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Electron relaxation in a double quantum dot through two-phonon processes

V. N. Stavrou* and Xuedong Hu
Department of Physics, State University of New York at Buffalo, Buffalo, New York 14260, USA

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We theoretically study the relaxation of electron orbital states of a double quantum dot due to two-phonon processes. In particular, we calculate how the relaxation rates depend on the separation distance between the quantum dots, the strength of quantum dot confinement, and the lattice temperature. Enhancement of the relaxation rates by specific interdot distances and lattice temperatures, and the relative strength of different scattering channels are discussed. Our results show that although at low temperatures ($T\sim 1$ K) two-phonon processes are almost four orders of magnitude weaker compared to one-phonon processes in relaxing electron orbital states, at room temperature they are as important as one-phonon processes. Furthermore, at higher temperatures processes involving absorbing a phonon and emitting another one are more important than emitting or absorbing two phonons due to an interplay of the finite thermal occupation of phonon modes and the phonon density of state considerations.

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I. INTRODUCTION

Electronic states in artificial atomic and molecular devices, such as semiconductor single and coupled quantum dots, have been studied theoretically and experimentally for the past two decades.[1] More recently, single electrons trapped in a semiconductor double quantum dot (QD) have been suggested as candidates of quantum bits (qubits), where the location of the electron in the two dots represents the two states of the qubit.[2–8] These proposals have prompted careful experimental investigations of the electronic states in a coupled double dot[9,10] during the past few years, which in turn motivated more detailed theoretical studies of the decoherence properties of these electronic devices.[11–14] The results generally show that for the current generation devices, charge noise induced electron relaxation is more important than that due to electron-phonon coupling. However, since the latter is intrinsic while charge noise is often from extrinsic origins, it is crucial to thoroughly characterize the electron-phonon interaction induced charge relaxation in order to establish the lower limit of the quantum decoherence rate in semiconductor nanostructures.

So far all the calculations on electron relaxation due to electron-phonon interaction have focused on one-phonon processes, specifically single-phonon emission process. Here we calculate the relaxation rates due to two-phonon processes and investigate whether there exists any regime where two-phonon processes might be as important as one-phonon processes to charge relaxation. Although the current study is motivated by considerations of charge qubit based solid state quantum computing, we emphasize that the results obtained are relevant to semiconductor nanodevices, in general. Indeed, our results turn out to be more interesting at higher device temperatures, where different two-phonon processes are stronger and display unexpected crossover behaviors.

Two-phonon processes in a single three-dimensional isotropic GaAs QD were studied in the context of electron relaxation in photoluminescence experiments more than a decade ago,[15] where processes of the most interest involve a longitudinal acoustic (LA) and a longitudinal optical (LO) phonon in the bulk phonon approximation.[16] In this stuation the LO phonon density of states is singular, so that two-phonon processes could be a dominant relaxation mechanism. Here we study the two-phonon processes in the context of the relaxation of a single electron in the first excited state of a double dot, where the electron energy splitting ($\leqslant 1$ meV) is much smaller than that studied before ($\geqslant 36$ meV, when LO phonons are generally involved) so that our focus will be on two-acoustic-phonon processes. In these processes phonon density of states, qubit energy splitting, and lattice temperature together lead to interesting behaviors in the overall relaxation rates and relative strength of the different scattering channels.

The paper is organized as follows. In Sec. II we first briefly describe the electronic states we study and the electron-phonon coupling in our system. We then give the expressions for the two-phonon relaxation rates. In Sec. III we present our results, showing how the relaxation rates depend on double dot parameters such as an interdot distance and a single dot confinement, and how these rates vary with temperature. We also discuss the physical mechanisms behind these obtained behaviors. In Sec. IV we draw some conclusions.

II. THEORETICAL FORMALISM

Our model system consists of two coupled QDs separated by a distance of 2α and containing one electron. Each of the QDs is described by a two-dimensional (2D) harmonic well.[17] The single-dot one-electron wavefunctions are 2D harmonic oscillator functions[1,18] and are described in terms of the principal quantum number $n=0,1,2,\ldots$, and the angular momentum quantum number $m=0,\pm 1,\pm 2,\ldots$, as

$$\psi_{\parallel}^{(n,m)}(\tilde{\rho},\theta)=\sqrt{\frac{n!}{\pi l^2(n+|m|)!}}\,\tilde{\rho}^{|m|}e^{-\tilde{\rho}^2/2}e^{im\theta}\mathcal{L}_n^{|m|}(\tilde{\rho}^2), \quad (1)$$

where $\mathcal{L}_n^{|m|}(\tilde{\rho}^2)$ are the Laguerre polynomials, and $\tilde{\rho}=|\mathbf{r}_{\parallel}|/l$ is a scaled radius, with $l=\sqrt{\hbar/m^{*}\omega_0}$. The eigenenergies are

