

Electron-phonon interaction in quantum dots: A solvable model

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The relaxation of electrons in quantum dots via phonon emission is hindered by the discrete nature of the dot levels (“phonon bottleneck”). In order to clarify the issue theoretically we consider a system of N discrete fermionic states (dot levels) coupled to an unlimited number of bosonic modes with the same energy (dispersionless phonons). In analogy to the Gram-Schmidt orthogonalization procedure, we perform a unitary transformation into new bosonic modes. Since only $N(N+1)/2$ of them couple to the fermions, a numerically exact treatment is possible. The formalism is applied to a GaAs quantum dot with only two electronic levels. If close to resonance with the phonon energy, the electronic transition shows a splitting due to quantum mechanical level repulsion. This is driven mainly by one bosonic mode, whereas the other two provide further polaronic renormalizations. The numerically exact results for the electron spectral function compare favorably with an analytic solution based on degenerate perturbation theory in the basis of shifted oscillator states. In contrast, the widely used self-consistent first-order Born approximation proves insufficient in describing the rich spectral features.

I. INTRODUCTION

Since the development of quantum well lasers there have been continuous attempts to manufacture laser structures with even more reduced dimensions. The idea behind was to increase the efficiency by enhancing the density of states. However, zero-dimensional quantum structures (quantum dots) are characterized by a discrete spectrum, and the recombination probability does not depend on the radiative rate alone. Rather, the relaxation pathway into the ground state becomes decisive. Looking at the nearly monoenergetic longitudinal-optical (LO) phonons, an efficient relaxation between two dot levels seems to be possible only if level distance and LO energy match (resonance condition). This type of argument has been called *phonon bottleneck*.^{1,2}

From the experimental side there is an ongoing intense debate on whether or not the phonon bottleneck is seen in the data.³ However, the recently found ground-state lasing in quantum dots under cw conditions seems to prove that the phonon bottleneck is not an obstacle when trying to increase the laser efficiency by dimensional reduction.⁴

Nevertheless the theoretical concepts are still controversial. Obviously, the bottleneck argument relies on the assumption of strict energy conservation in the electron-phonon scattering, as dictated by Fermi’s golden rule. A next step towards a realistic description seems to incorporate the intrinsic lifetime broadening of dot levels. Král and Khás⁵ went along this way by calculating the complex electron self-energy due to the LO-phonon interaction. They claimed that the convolution of initial and final state spectral functions gives rise to a broadening that is able to circumvent the phonon bottleneck. Arakawa and co-workers^{6,7} have treated the electronic transition and the LO modes in closed form by wave function evolution. They pointed out that the final decay of the LO phonon into acoustic phonons is decisive for

the relaxation process. A different argument includes Auger-like electronic excitation for overcoming the sharp energy selection inherent to the LO-phonon relaxation.^{8–10}

A more general question concerns the appropriate theoretical tools for describing relaxation in zero-dimensional systems. Nonequilibrium Green’s functions are often too tedious to be used in realistic models. Therefore one is tempted to look for the one-particle Green’s functions and their broadening as a signature for relaxation.^{5,11} However, standard self-energy approaches as the self-consistent first-order Born approximation⁵ have to be questioned since they rely implicitly on the existence of an electronic continuum that is missing in zero-dimensional systems.

In this paper we want to look closely at this question and to qualify the standard approximation schemes (as the self-consistent Born approximation) in application to quantum dots. To compare with, we present results from an exact diagonalization of the electron-phonon Hamiltonian. This can be achieved even for an unlimited number of phonon modes provided they have *no dispersion*, and uses a unitary transformation among the phonons. If N electronic dot states (fermions) are considered, only $N(N+1)/2$ of the new modes (bosons) couple to the electrons, and for moderate numbers of N the transformed Hamiltonian can be easily diagonalized numerically. An upper limit of the boson occupation numbers can be fixed in accordance with temperature. For $N=1$, this exact solution is known for a long time as *independent boson model*.¹² Here we present the extension to a finite number of levels with the important interlevel coupling (phonon transition). A *single* boson mode in resonance with an equidistant series of electronic levels has been treated exactly in Ref. 13 predicting the phonon staircase effect.

A related problem is the electron-phonon coupling in semiconductor point defects. Both the internal defect transi-

tions in the near infrared and the phonon satellites of interband transitions show a rich spectrum. Even away from strict resonance with an electronic transition, the measured transition energies differ from the bare phonon value. This has been called the LO-phonon–donor bound state, similar to the exciton-phonon complex introduced earlier.¹⁴ In first attempts for a quantitative understanding, perturbation theory has been used.¹⁵ The crystallographic symmetry of the defect dictates what kind of lattice distortion (local phonon mode) couples to the electronic transitions. Taking into account only a few of these symmetry-adapted lattice modes, a full numerical diagonalization is possible nowadays.¹⁶ This is in particular important when dealing with strongly polar material. We show that semiconductor quantum dots behave similarly with respect to the lattice coupling, underlining once more that the quantum dot behaves as a kind of mesoscopic atom. Note, however, the quite different length scales involved. Whereas the local modes in the defect problem are constructed using large parts of the Brillouin zone, only a minor part around the Γ point is involved in the quantum dot case. Consequently, the LO-phonon dispersion can be safely neglected here.

