LO-phonon overheating in quantum dots

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Longitudinal optical phonons have been used to interpret the electronic energy relaxation in quantum dots and at the same time they served as a reservoir, with which the electronic subsystem is in contact. Such a phonon subsystem is expected to be passive, namely, in a long-time limit the whole system should be able to achieve such a stationary state, in which statistical distributions of both subsystems do not change in time. We pay attention to this property of the LO phonon bath. We show the passivity property of the so far used approximations to electronic transport in quantum dots. Also we show a way how to improve the passivity of LO phonon bath using canonical Lang-Firsov transformation.

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1 Introduction

Semiconductor nanostructures, like quantum dots, appear to bring new properties due to their restricted size in all three dimensions. The size of them is often not much larger that the charge carrier’s mean free path. Electronic energy relaxation in quantum dots does not seem a fully resolved question, both in experiment and theory [1]. The rapidity of electronic relaxation may be ascribed to the coupling of electrons with longitudinal optical (LO) phonons [2, 3], although the relaxation effect in quantum dots was also interpreted with help of Auger mechanism. Besides this, theory of polarons in quantum dots has developed recently, which concept, together with the finite lifetime of LO phonons, has became another way of explanation of the rapid relaxation of electronic energy in quantum dots [4].

The quantum transport theory, building in the concept of nonequilibrium Green’s functions [5], or alternatively on using the theory of nonequilibrium statistical operator [6], used the self-consistent Born approximation to the electronic self-energy. This approximation to the quantum transport of charge carriers in quantum dots was able to provide explanation of the rapidity of electronic relaxation [2, 3], and of some properties of the optical transitions, like the line-shape [7], but it also recently provided arguments in favor of electronic level occupation up-conversion and incomplete depopulation in quantum dots, which effect appears in remarkable agreement with a number of experimental papers (see e.g. [8, 9]). The self-consistent Born (SCB) approximation to electronic self-energy, which we are going to use here, means that the electron is bound to a cloud of virtual LO phonons. It is already at this level that we consider explicitly the motion of the reservoir and

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its entanglement with the electronic subsystem. This optical phonon cloud can be viewed as coherent phonons coupled to an electron. In analogy with coherent light, this coherent phonons may act as an effective classical time-dependent field, in which the electron moves. Due to this feature we can meet with the off-shell property of the collision integral of the electronic quantum transport equation in the SCB approximation.

Because this self-consistent Born approximation to the electron quantum transport equation leads to an off-shell formulation of the transport equation, a question naturally arises concerning an overall energetic balance in the processes of electronic energy relaxation via electron-LO-phonon scattering in quantum dots. Within this subject we can raise questions about the passivity of the phonon reservoir. In particular, we may ask, whether the whole system of electrons and phonons, can evolve to a long-time limit, such that in this limit the density matrices of both subsystems are in a steady state. If this happens to be the case, we can speak about a passivity of the phonon reservoir. Naturally, this passivity property is a desired characteristics of the theory describing the system under consideration, while the opposite case means a difficulty.

The questions of passivity is met in the Gaussian white noise problem [10] and in the theory of interacting harmonic oscillators [11, 12]. In this work we demonstrate, using quantum transport equations, that the problem of a non-passive reservoir occurs in quantum dots. We shall identify a significant source of such non-passivity with certain terms in the electron-phonon interaction operator of the system. These terms are those, which play a leading role in the creation of the polaron well of the charge carrier in a dot. The existence of the overheating in quantum dots shows that the formulation of the phonon kinetic equation need to be approached with care. We shall show a possibility of a partial elimination of this effect. This will be done by changing the basis set of electron and phonon eigenstates, switching from the basis from noninteracting electron and phonons to a set of dressed states of electron and phonons, which assumes that the crystal lattice accommodates to the presence of electrons in quantum dot. The transition to the new dressed basis is achieved by performing the well-known canonical Lang-Firsov transformation [13] of the Hamiltonian of the system, which exactly diagonalizes a certain part of the full electron-phonon Hamiltonian, including those terms of the electron-phonon coupling, which lead to a formation of the electronic polaron well. The result of the transformation then is a Hamiltonian for new quasiparticles interacting by a new interaction operator. The advantage of this new picture then is that the new quasiparticles do not tend to develop quickly a polaron potential well around them. The phonon kinetic equation may then become free of corresponding spurious effects like an excess heating of phonons.