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$$E_{nm} = (2n + |m| + 1)\hbar\omega_0. \tag{2}$$

In our calculations the vertical confinement length is kept sufficiently small so that the vertical degree of freedom is frozen out.[18] Specifically, we have chosen a 10 nm well width, so that the first excited subband along the growth direction is about 170 meV above the ground subband. Even though excited states are more important for two-phonon processes compared to single-phonon processes, the contribution from the excited subband should still be dominated by the excited states within the ground subband (a few to a few tens of meV above the ground state). Along the growth direction we assume an infinite quantum well (QW) confinement, so that the electron wave function has the form

$$\psi_z(z) = \frac{1}{\sqrt{L_z}}\cos(\pi z/2L_z). \tag{3}$$

In a real quantum well, the electron wave function always has its tails inside the barrier materials, especially for thinner wells. For a GaAs QW, the barrier materials are either $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ or pure AlAs. Since the mass difference between Al and As atoms is greater than that between Ga and As atoms, we anticipate a slightly stronger piezoelectric coupling[19] in the barrier regions for thinner wells because of the larger wave function tail inside the barrier. In a realistic structure, however, the change should be quite small because the tail electron probability is only a small fraction of the electron wave function. Combining both considerations above (freezing out of the vertical degree of freedom and barrier effect on the interaction strength), our particular choice of the vertical confinement length and wave function form should not have any significant bearing on our results.

For two QDs that are horizontally coupled, we use a simple in-plane confinement of two parabolic wells separated by an interdot distance 2α:

$$V_c = \tfrac{1}{2}m^*\omega_0^2 \min\{(x-\alpha)^2 + y^2, (x+\alpha)^2 + y^2\}. \tag{4}$$

The single-electron wave function for the lateral direction can, in general, be expressed as a superposition of the single-dot wave functions:

$$|\Psi_{\parallel}\rangle = \sum_k C_k|\psi_{\parallel,L}^k\rangle + D_k|\psi_{\parallel,R}^k\rangle, \tag{5}$$

and the total wave function of the system of the coupled QDs can be written as

$$\Psi(\mathbf{r}) = \Psi_{\parallel}(\mathbf{r}_{\parallel})\psi_z(z). \tag{6}$$

In the present study, the in-plane wave functions for the coupled QD are calculated numerically by direct diagonalization, using parameters of a GaAs QW.

Electrons in GaAs interact with both acoustic and optical phonons. Here we do not consider contributions from the optical phonons to the electron relaxation due to the small energy splitting in the double-dot system we study. We calculate two-phonon relaxation rates caused by both deformation potential and piezoelectric interactions. The Hamiltonian that describes the electron-acoustic-phonon interactions is given by

$$H_{ep} = \sum_{\mathbf{q}} \left(\frac{\hbar}{2\rho_m V\omega_{\mathbf{q}}}\right)^{1/2} \mathcal{M}(\mathbf{q})\rho(\mathbf{q})(a_{\mathbf{q}} + a_{-\mathbf{q}}^{\dagger}), \tag{7}$$

where ρ_m is the mass density of the host material, $\omega_{\mathbf{q}}$ is the frequency of the phonon mode with wave vector $\mathbf{q}$, V is the volume of the sample, $a_{\mathbf{q}}$ and $a_{-\mathbf{q}}^{\dagger}$ are phonon annihilation and creation operators, and $\rho(\mathbf{q})$ is the electron density operator. The interaction strength $\mathcal{M}(\mathbf{q})$ is defined by

$$\mathcal{M}(\mathbf{q}) = D|\mathbf{q}| + i\mathcal{M}_p(\hat{\mathbf{q}}), \tag{8}$$

where the first term represents the deformation potential interaction with deformation constant D, and the second term, which is imaginary, describes the piezoelectric interaction. For zinc blende crystals (e.g., GaAs), the piezoelectric term $\mathcal{M}_p(\hat{\mathbf{q}})$ takes the form[19,20]

$$\mathcal{M}_p(\hat{\mathbf{q}}) = 2ee_{14}(\hat{q}_x\hat{q}_y\xi_z + \hat{q}_y\hat{q}_z\xi_x + \hat{q}_x\hat{q}_z\xi_y), \tag{9}$$

where e is the electronic charge, e_{14} is the piezoelectric constant, and ξ is the unit polarization vector of the LA phonons. Only longitudinal acoustic phonons are considered in this study as they are most strongly coupled to the electron.[19]