In Sec. II the method is outlined and applied to the most simple but nontrivial case of two dot levels. Results for the spectral function are given in Sec. III. It shows a kind of avoided level crossing if level distance and phonon energy nearly coincide, which resembles the phonon-polariton feature. Still, the exact spectral function consists of a series of sharp lines. The self-consistent first-order Born approximation fails in this respect by exhibiting broad spectral features. However, we are able to derive a simple analytical approximation that almost coincides with the exact results. This employs nondegenerate perturbation theory for those electron-phonon states which are strongly coupled near resonance. It is called the rotating-wave approximation since it resembles a similar treatment of the Jaynes-Cummings model in quantum optics. Some consequences of the present work on the general description of relaxation in quantum dots are given as well, and conclusions are drawn in Sec. IV. In the Appendix, the coupling constants are calculated adopting parameter values for an idealized GaAs quantum dot with parabolic confinement.

II. THE MODEL AND THE TRANSFORMATION

Let us consider N discrete electronic levels $i=0, \dots, N-1$ coupled to M phonon modes $\mathbf{q}_1, \dots, \mathbf{q}_M$ of fixed energy $\hbar\omega_0$. The Hamiltonian reads

$$H = \sum_i \epsilon_i c_i^\dagger c_i + \sum_{\mathbf{q}} \hbar\omega_0 b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \sum_{i,j,\mathbf{q}} M_{\mathbf{q}}^{i,j} (b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger) c_i^\dagger c_j. \quad (1)$$

Here, the $c_i(c_i^\dagger)$ denote the fermionic creation (annihilation) operators, respectively, and $b_{\mathbf{q}}, b_{\mathbf{q}}^\dagger$ are the corresponding bosonic operators. The coupling constants between phonons and electrons, $M_{\mathbf{q}}^{i,j}$, depend explicitly on the fermionic states involved (transitions between dot levels). For H to be Hermitian, $(M_{\mathbf{q}}^{i,j})^* = M_{-\mathbf{q}}^{j,i}$ must hold. For simplicity, the spin is neglected because it is conserved by the electron-phonon interaction.

Since all M bosonic modes couple to the electronic levels, a straightforward diagonalization is not feasible. We proceed by mapping the phonon operators $\{b_{\mathbf{q}}\}$ onto a new set of bosonic operators $\{B_\lambda\}$, with the goal that only a limited number couples to the electrons.

We start with an arbitrary linear combination of the operators $b_{\mathbf{q}}$ written as A_λ with $\lambda = 1, \dots, M$, which also span the bosonic Hilbert space. Following the well-known Gram-Schmidt orthogonalization procedure,

$$\tilde{B}_\lambda = A_\lambda - \sum_{\alpha=1}^{\lambda-1} [A_\lambda, B_\alpha^\dagger] B_\alpha, \quad B_\lambda = \tilde{B}_\lambda / \sqrt{[\tilde{B}_\lambda, \tilde{B}_\lambda^\dagger]}, \quad (2)$$

we arrive at a new set B_λ whose members obey the canonical Bose commutation relations, $[B_\lambda, B_{\lambda'}] = \delta_{\lambda,\lambda'}$. Then, the transformation matrix U with $B_\lambda = \sum_{\mathbf{q}} U_{\lambda,\mathbf{q}} b_{\mathbf{q}}$ is unitary, and we have

$$\sum_{\lambda} B_\lambda^\dagger B_\lambda = \sum_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}}. \quad (3)$$

Since the bosonic modes have the *same energy*, the last relation shows that the free boson term in the Hamiltonian equation (1) remains diagonal.

The first linear combinations are chosen as

$$A_{\lambda(i,j)} = \sum_{\mathbf{q}} M_{\mathbf{q}}^{i,j} b_{\mathbf{q}}, \quad (4)$$

where $\lambda(i,j)$ runs over the $N(N+1)/2$ pairs (i,j) with $i \geq j$. The remaining A_λ can be taken arbitrarily but linearly independent. It follows from the prescribed one-to-one mapping that the electron-phonon interaction contains only the restricted set $\lambda(i,j)$ of the new operators. This reduces the numerical labor enormously since the relevant Hilbert space now contains N fermionic and only $N(N+1)/2$ bosonic degrees of freedom.

A. Reduction to a two-level system

The model can be used to describe electrons in a quantum dot that are coupled to LO phonons. Under the assumption that the third electronic level is energetically well above the lowest two, we will limit the number of states to $N=2$. Choosing $A_1 = \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,0} b_{\mathbf{q}}$, $A_2 = \sum_{\mathbf{q}} M_{\mathbf{q}}^{0,0} b_{\mathbf{q}}$, $A_3 = \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,1} b_{\mathbf{q}}$ we accomplish that the transition matrix element couples only to *three* non trivial bosonic modes. We obtain

$$\begin{aligned} H = & \epsilon_0 c_0^\dagger c_0 + \epsilon_1 c_1^\dagger c_1 + \sum_{\lambda} \hbar\omega_0 B_\lambda^\dagger B_\lambda + (C_1 B_1 + C_1^* B_1^\dagger) (c_1^\dagger c_0 \\ & + c_0^\dagger c_1) + (C_2 B_1 + C_2^* B_1^\dagger + C_3 B_2 + C_3^* B_2^\dagger) c_0^\dagger c_0 \\ & + (C_4 B_1 + C_4^* B_1^\dagger + C_5 B_2 + C_5^* B_2^\dagger + C_6 B_3 + C_6^* B_3^\dagger) c_1^\dagger c_1 \end{aligned} \quad (5)$$

with the six coupling constants C_i , which follow from the transformation (2). If we further assume that the electronic wave functions in the quantum dot exhibit a well-defined parity, the constants C_2 and C_4 vanish, and B_1 couples only to the transition $0 \rightarrow 1$. The model will therefore show prominent features of the Jaynes-Cummings model¹⁷ that has been

introduced to describe a single photon mode coupled to an atomic transition. In the present case, however, the detuning has to be defined as $\Delta = \hbar\omega - (\epsilon_1 - \epsilon_0)$. Further details regarding the explicit calculation of the coupling constants C_i are given in the Appendix. We adopt a parabolic confinement potential with extensions $y_0 = z_0$ and $x_0 > y_0$, having in mind an anisotropic harmonic quantum dot. In this case, all the lowest dot levels have equal energy separation, and the mentioned truncation to just two levels is not realistic. Nevertheless it will be applied here to keep the numerics at a reasonable level.

