ref. 20. For all systems studied the background spectra are strongly asymmetric,

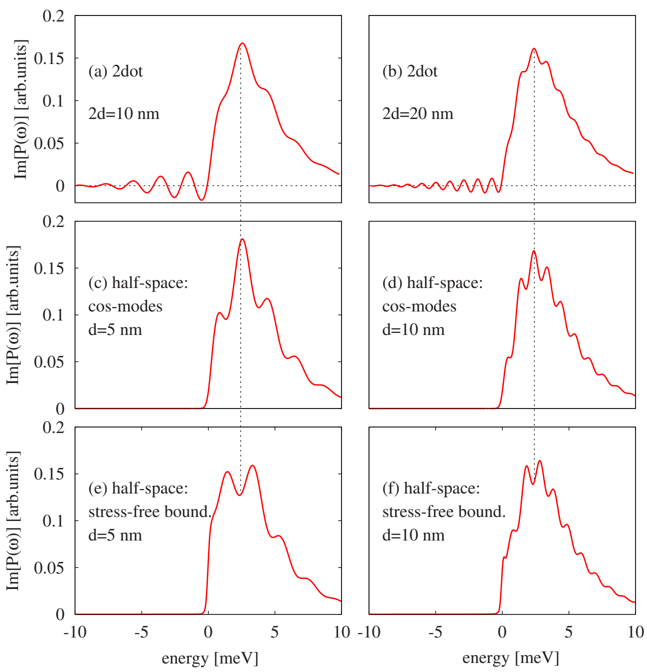

FIG. 5. (Color online) Optical spectra (imaginary part of the Fourier transform of the optical polarization): (a) and (b) for a two-QD model with different dot distances $2d$; (c) and (d) for a single-QD in a half space located at different distances d from the surface calculated with cos-phonon modes; (e) and (f) same as (c) and (d) but for phonon modes corresponding to a stress-free surface. All curves are calculated for a temperature $T=1$ K.

which is typical for low temperatures.[1,2,10,33] Comparing the spectra obtained with the different boundary conditions in the half-space geometry, we notice that the spectral oscillations have an opposite phase. The positions of the maxima in the spectra obtained with the cos modes coincide with the positions of the minima in the case of the stress-free surface boundary condition as indicated by the thin vertical dashed lines in Fig. 5. This opposite phase is a direct consequence of the opposite sign of the frequency shift seen in Fig. 4. There we have found that the frequency shift in the two-QD model agrees with the half-space result using cos modes. Accordingly, we now also find that the background spectra in the two-QD case have their maxima at the same positions as the spectra in the half-space case with cos-phonon modes.

However, there is a striking qualitative difference between the two-QD curves and the spectra for a QD in half space, either using cos-phonon modes or the modes corresponding to the stress-free surface; the frequency domain signals in the two-QD case show negative values for frequencies below the polaron shifted exciton line, i.e., for negative energies in Fig. 5. In addition the spectral oscillations for positive energies are less pronounced than for negative energies and also much weaker than for a QD near a surface. Therefore, as for the time-domain signals the impact of a strain wave on the spectra also depends strongly and qualitatively on the fact whether the strain wave originated in the same or in another QD. This holds even when the corresponding lattice displacements are identical.

It is possible to trace back the physical origin of the differences in the above considered spectra to the fact that the phonon wave packets affect the optical response of the dots via different quantum-mechanical subspaces. In order to see this more clearly, we first note that the optical polarization of QD α in the two-QD case consists of two terms that are weighted according to the pulse area f_β describing the excitation of QD β [cf. Eq. (3)]. In the first term the ground state is connected to an exciton in QD α, i.e., this is a ground state-to-exciton transition (GET). The second term can be interpreted as an exciton-to-biexciton transition (EBT) because it is only present if there is an exciton in QD β. Indeed, the probability for the creation of an exciton in QD β is given by $\sin^2(f_\beta/2)$, while $\cos^2(f_\beta/2)$ is the probability that this dot remains in its ground state. At this point it is important to realize that the carrier-phonon coupling is nonzero in a given dot only when carriers are present. This implies for the two-QD model that in a subspace, where only one of the two excitons is excited, only the dot that contains the exciton is coupled to the phonon modes. Thus, a strain wave generated by the excitation of dot β can affect the polarization of dot α only in the electronic subspace where both dots are excited. In other words in the two-QD model the strain wave from dot β can modify only the EBT part of dot α while the GET is unaffected. The EBT part of the polarization is by its nature a nonlinear signal, which is at least of third order in the driving laser field. Thus, the passage of the strain wave through QD α only affects its nonlinear response. In contrast, when a QD is located near a surface the strain wave generated by the optical excitation of the dot interacts after the reflection at the surface with the dot, which is still excited. Being a two-level model, there is only a GET, which is modified when the reflected wave passes the dot. From Eq. (1) it can be seen that for any excitation strength, the polarization of the QD in a half space is proportional to the linear polarization with a pulse-area dependent prefactor. Thus, the corresponding spectra are proportional to the linear absorption spectrum, which forbids negative values when the system is initially in a thermal state. For the EBT polarization, on the other hand, as a nonlinear signal there is no such restriction to positive values in the frequency domain because it cannot be interpreted as an absorption spectrum.