Using the second-order perturbation theory, scattering rates due to the emission and/or absorption of two LA phonons can be obtained (these are separate contributions to the electron relaxation, in addition to the single-phonon contributions)

$$\Gamma_{++} = \frac{\pi}{\hbar}\sum_{\mathbf{q},\mathbf{k}} \left| \sum_s \left(\frac{M_{\mathbf{q}}^{is}M_{\mathbf{k}}^{sf}}{E_i - E_s - E_{\mathbf{q}}} + \frac{M_{\mathbf{k}}^{is}M_{\mathbf{q}}^{sf}}{E_i - E_s - E_{\mathbf{k}}} \right) \right|^2 \times (N_{\mathbf{q}} + 1)(N_{\mathbf{k}} + 1)\delta(E_i - E_f - E_{\mathbf{q}} - E_{\mathbf{k}}), \tag{10}$$

$$\Gamma_{--} = \frac{\pi}{\hbar}\sum_{\mathbf{q},\mathbf{k}} \left| \sum_s \left(\frac{M_{\mathbf{q}}^{is}M_{\mathbf{k}}^{sf}}{E_i - E_s + E_{\mathbf{q}}} + \frac{M_{\mathbf{k}}^{is}M_{\mathbf{q}}^{sf}}{E_i - E_s + E_{\mathbf{k}}} \right) \right|^2 \times N_{\mathbf{q}}N_{\mathbf{k}}\delta(E_i - E_f + E_{\mathbf{q}} + E_{\mathbf{k}}), \tag{11}$$

$$\Gamma_{+-} = \frac{2\pi}{\hbar}\sum_{\mathbf{q},\mathbf{k}} \left| \sum_s \left(\frac{M_{\mathbf{q}}^{is}M_{\mathbf{k}}^{sf}}{E_i - E_s - E_{\mathbf{q}}} + \frac{M_{\mathbf{k}}^{is}M_{\mathbf{q}}^{sf}}{E_i - E_s + E_{\mathbf{k}}} \right) \right|^2 \times N_{\mathbf{k}}(N_{\mathbf{q}} + 1)\delta(E_i - E_f - E_{\mathbf{q}} + E_{\mathbf{k}}), \tag{12}$$

where the matrix elements are defined as in Ref. 15. Here emission of two phonons (+LA+LA), absorption of two phonons (−LA−LA), and emission and absorption of one phonon each (+LA−LA, for overall emission, and −LA +LA, for overall absorption) are indicated by subscripts $++$, $--$, $+-$, and $-+$ [Γ_{-+} takes exactly the same form as Γ_{+-}. The difference is that for Γ_{+-} in Eq. (12) $E_{\mathbf{q}} > E_{\mathbf{k}}$, while for Γ_{-+}, $E_{\mathbf{q}} < E_{\mathbf{k}}$]. The later three processes vanish at $T = 0$ K because they involve absorption of phonons. However, since experimental temperature is often in the same order as the qubit energy splitting, we include all two-phonon processes in our calculation. Indices i and f represent the initial and final electronic states, which are the first excited and ground double-dot states, respectively. Index s refers to intermediate electronic states which are among the 13 higher energy states we include in the present calculation (the summation over s excludes the initial and final states, and the sum converges).

TABLE I. Material parameters (Ref. 20) used in our calculation (m_e is the bare electron mass).

	m/m_e	D (eV)	c_s (m/s)	ρ (K g/m^3)	e_{14} (V/m)	ϵ_s	ϵ_∞
GaAs	0.067	8.6	3700	5300	1.38×10^9	12.9	10.89

The matrix elements are calculated as in Ref. 14. $N_{\mathbf{k}}$ is the Bose-Einstein distribution function for the **k** phonon mode with energy $E_{\mathbf{k}}=\hbar\omega_{\mathbf{k}}$. In other words, in the calculation of the matrix elements the phonons are treated as thermal. The integrals over **k** and **q** in Eqs. (10)–(12) are calculated by the Monte Carlo method. It is worth emphasizing that the abovementioned scattering rates are separate contributions to the charge qubit energy relaxation rate.