By means of an appropriate rotation of the operators B_2 and B_3 , i.e.,

$$\begin{aligned} B_2 &\leftarrow [(C_5 - C_3)B_2 + C_6 B_3]/\gamma, \\ B_3 &\leftarrow [-C_6^* B_2 + (C_5^* - C_3^*)B_3]/\gamma, \end{aligned} \quad (6)$$

with $\gamma^2 = |C_5 - C_3|^2 + |C_6|^2$, we compel the new mode B_3 to couple only to the fermionic particle number operator $\mathcal{N} \equiv c_0^\dagger c_0 + c_1^\dagger c_1$. If we leave out the bosonic modes that do not couple to the fermionic levels at all, we obtain

$$\begin{aligned} H = & \epsilon_0 c_0^\dagger c_0 + \epsilon_1 c_1^\dagger c_1 + \hbar\omega_0 B_1^\dagger B_1 \\ & + (C_1 B_1 + C_1^* B_1^\dagger)(c_1^\dagger c_0 + c_0^\dagger c_1) \\ & + \hbar\omega_0 B_2^\dagger B_2 + (\gamma B_2 + \gamma B_2^\dagger)c_1^\dagger c_1 - (\eta^* B_2 + \eta B_2^\dagger)\mathcal{N} \\ & + \hbar\omega_0 B_3^\dagger B_3 - (\kappa^* B_3 + \kappa B_3^\dagger)\mathcal{N}, \end{aligned} \quad (7)$$

with the new parameters

$$\eta = C_3^*(C_3 - C_5)/\gamma, \kappa = C_3^* C_6^*/\gamma. \quad (8)$$

The Hamiltonian conserves the electron number, $[H, \mathcal{N}] = 0$, and consequently the Hilbert space can be decomposed according to the electron number (zero, one, or two). In the subspace of zero or two fermions, the Hamiltonian can be diagonalized easily because the nondiagonal transitions $0 \leftrightarrow 1$ are impossible here. More demanding is the subspace of one fermion, which can, however, be rationalized a lot by introducing shifted operators

$$\mathcal{B}_2 = B_2 - \eta\mathcal{N} + \gamma c_1^\dagger c_1, \mathcal{B}_3 = B_3 - \kappa\mathcal{N}. \quad (9)$$

Note that from Eq. (9) onward, the LO energy $\hbar\omega_0$ is taken as unit of energy in the remainder of this section. We want to stress that the shifted bosonic operators \mathcal{B}_2 and \mathcal{B}_3 still obey the canonical commutation rules, but do not commute with the fermion operators. The Hamiltonian equation (7) is now represented as

$$\begin{aligned} H = & \epsilon_0 c_0^\dagger c_0 + \tilde{\epsilon}_1^{\mathcal{N}} c_1^\dagger c_1 - E_p^{\mathcal{N}} + B_1^\dagger B_1 + \mathcal{B}_2^\dagger \mathcal{B}_2 + \mathcal{B}_3^\dagger \mathcal{B}_3 \\ & + (C_1 B_1 + C_1^* B_1^\dagger)(c_1^\dagger c_0 + c_0^\dagger c_1), \end{aligned} \quad (10)$$

with the modified energies depending of the number of particles

$$\tilde{\epsilon}_1^{\mathcal{N}} = \epsilon_1 + 2 \operatorname{Re}(\gamma\eta^*)\mathcal{N} - |\gamma|^2, \quad (11)$$

$$E_p^{\mathcal{N}} = (|\eta|^2 + |\kappa|^2)\mathcal{N}^2. \quad (12)$$

The left-hand side of Fig. 1 shows the (dimensionless)

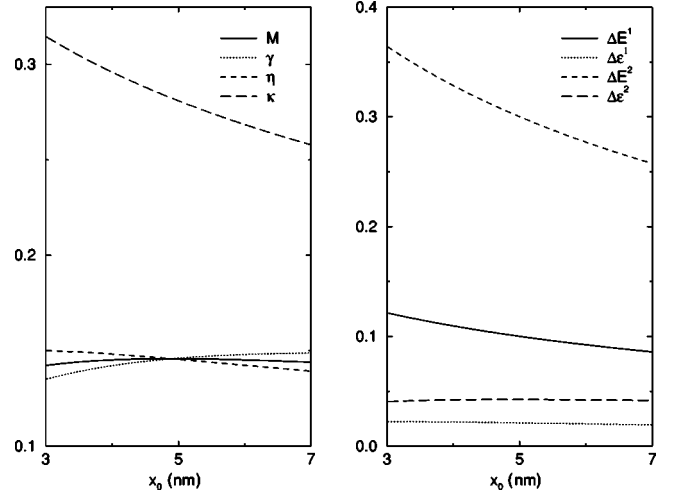


FIG. 1. Left: The parameters C_1 , γ , η , and κ in units of the LO energy as a function of the dot size x_0 . Right: The relative polaronic shifts ΔE^N and the relative shifts of the level spacing $\Delta \epsilon^N$.

parameters calculated for a parabolic confinement potential with extensions of $y_0 = z_0 = 3$ nm as a function of x_0 . All input parameters refer to GaAs, e.g., $\hbar\omega_0 = 36.7$ meV (see the Appendix). The coupling constants are almost independent of the dot size and of order $0.1\hbar\omega_0$. This value of 3.6 meV compares favorably with the polaron shift of electrons in bulk GaAs, 2.3 meV.