The fact that strain waves in the two-QD system affect only the EBT and not the GET can be visualized when considering two-QDs, where the electrostatic interactions lead to a sizable value of the biexcitonic shift $\hbar\Delta_B$. In this case the GET and EBT signals are spectrally separated. When $\hbar\Delta_B$ is larger than the total width of a typical background spectrum then the spectra resulting from GET and EBT can be analyzed separately. Figure 6 shows results for the two-QD system with distances of 10 and 20 nm, where we have assumed that $\hbar\Delta_B$ is large enough such that both spectra are completely separated. The energy is again measured with respect to the ZPL of the corresponding transition. As can be seen from Figs. 6(a)–6(d), for the present parameters this requires a value for $\hbar\Delta_B$ of at least about 10 meV. The right panel [Figs. 6(b) and 6(d)] shows the spectra corresponding to the GET signal and the left panel [Figs. 6(a) and 6(c)] shows those corresponding to the EBT signal. As expected, the GET spectra are not affected by the passage of the strain waves. Therefore, they are independent of the distance and have the same shape as the linear absorption spectrum of a

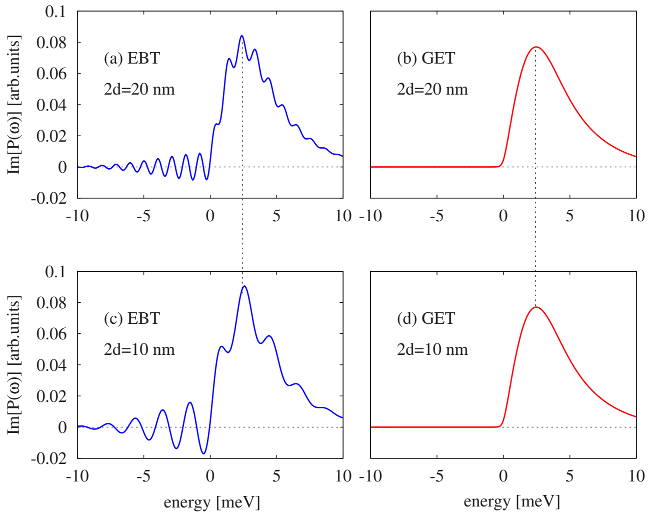

FIG. 6. (Color online) Optical spectra corresponding to the GET (right panel) and EBT (left panel) polarization for a two-QD model with different dot distances $2d$ at $T=1$ K. The biexcitonic shift $\hbar\Delta_B$ has been assumed to be large enough to completely separate GET and EBT spectra.

single two-level QD in an infinite crystal. In contrast, the EBT spectra exhibit strong spectral oscillations leading to negative values below the EBT resonance.

It is worth noting that when the carrier-phonon coupling is treated on the mean-field level then obviously identical displacement fields yield identical impacts on the optical transitions; since on this level of the theory, the coupling only depends on the displacement field and not on higher phonon correlations or carrier-phonon entanglements. The mean-field theory is often invoked when nonthermal phonons are excited in such a way that nonzero lattice displacements occur. Nonzero expectation values of the lattice displacement require nonzero phonon amplitudes $\langle\hat{b}_\xi\rangle$, where $\hat{b}_\xi$ is an annihilation operator for a phonon in mode ξ. Thus the state of the phonon system needs to exhibit at least a partial coherence. For the limiting case of a phonon field in an exact coherent state, the mean-field factorization of the carrier-phonon coupling would be exact. The fully coherent state is also the quantum-mechanical state of the phonon field that comes closest to the model of a classical sound wave, which is another rationalization of the mean-field approximation. However, for the two-QD model it has previously been shown[25] that the nonthermal phonons, that are generated by an ultrafast optical excitation, can only be in a fully coherent state when the electronic system ends up either in the biexciton or in the ground state and not in any superposition state. But in these cases the optical polarization must vanish. Consequently, in all cases where the resulting optical polarization is nonzero the phonon system is not in a fully coherent state. Thus, it can be concluded that the different impacts of strain waves on the optical properties of a two-QD system and a QD in a half space also reflect the different carrier-phonon correlations that occur in these systems. Neglecting these correlations by invoking the mean-field approximation would result in the prediction that the optical responses should be the same once the displacement fields become identical. This is, however, in clear contrast to our above findings.