Throughout the paper the QW width is fixed at $2L_z=10$ nm and the material parameters are taken from Bruus *et al.*[20] Table I presents the material parameters that are used in the current calculations.

III. RESULTS AND DISCUSSIONS

At low temperatures, which are generally the operating temperature of the current generation nanoelectronic devices, two-phonon absorption processes should be much weaker than emission processes. Therefore, we first evaluate the rates for an electron in the first excited state of the double dot as the initial state to relax to the final ground state via two-phonon processes. Two channels are included here: (a) emission of two LA phonons (+LA+LA) and (b) emission of a high energy LA phonon and absorption of a lower energy LA phonon (+LA−LA).

In Figs. 1 and 2 we present the calculated relaxation rates due to the two overall emitting processes as functions of half interdot distance α and single-dot confinement energy $\hbar\omega_0$. For both figures the lattice temperature is fixed at $T=1$ K, a usual operating temperature for gated semiconductor quantum dots. The results presented in Fig. 1 have some important features. The most prominent is that the order of magnitude for both processes is much smaller compared to the one-phonon processes.[14] Therefore, at low temperatures it should be sufficient to consider only the single-phonon processes when studying phonon-induced electron relaxation and the related charge qubit decoherence.

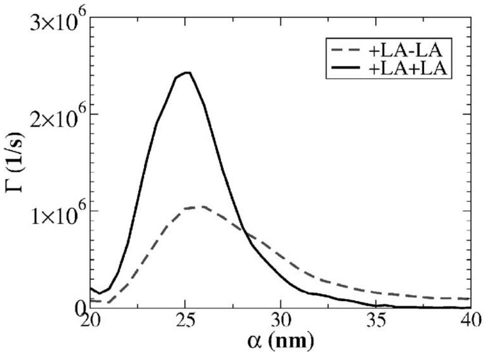

FIG. 1. Relaxation rates of an electron in a double dot due to two different two-phonon processes (+LA+LA and +LA−LA) as functions of the half interdot distance α. The confinement strength is $\hbar\omega=3$ meV, lattice temperature $T=1$ K, and QW width $2L_z=10$ nm. The oscillatory behavior of the +LA+LA process at a larger α is an artifact of the integration process.

Both curves in Fig. 1 display a clear peak at an intermediate interdot distance α. This feature is an interplay between the phonon density of states (DOS) and the magnitude of the electron-phonon matrix element. When interdot distance increases, the electron energy splitting decreases monotonically. This leads to a decreasing number of phonon modes that can be involved in the relaxation process since the phonon DOS varies as ω^2 as a function of phonon frequency. On the other hand, the magnitude of the electron-phonon matrix element increases with increasing α (and eventually saturates).[14] These two opposite trends dictate that a maximum should develop at an intermediate interdot distance for the electron relaxation rate.

Another interesting feature of Fig. 1 is the crossover between the two curves representing the +LA−LA and +LA+LA processes. The physics behind this crossover is more complicated than the maxima of the two curves at an intermediate value of α. Qualitatively, for +LA+LA processes, both phonons involved have to have energies smaller than the qubit energy splitting, while for +LA−LA processes the emitted phonon has to have energy larger than the qubit energy splitting. As bulk acoustic phonons have a DOS proportional to ω^2, the +LA−LA processes should be favored over +LA+LA processes in terms of the DOS consideration. On the other hand, +LA−LA processes involve absorption of one phonon, which is thermally constrained [a phonon thermal population factor of $(N_{\mathbf{q}}+1)N_{\mathbf{k}}$ versus $(N_{\mathbf{q}}+1)(N_{\mathbf{k}}+1)$ for the two-phonon emission]. The energy of the absorbed