The right-hand side of Fig. 1 shows the polaronic shift due to the phonons B_2 and B_3 , Eq. (12), and the renormalization of the level spacing, Eq. (11). Since both depend on the number of electrons present, we display the relative shifts when adding one electron, $\Delta E^N = E_p^N - E_p^{N-1}$ and $\Delta \epsilon^N = \tilde{\epsilon}_1^N - \tilde{\epsilon}_1^{N-1}$.

B. Solution of the model

The eigenvectors $|N; n_1, n_2, n_3\rangle$ for the N -electron Hilbert space with $N=0,2$ are simple and given by

$$\begin{aligned} |0; n_1, n_2, n_3\rangle &\equiv |V\rangle |n_1\rangle_0 |n_2\rangle_0^2 |n_3\rangle_0^3, \\ |2; n_1, n_2, n_3\rangle &\equiv c_1^\dagger c_0^\dagger |V\rangle |n_1\rangle_0 |n_2\rangle_{-2\eta+\gamma}^2 |n_3\rangle_{-2\kappa}^3, \end{aligned} \quad (13)$$

with the corresponding eigenvalues

$$E_{n_1, n_2, n_3}^N = (\epsilon_0 + \tilde{\epsilon}_1^2 - E_p^2)N/2 + n_1 + n_2 + n_3. \quad (14)$$

Here, $|V\rangle$ denotes the electron vacuum, and

$$|n\rangle_\alpha^\lambda \equiv (n!)^{-1/2} (B_\lambda^\dagger + \alpha)^n |0\rangle_\alpha^\lambda \quad (15)$$

are the shifted oscillator eigenstates (or coherent states) whose vacuum is defined as $(B_\lambda + \alpha)|0\rangle_\alpha^\lambda = 0$. We will omit the upper index $\lambda = 1, 2, 3$ of the bosonic states from now on since confusions are unlikely.

Considering the $N=1$ Hilbert space the Hamiltonian can be easily diagonalized numerically since only *two* bosonic modes are involved. An approximate analytical solution is possible as well, where the transition matrix element is treated in degenerate perturbation theory, using the coherent state equation (15) as basis. In order to show the basic features of this analytic solution we neglect for the moment the

third bosonic mode \mathcal{B}_3 , which reduces the number of quantum numbers that have to be kept track of. Since this mode commutes with the rest of the system, it can easily be incorporated afterwards. In the same spirit we neglect the polaronic shift (12) and the shift of the level spacing (11), which are restored, however, in the numerics.

In the resonant situation (zero detuning Δ) and neglecting terms of the order $\gamma^2 C_1$, *degenerate* perturbation theory leads to the level repulsion (or avoided level crossing)

$$E_{n_1, n_2}^{\pm} = \epsilon_0 + n_1 + n_2 \pm \sqrt{n_1} |{}_0\langle n_2 | n_2 \rangle_{\gamma} C_1| \quad (16)$$

for $n_1 \geq 1$, $n_2 \geq 0$. The overlap integrals of two displaced oscillators appearing in Eq. (16) are also known as Franck-Condon factors, which were first introduced in the theory of excited molecules. More generally they are given by

$$\begin{aligned} {}_0\langle n | m \rangle_{\gamma} &= \frac{\gamma^{n+m}}{\sqrt{n!m!}} e^{-\gamma^2/2} \sum_{k=0}^{\min(n,m)} (-1)^{k+n} \\ &\times \frac{1}{k!} \frac{n!}{(n-k)!} \frac{m!}{(m-k)!} \gamma^{-2k} \end{aligned} \quad (17)$$

and are related to the associated Laguerre polynomials. Since the Franck-Condon factors ${}_0\langle n_2 | n_2 \rangle_{\gamma}$ are oscillating as functions of n_2 we find a complex level structure if many bosons are present, i.e., at elevated temperatures. Introducing a small external broadening will lead to a continuous but still highly structured spectral function.

The energy splitting of Eq. (16) can be recovered in a truncated Hamiltonian where only the nearly resonant transitions $0 \rightarrow 1$ with phonon absorption and $1 \rightarrow 0$ with phonon emission are kept,

$$\begin{aligned} H^{RW} &= \epsilon_0 c_0^{\dagger} c_0 + \epsilon_1 c_1^{\dagger} c_1 + B_1^{\dagger} B_1 + B_2^{\dagger} B_2 + C_1 B_1 c_1^{\dagger} c_0 \\ &+ C_1^* B_1^{\dagger} c_0^{\dagger} c_1. \end{aligned} \quad (18)$$

In analogy to the optical equivalent in the Jaynes-Cummings model, we use the term *rotating-wave approximation* (RW). H^{RW} can be represented by 2×2 matrices¹⁸ in the basis

$$\begin{aligned} |n_1, n_2\rangle^0 &\equiv c_0^{\dagger} |V\rangle |n_1\rangle_0 |n_2\rangle_{-\eta}, \\ |n_1, n_2\rangle^1 &\equiv c_1^{\dagger} |V\rangle |n_1 - 1\rangle_0 |n_2\rangle_{-\eta + \gamma}. \end{aligned} \quad (19)$$

Notice that the shift of \mathcal{B}_2 depends on the electronic level, a feature that gives rise to the Franck-Condon factors. The corresponding eigenfunctions are

$$|n_1, n_2\rangle^{\pm} = c_{n_1, n_2}^{\pm} |n_1, n_2\rangle^0 \pm c_{n_1, n_2}^{\mp} |n_1, n_2\rangle^1 \quad (20)$$

with the eigenvalues

$$E_{n_1, n_2}^{\pm} = \frac{1}{2} (\epsilon_0 + \epsilon_1) + n_1 + n_2 - 1/2 \pm R_{n_1, n_2} / 2, \quad (21)$$

with the Rabi splitting $R_{n_1, n_2}^2 = \Delta^2 + n_1 |{}_0\langle n_2 | n_2 \rangle_{\gamma} C_1|^2$ and the weight factors $c_{n_1, n_2}^{\pm} = [(R_{n_1, n_2} \pm \Delta) / 2R_{n_1, n_2}]^{1/2}$.