Considering electronic relaxation and phonon generation in the system of a single quantum dot with a single spin-less electron and with the influence of holes in the valence band states of quantum dot completely neglected [2, 8, 9], we shall demonstrate the existence of the phonon overheating effect in quantum dots and show how to eliminate it to a considerable degree. We also show that the electronic relaxation characteristics, given by the electronic transport equation, remain nearly
unchanged by the treatment of the overheating effect with help of the Lang-Firsov transformation.

2 Hamiltonian

The electronic eigenstates of this single electron in a quantum dot will be approximated by electronic eigenstates in a cubic shaped quantum dot, with infinitely deep potential. The present model assumes only two nondegenerate electron states $\psi_n(r)$ in this dot, namely the ground state ($n=0$) and one of the lowest-energy triple-degenerate exited states ($n=1$). The bath of LO phonons will be approximated by the bulk modes of LO phonons of the full sample. The Hamiltonian of the whole system is:

$$H = H_e + H_{ph} + H_1,$$

(1)

in which $H_e = \sum_{i=0,1} E_i c_i^+ c_i$, with $c_i$ being annihilation operator of electron in state $i$. We set the energy of the ground state $E_0 = 0$. $H_{ph} = \sum_q E_{LO} b_q^+ b_q$, with $b_q$ being LO phonon annihilation operator in state with phonon momentum $q$. The interaction of electron with modes $q$ of phonons, with phonon energy $E_{LO}$, is

$$H_1 = \sum_{q,m,n=0,1} A_q \Phi(n,m,q) (b_q - b_{-q}^+) c_n^+ c_m.$$

(2)

The Fröhlich’s coupling $H_1$ between the electron and LO phonons contains the coupling constant $A_q$, which is $A_q = (-ie/q)|E_{LO}(\kappa^{-1}_\infty - \kappa^{-1}_0)|^{1/2}(2\varepsilon_0 V)^{-1/2}$, where $\kappa_\infty$ and $\kappa_0$ are, respectively, high-frequency and static dielectric constants, $\varepsilon_0$ is permittivity of free space, $-e$ is the electronic charge, $q = |q|$, and $V$ is volume of the sample. $\Phi$ is the form-factor, $\Phi(n,m,q) = \int d^3r \psi_n^*(r) e^{iqr} \psi_m(r)$.

This Hamiltonian consists of the electronic subsystem, $H_e$, the bath $H_{ph}$ and their coupling $H_1$. A remarkable feature of this Hamiltonian is the presence of two different components of interaction operator. This operator can be written as $H_1 = H_1^{(t)} + H_1^{(l)}$. The transverse part $H_1^{(t)}$ contains that part of $H_1$, in which the electronic orbital indexes $n,m$ are equal to each other, which meaning is that electron emits or absorbs phonon without changing orbital state. The longitudinal term $H_1^{(l)}$ contains terms, in which the electronic orbital indexes are different, expressing transition between different orbital states, during emission or absorption of a phonon. The former term $H_1^{(t)}$ plays a role in processes of development of a polaron well, after the electron is placed into an orbital. In the formulation of phonon quantum transport equation for the distribution function $< b_q^+ b_q'>$ we do not take into account the process of the development of such a polaron well in the form of a deformation of the potential in which the harmonic oscillators of the phonons move in the presence of the charge carrier. The deformation of the phonon field could be described by a quantity like the average $<b_q>$, for which we are not currently considering an appropriate transport equation. Therefore, it would not be unexpected if the term $H_1^{(t)}$ led to a conflicting effect in the standard phonon transport equation for the density matrix $<b_q^+ b_q'>$. The transverse term of the
electron-phonon coupling may contribute to this effect already in the first order of perturbation, while the longitudinal term \( H_1^{(1)} \) can contribute to such an effect only at higher orders.

We need to describe the electron-phonon system with having the phonon bath passive. This means that energy does not permanently come into or leave the bath in the long-time limit. The bath energy is related to the total phonon number \( n \). The longitudinal term is the electron-phonon coupling may contribute to this effect already in the first order of perturbation, while the longitudinal term \( H_1^{(1)} \) can contribute to such an effect only at higher orders.

We shall show the result of the Lang-Firsov transformation of the Hamiltonian \( H \) and we shall also present the transformation of the remaining longitudinal part of the coupling \( H_1^{(1)} \). We shall calculate the magnitude of the permanent phonon generation in the phonon bath within the quantum transport equation based on the full interaction operator \( H_1 \). Then we shall show how much the Lang-Firsov transformation helps to suppress the phonon overheating effect.