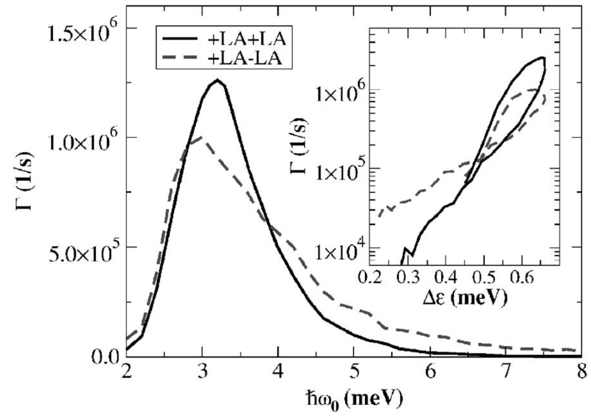

FIG. 2. Relaxation rates of an electron due to two different two-phonon (overall emission) processes (+LA+LA and +LA−LA) as a function of the confinement strength. The same relaxation rates as a function of the charge qubit energy splitting $\Delta\varepsilon$ are shown in the inset. Here the half interdot distance is fixed at $\alpha=25$ nm, and the lattice temperature is $T=1$ K.

phonon has to be relatively small compared to the thermal energy (in the current case ~0.1 meV), which also means a relatively small phonon DOS as well. This argument favors the +LA+LA processes. Furthermore, we mentioned above that the electron-phonon interaction matrix element increases with increasing α. Therefore, which process yields a higher relaxation rate is determined by considering all three factors of lattice temperature, the electron-phonon matrix element, and phonon DOS. For example, when the half interdot distance $\alpha > 35$ nm, the splitting between the two energy levels of the double-dot system is less than 0.2 meV so that +LA +LA processes (represented by the solid curve in Fig. 1) are strongly suppressed due to the phonon DOS consideration. On the other hand, even though $T=1$ K is a low temperature, it is still finite (thermal energy ~0.1 meV), so that populations of phonons with energy $\lesssim 0.1$ meV are finite and absorption of such phonons is possible, which in turn leaves the +LA−LA relaxation channel open (represented by the dashed curve in Fig. 1). In essence, the quadratically larger DOS of the emitted phonon in the +LA−LA process is the most important reason why the +LA−LA process is faster at larger interdot distance α.

The results presented in Fig. 2 are obtained at a fixed interdot distance of $2\alpha = 50$ nm. Here the maxima of the two relaxation rate curves are mostly determined by the electron energy splitting (as shown in the inset) and the phonon DOS consideration. The matrix element plays a less important role because it is not as sensitive to the single-dot confinement energy as it is to interdot distance. Again the two rates have a crossover, with +LA+LA processes faster at larger electron energy splittings while +LA−LA processes faster for smaller energy splittings. The physical mechanism is similar to that in Fig. 1: as the energy splitting between the electronic levels varies, so does the involved phonon DOS. In the inset we plot the same relaxation rates as functions of the energy splitting $\Delta\varepsilon$ between the two double-dot levels. Here $\Delta\varepsilon = \Delta\varepsilon(\omega_0)$ is calculated as we vary the confinement energy $\hbar\omega_0$ and fix the half interdot distance at 25 nm. The loop structure in the inset of Fig. 2 reflects a nonmonotonic dependence of the energy splitting $\Delta\varepsilon$ on the confinement strength $\hbar\omega_0$. In other words, $\Delta\varepsilon$ could be small when $\hbar\omega_0$ is both small (when the double-dot states are the same as the states of a large single dot, so that energy levels are dense) and large (when the inter dot overlap is small). The curve shape in the inset is also consistent with what is presented in the main curve, where each relaxation rate value correpsonds to two different confinement energies, which in turn generally correspond to different double-dot energy splittings.

In Fig. 3 we present the electron relaxation rates as functions of the lattice temperature for all the one-phonon and two-phonon processes. At $T=1$ K, the strongest two two-phonon processes are the +LA+LA and +LA−LA overall emission processes, but the corresponding relaxation rates are more than three orders of magnitude smaller than the one-phonon emission process. The other two two-phonon processes are thermally suppressed because of the involvement of absorption of higher energy phonon(s). On the other hand, at room temperature (corresponding to a thermal energy in the order of 30 meV, much larger than the electronic energy splitting of <1 meV), the strongest two-phonon process, the +LA−LA (overall emission) process is only one order of magnitude smaller than the one-phonon processes (both emission and absorption). In other words, if the electron orbital states of a double quantum dot are to be used for an electronic device (with low charge fluctuations) at high temperatures such as room temperature, two-phonon relaxation processes should also be taken into consideration.