In this approximation, H^{RW} has been mapped onto the Jaynes-Cummings model plus an additional bosonic mode that merely renormalizes the coupling constant $C_1 \rightarrow {}_0\langle n_2 | n_2 \rangle_{\gamma} C_1$.

III. THE SPECTRAL FUNCTION

In this section we calculate the spectral function of our system. We contrast two different approaches and also provide the full solution via numerical diagonalization of a matrix spanned by only two bosonic modes.

A. Analytic expressions

The spectral function of the electron-phonon Hamiltonian equation (1) is usually calculated by means of finite-temperature Green's functions (Chap. 6 in Ref. 12). The self-consistent first-order Born approximation for the (retarded) self-energy is often used provided the assumption of weak coupling holds. With some simplification it has been applied to the present quantum dot problem with two levels by Král and Khás.⁵ There are two diagrams of first order in the phonon propagator that can be classified as the Hartree and exchange self-energies, $\Sigma = \Sigma^H + \Sigma^X$. Usually, the Hartree term is neglected in view of a constant electron charge density, which is compensated by a positive background. However, in the case of quantum dots being localized in space, the situation is different: In a phonon-assisted transition between levels, the charge structure of the electron (given by the confinement functions) changes, and a classical electrostatic contribution to the lattice deformation appears. Explicitly, the Hartree self-energy of level i is given by ($\hbar = 1$)

$$\Sigma_i^H = -\frac{2}{\omega_0} \sum_{j, \mathbf{q}} n_j M_{\mathbf{q}}^{i,i} M_{-\mathbf{q}}^{j,j}. \quad (22)$$

The electronic occupations numbers n_i have to be determined via the spectral function,

$$n_i = \int_{-\infty}^{\infty} d\omega A_i^B(\omega) f(\omega). \quad (23)$$

Under equilibrium conditions, the electrons are distributed over energy according to the Fermi function $f(\omega) = (e^{\beta(\omega - \mu)} + 1)^{-1}$ with inverse temperature $\beta = 1/k_B T$ and chemical potential μ . The spectral function and self-energy are related as usual via

$$A_i^B(\omega) = -\frac{1}{\pi} \text{Im} \frac{1}{\omega - \epsilon_i - \Sigma_i(\omega)}. \quad (24)$$

The exchange self-energy reads

$$\begin{aligned} \Sigma_i^X(\omega) &= \sum_{j, \mathbf{q}} M_{\mathbf{q}}^{i,j} M_{-\mathbf{q}}^{j,i} \int_{-\infty}^{\infty} d\omega' A_j^B(\omega') \left[\frac{N(\omega_0) + f(\omega')}{\omega + \omega_0 - \omega' + i0} \right. \\ &\quad \left. + \frac{N(\omega_0) + 1 - f(\omega')}{\omega - \omega_0 - \omega' + i0} \right]. \end{aligned} \quad (25)$$

Here, $N(\omega) = (e^{\beta\omega} - 1)^{-1}$ is the Bose function and gives the phonon occupation. For the self-consistent first-order Born

approximation, Eqs. (22)–(25) have to be solved in an iterative manner until convergence for the total self-energy $\Sigma_i(\omega)$ is reached.

This approximate result will be compared with the spectral function based on the numerically exact eigenfunctions and eigenvalues. As a starting point we use the general definition

$$A_{i,j}(\omega) = \frac{1}{\pi} \text{Re} \int_0^\infty dt e^{i\omega t} \text{Tr} \{ \rho [c_i(t), c_j^\dagger(0)]_+ \} \quad (26)$$

with ρ as the density matrix. Expanding into exact eigenstates we obtain

$$A_i(\omega) = \sum_{N,\nu,\mu} (\rho_\nu^N + \rho_\mu^{N-1}) | \langle N-1; \mu | c_i | N; \nu \rangle |^2 \times \delta(\epsilon_\nu^N - \epsilon_\mu^{N-1} - \omega), \quad (27)$$

where $H|N; \nu\rangle = \epsilon_\nu^N |N; \nu\rangle$ denotes the eigenvalue problem in the N -electron subspace. In equilibrium, ρ is diagonal, $\langle N; \nu | \rho | N; \mu \rangle = \delta_{\nu,\mu} \rho_\nu^N$. Due to the well-defined parity of the Hamiltonian equation (10), the spectral function has only elements diagonal in the sublevel index. Parity means here that the electron- B_1 subspace decomposes into two subsets defined by the states $\{c_0^\dagger | V \rangle | 2n_1 \rangle_0, c_1^\dagger | V \rangle | 2n_1 + 1 \rangle_0\}$ and $\{c_0^\dagger | V \rangle | 2n_1 + 1 \rangle_0, c_1^\dagger | V \rangle | 2n_1 \rangle_0\}$, respectively.