Finally, let us comment on possible experiments to measure the influence of traveling-wave packets. For the single-QD system, the influence is already present in the linear response. Thus the oscillations in the spectra should be visible in single-dot absorption as well as in single-dot luminescence spectra, if the QD is sufficiently close to the surface. Because of the long exciton lifetime the phonon background at low temperatures is rather weak, however, in particular in luminescence experiments it has clearly been observed.[1] Alternatively, by using four-wave-mixing (FWM) experiments, the time domain can be probed directly and the initial decay of the signal caused by the phonon interaction has been quantitatively described by the pure dephasing model.[23] The dip in the linear polarization seen in Fig. 2(b) will be present in the time-resolved FWM signal of a single-QD and even in the time-integrated FWM signal of a QD ensemble if all QDs have the same distance from the surface. This condition will typically be fulfilled for QDs grown on a wetting layer. For the two-QD system the situation is more challenging because here only nonlinear signals are affected by the wave packet. Nevertheless, with the increasing sensitivity of single-QD spectroscopy features, such as the spectral oscillations in the EBT spectra (see Fig. 6), might become detectable. In FWM signals, essentially no signatures are expected for positive delay times neither in the time-resolved signal from a single pair of QDs nor in the time-integrated signal from an ensemble of QD pairs with fixed dot distance because of the absence of a signature in the modulus of the polarization [Fig. 2(b)]. Thus, in a FWM experiment the different behavior of the two cases, despite the fact that the strain fields traveling across the dots are essentially the same, should be clearly evident. In the two-QD case it has been found that the coherence between the ground state and the biexciton state, which is not optically active and therefore has not been discussed here, is affected by the passage of the wave packet.[34] It is known that such a biexcitonic coherence gives rise to a FWM signal at negative delay times. Therefore the time-resolved FWM signal from a single pair of QDs at negative delay times should display signatures of the wave packet traveling from one dot to the other, while from the single-QD in a half space, being essentially a two-level system, no FWM signal will be emitted for negative delay times.

IV. CONCLUSIONS

In this paper we have considered strain waves of different origins that propagate across a QD. We have analyzed the influence of these strain waves on the optical response of the QD and compared two situations: (i) a single-QD in a half space generating, by an optical excitation, a strain wave that after reflection at the surface re-enters the dot; and (ii) a two-QD model where a strain wave generated by a nearby QD travels across the dot. Surprisingly, the impacts of the strain waves on the optical polarization are qualitatively different for these two models even when the corresponding displacement fields are identical in the half space of the QD.

While the amplitude of the polarization in the two-dot model is hardly affected, the strain wave in the half-space model leaves a distinct trace in the dynamics of $|\mathbf{P}(t)|$. The impact on the phase of the polarization, on the other hand, is similar in both cases. These qualitatively different signatures, in the polarization dynamics, turned out to be independent of the detailed boundary conditions for the phonons at the surface of the half space. Both types of phonon modes, either those obeying the common stress-free surface boundary conditions or cos-phonon modes that yield the same displacement field as in the two-QD case, give rise to qualitatively the same behavior. Furthermore, it was demonstrated that the corresponding spectra are noticeably influenced by the passage of the phonon wave packets for both systems and both show strong spectral oscillations. However, also in the frequency domain, the results clearly differ for these systems. In particular the spectra for the two-QD model may exhibit negative values, which are excluded in the half-space case. The physical origin of these differences could be traced back to the fact that the phonons traveling across the dot affect the corresponding optical response via different quantum-mechanical subspaces, namely, reflected phonon wave packets couple to the single-exciton subspace while wave packets generated by a second dot enter only via their influence on the two-exciton manifold. Hence, the optical response in principle allows one to distinguish whether stress waves are originated from the same or another dot.

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[30] Note that in the present case the single-exciton Hamiltonian without phonons is already diagonal. Thus, here the transformation that was needed in Ref. 25 to diagonalize the coupling between the two single-exciton states resulting from the exchange interaction is redundant.
[31] We have used rather small dots, because the coupling to phonons increases with decreasing dot size (Ref. 2). However, in the present case where only the strain waves emitted in the z direction may return to the dot or travel across the other dot there is a strong dependence on the vertical size but only a weak dependence on the lateral size.
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