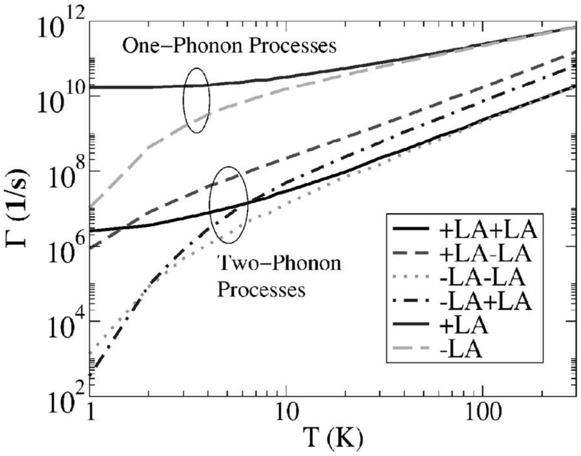

FIG. 3. Relaxation rates of an electron due to all the two-phonon processes [+LA+LA, +LA−LA (overall emission), −LA−LA (two-phonon absorption), and −LA+LA (overall absorption)] and one-phonon processes [+LA (one-phonon emission) and −LA (one-phonon absorption)] as functions of the lattice temperature. Here the confinement strength is $\hbar\omega = 3$ meV, and the half interdot distance is $\alpha = 25$ nm.

In Fig. 3 at intermediate temperatures, the relaxation rates have crossovers between the one-phonon-emission–one-phonon-absorption (+LA−LA and −LA+LA) processes and the two-phonon absorption or emission (+LA+LA or −LA −LA) processes. The physical mechanism for these crossovers is similar to what we have discussed for Figs. 1 and 2 in that the thermal suppression of the higher energy phonon populations (which are only involved in +LA−LA and −LA+LA processes and have a higher density of state) is gradually removed as temperature increases, while the bulk phonon DOS of the system is unchanged. This removal of the thermal constraint strongly favors the +LA−LA and −LA+LA processes at higher temperatures.

At room temperature the +LA+LA and −LA−LA processes have almost the same relaxation rates because the thermal factors in Eqs. (10) and (11) are approximately the same for phonons with an energy smaller than 1 meV (recall that for two-phonon emission or absorption processes each of the phonons involved should have an energy smaller than the electron energy splitting). Similarly, +LA−LA (overall emission) and −LA+LA (overall absorption) processes also have similar rates at high temperatures. The discrepancy is mostly due to the involvement of higher energy phonons in these processes, for which the factors N_k and (N_k+1) could be sufficiently different.

For two-phonon processes both deformation potential and piezoelectric interactions are included in the matrix element—they cannot be treated separately as in one-phonon processes. According to our one-phonon calculations in Ref.

14, for larger energy splittings between the energy levels (>0.5 meV) the deformation potential (whose matrix element is proportional to $\sqrt{\omega_q}$) is more important than the piezoelectric interaction (whose matrix element is proportional to $1/\sqrt{\omega_q}$), while for smaller energy splittings the piezoelectric interaction dominates. This result should carry over to two-phonon processes, so that two-phonon emission or absorption processes should mostly be dominated by the piezoelectric interaction as long as the qubit energy splitting is not too large. However, for the +LA−LA type of processes, even when the energy splitting between the initial and final electronic levels is small, the energies of the two phonons involved in the transition do not have to be small, especially at higher temperature. For example, if the energy splitting between the two levels in consideration is 0.1 meV and temperature is in the order of 1 K so that population of higher energy phonons ($\omega > 0.1$ meV) is suppressed, the piezoelectric interaction should dominate in all the one- and two-phonon processes. On the other hand, if the temperature is raised to 100 K, so that absorption of meV phonons becomes uninhibited, the deformation potential should be more important in the +LA−LA and −LA+LA processes. Therefore, the deformation potential interaction could play a more important role in some of the two-phonon processes than in one-phonon processes for small energy splitting situations.

IV. CONCLUSIONS

In this study we have investigated two-acoustic-phonon processes (including both phonon emissions and absorptions) induced electron orbital relaxation in a semiconductor double quantum dot. Our results show interesting dependence of the relaxation rates on the lattice temperature and system configuration parameters such as the interdot distance and single-dot confinement energy. We find that acoustic phonon density of states, electron-phonon coupling matrix elements, and phonon thermal distributions together lead to crossovers between different two-phonon scattering channels. The two-phonon processes are, in general, much weaker than one-phonon processes at relatively low temperatures, although their magnitudes become similar at the room temperature.

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*Electronic address: vasileios-stavrou@uiowa.edu

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