For the spectral function in the rotating-wave approximation, an explicit result can be given using the results of Eqs. (20) and (21). Again the mode B_3 is neglected, which can be easily incorporated afterwards. To complete the spectrum of H^{RW} we have to include the noninteracting states $|0, n_2\rangle^0 \equiv c_0^\dagger | V \rangle | 0 \rangle_0 | n_2 \rangle_{-\eta}$ with eigenvalues $\epsilon_0 + n_2 \omega_0$, which resemble the ground state for fixed n_2 .

The spectral function in RW has two contributions, $A_0^{RW} = A_0^{RW,10} + A_0^{RW,21}$, which refer to the transition between one (zero) particles and two (one) particles, respectively.

$$\begin{aligned} A_0^{RW,10} &= \sum_{n_2, n_2'=0} (\rho_{0, n_2}^1 + \rho_{0, n_2'}^0) | \langle 0 | n_2' | n_2 \rangle_\eta |^2 \\ &\times \delta(\epsilon_0 + (n_2 - n_2') \omega_0 - \omega) + \sum_{n_1=1; n_2, n_2'=0} (\rho_{n_1, n_2}^1, - \\ &+ \rho_{n_1, n_2'}^0) | c_{n_1, n_2}^- |^2 | \langle 0 | n_2' | n_2 \rangle_\eta |^2 \\ &\times \delta(\epsilon_0 - \Delta/2 - R_{n_1, n_2}/2 + (n_2 - n_2') \omega_0 - \omega) \\ &+ \sum_{n_1=1; n_2, n_2'=0} (\rho_{n_1, n_2}^1, + + \rho_{n_1, n_2'}^0) \\ &\times | c_{n_1, n_2}^+ |^2 | \langle 0 | n_2' | n_2 \rangle_\eta |^2 \\ &\times \delta(\epsilon_0 - \Delta/2 + R_{n_1, n_2}/2 + (n_2 - n_2') \omega_0 - \omega), \quad (28) \end{aligned}$$

where $\rho_{0, n_2}^1 \equiv \langle 0 | n_2 | \rho | 0, n_2 \rangle^0$, $\rho_{n_1, n_2, \pm}^1 \equiv \langle n_1, n_2 | \rho | n_1, n_2 \rangle^\pm$, and $\rho_{n_1, n_2}^0 \equiv \langle 0; n_1, n_2 | \rho | 0; n_1, n_2 \rangle$ [see Eq. (13)]. An analogous expression can be given for $A_0^{RW,21}(\omega)$. The spectral function $A_0^{RW}(\omega)$ satisfies the strict sum rules for the zeroth and first moment, which give normalization and average energy, respectively.

Since the overlap between displaced oscillators does not vanish even for different quantum numbers, the double summation over n_2 and n_2' will account for the satellites of the spectral function that appear at multiples of the phonon frequency ω_0 . The approximation is good as long as the system is close to zero detuning, i.e., as long as degenerate perturbation theory works. Treating additionally the eigenstates in perturbation theory leads to marginal improvements only.

For the construction of the one-particle spectral function, a combination of states referring to different fermion subspaces are needed, since the spectral function gives the frequency-resolved probability for removal (or addition) of one electron. A signature of this general behavior is the occurrence of both the Rabi splitting and the bare detuning in the δ -function argument of Eq. (28), accompanied with the overlap between non-interacting and interacting states. Since, on the other hand, the phonon-assisted transition of an electron occurs exclusively in the subspace of one fermion, we have severe doubts on using a convolution of one-particle spectral functions for describing relaxation, as done in Ref. 5. We expect this deficiency to be dramatic in particular when only a few electronic levels are involved, as in the present case.

B. Numerical results

In the following figures we present our numerical results. The exact spectral function of the lower level $A_0(\omega)$ (solid line), obtained from numerical diagonalization, is compared with the simplified solution in rotating-wave approximation A_0^{RW} (dotted line) and with the self-consistent first-order Born approximation A_0^B (dashed line). Both A_0 and A_0^{RW} are convoluted with a Gaussian of variance $\sigma = \hbar \omega_0 / 70$.

However, we want to emphasize that the exact spectral function [and $A_i^{RW}(\omega)$ as well] consists of δ functions only. This was clear from the beginning since a finite perturbation cannot change the character of the spectrum of the unperturbed system. Since we started from *dispersionless* bosonic modes, the discrete electronic spectrum cannot be altered by the finite electron-phonon interaction.¹⁹ For the density matrix in Eq. (26) we assume the grand canonical equilibrium distribution $\rho \propto \exp[-\beta(H - \mu N)]$ and fix the chemical potential μ halfway in energy between the bare electronic levels. This is close to putting just one electron into the dot.

Figure 2 shows the spectral functions at $k_B T = \hbar \omega_0$ ($T = 426$ K) for the *bare* detuning zero, i.e., $\epsilon_1 = \hbar \omega_0$ (ϵ_0 is taken as the zero of energy in what follows). This level spacing corresponds to a dot size of $x_0 = 3.9$ nm.

The prominent feature is the splitting of the spectral function into some kind of doublet that can be traced back to the level repulsion in the (dominant) B_1 channel. Its value is related to the strength of C_1 (3.3 meV). Looking more closely it becomes apparent, however, that the upper and lower structure do not have an equal weight as one would expect for zero detuning. This clearly indicates the shift in level spacing (renormalized detuning). Furthermore, an overall shift with respect to the bare energy $\epsilon_0 = 0$ is obvious, which stems from the polaronic shift of the other modes. All these features can be seen also at $k_B T = \hbar \omega_0 / 4$ ($T = 106$ K), which is more relevant for realistic dot spectroscopy (Fig. 3). The right-hand panels of Figs. 2 and 3 show the first satellite