With help of canonical Lang-Firsov (LF) transformation \( S \) we introduce new particle operators of electron, \( A_n \), and of phonons, \( B_q \), by relations \( \epsilon_n = E_{LO}A_nS^+ \) and \( b_q = S B_qS^+ \). The operator \( \sigma = -\sum_{j,n}(A_q\Phi(j,j,q)/E_{LO})(B_q + B_{-q})A_j^+A_j \). This transformation diagonalizes exactly the independent boson part of the full Hamiltonian, \( H_{IB} \). Therefore, the full Hamiltonian has now the form

\[
H = \sum_n (E_n - \frac{\alpha_{nn}}{E_{LO}})A_n^+A_n + \sum_q E_{LO}B_q^+B_q - \frac{\kappa_{01}}{E_{LO}}A_1^+A_1A_0^+A_0 + \tilde{H}_1^{(1)},
\]

where \( \kappa_{01} = 2 \sum_q |A_q|^2 \ |\Phi(0,0,q)|^2 \), while the electronic polaron well depth is \( \alpha_{nn}/E_{LO} \), with \( \alpha_{nn} = \sum_q |A_q|^2 \ |\Phi(n,n,q)|^2 \). \( \tilde{H}_1^{(1)} \) is the longitudinal part of the electron-phonon interaction transformed by the canonical transformation \( S \). This operator comes out from the transformation as \( \tilde{H}_1^{(1)} = V_1 + V_2 \).

Before specifying these two operators, let us introduce an abbreviation, \( \gamma_{j,q} = A_q\Phi(j,j,q)/E_{LO} \), which is a quantity of the first order in the electron-phonon coupling \( A_q \). The operator \( V_2 \) reads:

\[
V_2 = 2 \sum_{j,q} A_q\Phi(0,1,q)\gamma_{j,-q}A_0^+A_1e^{\beta} + h.c.,
\]

where \( \beta = \sum_p (\gamma_{1,p} - \gamma_{0,p})(B_p + B_{-p}) \). Let us note that this part of the electron-phonon interaction is at least of the second order in the coupling constant \( A_q \). In the lowest order in the electron-phonon coupling constant it gives a phonon-less electronic level mixing. We shall neglect this effect. Because in \( V_2 \) the electron-phonon coupling starts in the third order of the expansion in powers of the coupling,
we shall neglect $V_2$ in comparison with $V_1$, which reads:

$$V_1 = \sum_q A_q \Phi(0, 1, q)(B_q - B_{-q}^+)A_0^+ A_1 e^{\beta} + h.c. \quad (5)$$

The effect of the Lang-Firsov transformation has thus been concentrated in the latter formula into the exponential factor, which now contains a complicated dependence on the phonon operators. The advantage of making the LF transformation is also in that we can further simplify the electron-phonon operator $V_1$. The expansion of $V_1$ in powers of the coupling starts with the first power of $A_q$. The lowest order term corresponds to putting $\beta$ equal zero. Confining ourselves to keeping only the leading order terms in $V_1$, we estimate the influence of the exponential factor. Namely, we approximate $e^{\beta}$ by an average of this quantity $< e^{\beta} >$, calculated with canonical ensemble at a temperature $T$. Calculating this average [13] we get $< e^{\beta} > = \exp[-\sum_p \gamma_{1,p} - \gamma_{0,p}^2 (B_p^+ B_p + 1/2)]$. Here $B_p^+ B_p$ has the meaning of an average number of phonons with wave vector $p$. We shall first of all assume that this number of phonons is small, comparable with the factor $1/2$. For the second, we see that the order of the square of difference of the coupling constants $\gamma_{n,p}$ is given approximately by the polaron constant [13], which is about 0.07 in GaAs. From these reasons we approximate the factors $e^{\beta}$ by the value of 1.

After having made these approximations we can see that the longitudinal part of the coupling has not changed upon making the LF transformation.

![Fig. 1. Long-time limit (steady state) of occupation $N_0$ of electronic lower-energy state at given temperature $T_{LO}$ of LO-phonons. Dashed line is calculated without LF transformation.](image)

We also neglect the polaron well depth component of new electron energy, $\alpha_{nn}/E_{LO}$, in [3] and the term proportional to $\kappa_0$ in the same equation. These terms are small on the scale of the electron energy level separations, and the optical phonon energy, in cases we are interested in. With the above made approximations,
the resulting Hamiltonian of the system, after making LF transformation, is the operator identical with the original Hamiltonian $H$ of equation (1), with the only change that the transverse coupling $H_{1}^{(t)}$ has been dropped. In other words, when estimating numerically the electronic relaxation in the dressed electron and phonon basis, the original Hamiltonian (1) can be used, with the transverse terms put equal zero. The removal of the transverse terms from the original Hamiltonian is done simply by putting the form-factors $\Phi(n, n, q)$ equal zero for $n = 0, 1$.