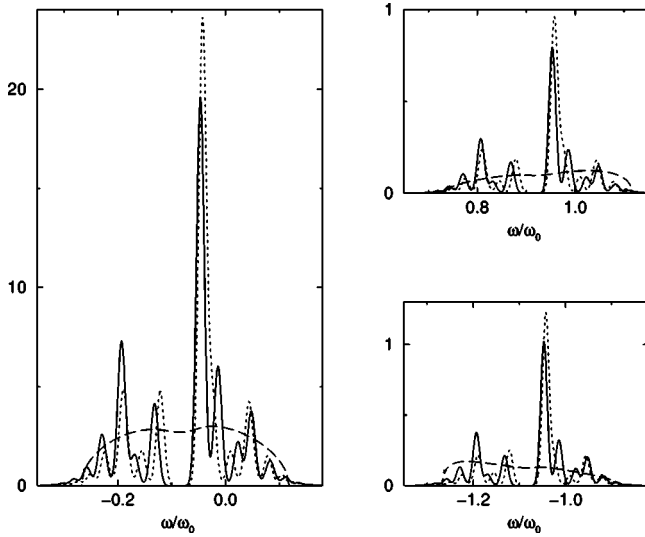


FIG. 2. The spectral functions A_0 (full), A_0^{RW} (dotted), and A_0^{B} (dashed) at $k_B T = \hbar \omega_0$ and zero detuning. The energy scale is in units of $\hbar \omega_0$, with $\epsilon_0 = 0$. The right panels show the first satellite structure at one phonon energy above (top) and below the main level (bottom).

structures. They are reduced in weight but have a similar appearance as in the main structure.

In Fig. 4 the spectral functions at $k_B T = \hbar \omega_0$ are compared for two nonzero detunings $\Delta = \hbar \omega_0 - \epsilon_1 + \epsilon_0$, which refer to dot sizes of $x_0 = 3.6$ nm (left) and $x_0 = 4.4$ nm (right), respectively. The spectral function $A_0^{\text{RW}}(\omega)$ shows good agreement with the exact solution in all features. In particular, the complex structure of lines having different weight and position is well reproduced. This means that not only the eigenvalues are correctly approximated by the reduction of the system into (many) 2×2 matrices but also the eigenvectors.

Turning to the self-consistent first-order Born approximation, $A_0^{\text{B}}(\omega)$, the agreement is not satisfying at all. At the higher temperature (Fig. 2), instead of the complex structure only two broad bands are seen that have nearly smeared out

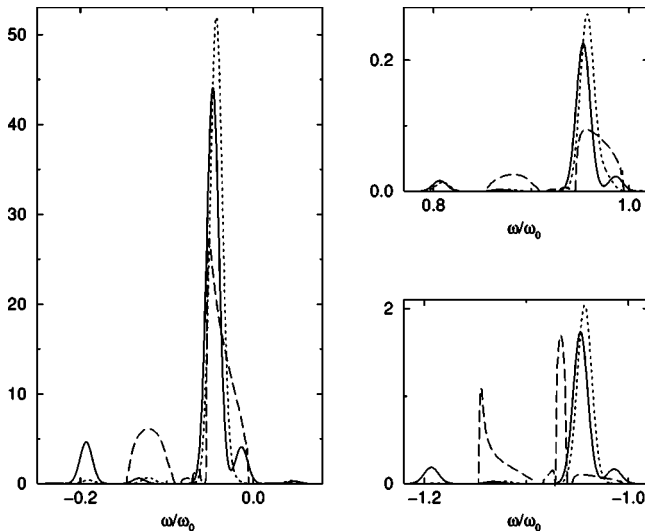


FIG. 3. The spectral functions at $k_B T = \hbar \omega_0 / 4$ and zero detuning. Other data as in Fig. 2.

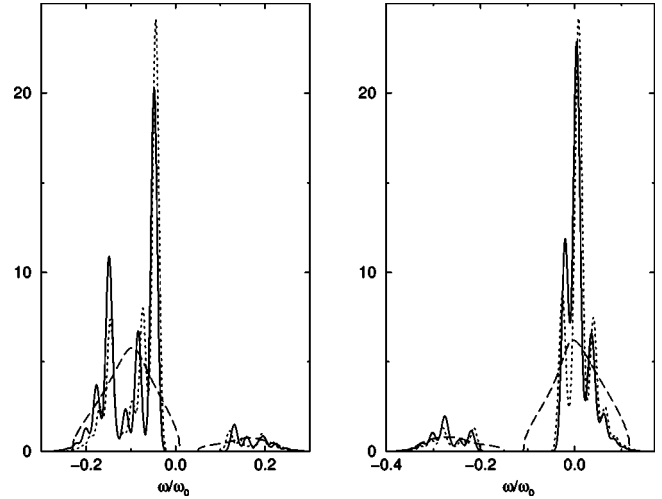


FIG. 4. The spectral functions at $k_B T = \hbar \omega_0$ for negative detuning ($\Delta = -0.2 \hbar \omega_0$, left panel) and positive detuning ($\Delta = 0.2 \hbar \omega_0$, right panel). Other data as in Fig. 2.

the level repulsion. For the lower temperature (Fig. 3), the Born approximation works somewhat better since here the unperturbed but shifted ground-state energy carries the dominant weight.

IV. CONCLUSIONS

We have presented a model that is suitable to describe the electron-phonon interaction in quantum dots and can be treated numerically *exact*. Thus we were able to compare the exact spectral function with the self-consistent first-order Born approximation. The first observation was that the exact spectral function consists of δ functions, whereas the Born approximation gives a continuous spectral function. But even after an artificial broadening of the exact spectral function, striking differences remain, which are due to the inability of the Born approach to exhibit all shifts and splittings in detail. We would like to mention that without including the Hartree self-energy into the Born calculation, the agreement would be even worse.

As an alternative we propose an analytic solution that employs the coherent states for representing the electron-phonon Hamiltonian. The rotating-wave approximation allows us to reduce the problem to an (infinite) number of decoupled 2×2 matrices. Close to resonance (zero detuning), this approximation works remarkably well. The appearance of Franck-Condon factors points to the great similarity of the electron-phonon interaction in molecules and quantum dots.

Starting from the full electron-LO-phonon Hamiltonian with Fröhlich interaction, we were able to reduce the problem in a quantum dot to a few boson modes only. Due to this considerable reduction in numerical labor, it was possible to construct the exact eigenstates and energies. In the present work, these results have been used to calculate the one-particle spectral function. However, an extension to two-particle expectation values that are relevant for, e.g., transition rates is possible. Using the density-matrix scheme, relaxation processes can be studied within the same reduced Hilbert space, a route that will be followed in future work.

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APPENDIX: DETERMINATION OF COUPLING CONSTANTS

With the transformations of Eq. (2) we obtain the following expressions for the six coupling constants:

$$\begin{aligned}
|C_1|^2 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,0} M_{-\mathbf{q}}^{0,1}, \\
C_2 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{0,0} M_{-\mathbf{q}}^{0,1} / C_1, \\
|C_3|^2 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{0,0} M_{-\mathbf{q}}^{0,0} - |C_2|^2, \\
C_4 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,1} M_{-\mathbf{q}}^{0,1} / C_1 + C_5 C_2 / C_3, \\
C_5 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,1} M_{-\mathbf{q}}^{0,0} / C_3, \\
|C_6|^2 &= \sum_{\mathbf{q}} M_{\mathbf{q}}^{1,1} M_{-\mathbf{q}}^{1,1} - |C_4|^2 - |C_5|^2 \\
&\quad + 2 \operatorname{Re}[(2C_2^* C_3 C_4 C_5^* - |C_2|^2 |C_5|^2) / C_3^2].
\end{aligned} \tag{A1}$$

The standard Fröhlich coupling for the electron–LO-phonon interaction is adopted and applied to the dot confinement states,

$$\begin{aligned}
M_{\mathbf{q}}^{i,j} &= \frac{A}{V^{1/2} q} \Phi_{\mathbf{q}}^{i,j}, \quad A^2 = \hbar \omega_0 \frac{e^2}{2\epsilon_0} \left(\frac{1}{\kappa_{\infty}} - \frac{1}{\kappa_0} \right), \\
\Phi_{\mathbf{q}}^{i,j} &= \int d^3 r \psi_i^*(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} \psi_j(\mathbf{r}).
\end{aligned} \tag{A2}$$

For simplicity we consider an anisotropic parabolic potential as dot confinement, with x as the long axis. The two energetically lowest wave functions read

$$\begin{aligned}
\psi_0(\mathbf{r}) &= (\sqrt{2\pi} x_0 y_0 z_0)^{-1/2} \exp \left[-\frac{1}{4} \left(\frac{x^2}{x_0^2} + \frac{y^2}{y_0^2} + \frac{z^2}{z_0^2} \right) \right], \\
\psi_1(\mathbf{r}) &= \frac{x}{x_0} \psi_0(\mathbf{r}),
\end{aligned} \tag{A3}$$

where $x_0 > y_0, z_0$ are the spatial extensions (variances) of the ground state. The relevant level distance is

$$\epsilon_1 - \epsilon_0 = \frac{\hbar^2 x_0^{-2}}{2m_e}. \tag{A4}$$

The matrix elements in Eq. (A2) read

$$\begin{aligned}
\Phi_{\mathbf{q}}^{0,0} &= \exp \left(-\frac{1}{2} (q_x^2 x_0^2 + q_y^2 y_0^2 + q_z^2 z_0^2) \right), \\
\Phi_{\mathbf{q}}^{1,0} &= i q_x x_0 \Phi_{\mathbf{q}}^{0,0}, \\
\Phi_{\mathbf{q}}^{1,1} &= (1 - q_x^2 x_0^2) \Phi_{\mathbf{q}}^{0,0}.
\end{aligned} \tag{A5}$$

The final integration over \mathbf{q} leads to $C_2 = 0$, $C_4 = 0$ on account of the well-defined parity of the wave functions. The other constants can be reduced to the following (elliptic) integrals,

$$I_n = \frac{A^2}{8\pi^{3/2} x_0} \int_1^{\infty} dt t^{-n} [t(t - \alpha_y)(t - \alpha_z)]^{-1/2}, \tag{A6}$$

with $\alpha_y = 1 - (y_0/x_0)^2 < 1$ and $\alpha_z = 1 - (z_0/x_0)^2 < 1$. We obtain finally

$$\begin{aligned}
C_1 &= (I_1/2)^{1/2}, \quad C_3 = (I_0)^{1/2}, \\
C_5 &= (I_0 - I_1/2) / C_3, \\
C_6 &= (I_0 - I_1 + 3I_2/4 - C_5^2)^{1/2}.
\end{aligned} \tag{A7}$$

For the numerical calculations we choose material constants of GaAs, i.e., LO-phonon energy $\hbar \omega_0 = 36.7$ meV, conduction band mass $m_e = 0.067 m_0$, and dielectric constants $\kappa_{\infty} = 10.7$, $\kappa_0 = 12.4$. According to Eq. (A4), a (long) dot extension of $x_0 = 3.9$ nm gives resonance between level spacing and LO energy. The effective dot extensions in y and z directions are taken equal, $y_0 = z_0 = 3$ nm, which renders the elliptic integrals (A6) to be simple logarithmic functions.

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