3 Electron relaxation and phonon heat generation

In order to demonstrate the overheating effect, we calculate first the heat generation in the phonon system with using the full original Hamiltonian (1). The kinetic equations for the electron was derived earlier [2, 8] with help of either nonequilibrium statistical operator or nonequilibrium Green’s functions [6, 5]. The derivation of the kinetic equation for the phonon system can be obtained in the same way. The state of the phonon system should be generally described with help of the density matrix $<b_{q}^{+}b_{q'}>$, or equivalently with help of the Wigner function. This generality would be good for expressing correctly the generation of phonons in the area of the quantum dot only. In order to reduce the computing demands as much as possible, while conserving reasonably much the predictivity of the model, we reduce somewhat the complexity of the system of phonon modes. In that sense, in order to get rid of the off-diagonal elements of the phonon density matrix, we identify the volume of the basic area of the sample with the volume of the quantum dot. In addition to this, we formally reduce the number of the modes, with which the electron interacts, to just a single one, with phonon annihilation operator $b$, the interaction constant of this mode with the electron being chosen such that the coupling of electron to this single mode provides the same relaxation rapidity as the original full set of LO phonon modes. We shall publish the argumentation leading to this simplification elsewhere. The kinetic equations for the electron distribution function in the excited state, $<c_{1}^{+}c_{1}>$, is derived in the self-consistent Born Approximation to electronic self-energy and in the random-phase approximation to the phonon self-energy. In the right hand sides of the kinetic equations, and in the equation for the self-energy, the phonon propagator is approximated by undressed phonon propagator. Instantaneous collisions approximation is used. Standard Kadanoff-Baym ansatz is used, which step is regarded as suitable enough for expressing the long-time limits of development of the system’s statistics. Other details of the derivation of the kinetic equations will also be described elsewhere.

We shall first show the electronic relaxation at a given temperature of the LO-phonon reservoir. We use the material constants of GaAs, with energy of LO phonon of 36.2 meV. The lateral size of the cubic dot is taken to be 16.4 nm. This size corresponds to the electronic energy level separation of 62.7 meV. The electronic transport equation calculated with the help of the full Hamiltonian in the original form (1) was solved first to find the electronic distribution among the two levels at a given lattice temperature. In Fig. 1 the dashed line shows the lattice temperature...
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Fig. 2. Plot of generation rate $d < b^+ b > / dt$ of LO-phonons as a function of temperature $T_{LO}$ of the lattice. The steady-state occupation $N_0$ of the lower-energy electronic state is shown in Fig. 1.

dependence of the occupation $N_0$ of the lower energy electron orbital, at which the relaxation rate $dN_0/dt$ is zero. In other words, at this $N_0$ the electronic subsystem reaches the long-time limiting steady state distribution. Let us remark that we have earlier shown [15] that these values of $N_0$ are well below the values given by the Fermi-Dirac distribution of electron in the present electronic two-level system. The dashed curve at Fig. 2 shows the generation rate of LO phonons $d < b^+ b > / dt$ at given phonon temperature and at the steady state electronic distribution given by the dashed line in Fig. 1. One can see that at these conditions the phonon system does not have a steady state. It permanently heats up with the rather high speed of about 1.5 to 1.8 phonons per picosecond (the dashed line). We ascribe this fast heating to the role of the transverse coupling terms in the original Hamiltonian [11], as discussed in an earlier section. From the above given reasons we go over to a different representation, using dressed electron and phonon basis, writing quantum transport equations for electron and the phonon transformed with help of Lang-Firsov transformation. In this new representation we use an approximate Hamiltonian, which equals the original operator [11] with the transverse terms dropped. When the same characteristics of electron and the phonon are calculated, we get the full lines in both Figs. 1 and 2. We see that in the new basis the phonon heating is considerably reduced, namely by the factor of about 7, with the present simple tool based on LF transformation.

Summing up, let us note the remarkable numerical result for the long-time limit of electronic distribution at given lattice temperature: this long-time limit practically does not depend on whether the calculation is done with or without the Lang-Firsov transformation procedure applied. This helps us to make a conclusion: the use of the original representation in undressed states, using the full Hamiltonian
having the property of the phonon overheating effect, appears to give us plausible data about the electronic relaxation in the quantum dot. From this reason, the earlier made conclusions about the electron relaxation effects [8, 9], may remain valid. As for the phonon kinetic equation, the transformation to the dressed basis with LF transformation helps to reduce the overheating effect considerably.

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