Incomplete electronic relaxation and population upconversion in quantum dots

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We calculate the electronic relaxation of a single electron in a quantum dot with two electronic orbital states, and with the electronic coupling to the longitudinal optical modes of the lattice vibrations included in the multiple scattering approximation to the electronic self-energy. This model shows that there is an incomplete electronic relaxation from the excited state to the ground state and also the incomplete increase of the population of the excited state after resonantly exciting the ground state. These theoretical results are compared with very similar recent findings reported in experimental papers.

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I. INTRODUCTION

Relaxation of the electronic energy in individual quantum dots and the question of the phonon bottleneck still remain to be of considerable current interest. The Fröhlich's mechanism of the electron-LO-phonon coupling in the polar semiconductors is regarded as very important. The electronic self-energy due to this electronic interaction was studied by Inoshita and Sakaki, who found that the spectral features of the electronic spectral density have very narrow width. The dependence of these very narrow spectral peaks on the energy parameter $E$ was shown to have the shape of the $E^{-1/2}$, in the limit of zero temperature.

The relaxation of the electron energy in quantum dots was studied later in the approximation of multiple electronic scattering on optical phonons expressed in the self-consistent Born approximation. It was theoretically shown that despite the fact that the spectral peaks of the electronic spectral density have zero width in the sense of full width at half maximum, the relaxation rate of an electron from the excited state in the quantum dot remains nonzero in the limit of zero temperature of the lattice. The relaxation rate obtained in this way was shown to have the order of magnitude of $1/P_{LO}^{8,9}$, in agreement with broad experimental data.

In the same approximation to the electronic self-energy it was demonstrated that the model gives the electronic relaxation even in cases when the electronic inter-level energy separation is not equal to the optical phonon energy. The dependence of the relaxation rate on the detuning, between the electronic energy level separation and the energy of the optical phonon, has a resonance structure. The existence of this resonance structure is in accord with the well-known experimental rule for detecting the presence of quantum dot in quantum dot samples with help of optical methods, basing on the detection of the resonance structure of the photoluminescence excitation signal with peaks separated by optical phonon energy $E_{LO}$.

The important role of Fröhlich's coupling has also been supported by the recent interpretation of the optical line broadening and of the continuous background observed in photoluminescence excitation spectra measured on self-assembled quantum dot samples at high levels of the electronic excitation.

Treating the electronic relaxation in the self-consistent Born approximation to the self-energy means that the electron makes the irreversible transition between two electronic states which are renormalized with respect to the electron-phonon coupling, with the electronic self-energy taken in the self-consistent Born approximation. This approximation takes into account multiple scattering processes of electron on the system of optical phonons and introduces in this way the multiphonon states into the relaxation rate formula and into the formula for the self-energy. The coherent multiphonon states created in this way are analogous to the classical macroscopic oscillations of the lattice and the irreversible process of the electronic relaxation in this system can be viewed as a quantum transition in a potential dependent on time. A nonconservation of energy in such a system then may be manifested as an absence of the phonon bottleneck in quantum dots. Because of the rapidity of the electronic relaxation in quantum dots, this property of the electronic relaxation between the electronic states in the dot restricts considerably the possible use of quantum dots in constructing quantum bits using the electronic orbital states in quantum dots as qubit states.

To our knowledge, in the theory the electronic relaxation has so far been studied with an emphasis to the relaxation rate from the excited state only, calculated at the initial stage of the relaxation, which was understood to be such instant of time, at which the electron is created at this state. The time dependence of the electronic relaxation in quantum dots has not yet been given much attention in theory. In this paper we present the numerical evaluation of the time dependence of the relaxation. We point out several features which are seen from the numerical data, concerning the final stage of the relaxation. In particular, we demonstrate theoretically the effect of the rapid decrease of the population of the ground state energy level in the quantum dot and an appearance of electronic population in the excited state, after resonantly exciting the electron to the ground state (an upconversion). The effect of the incomplete depopulation of the electronic excited state in quantum dot,
II. THE TRANSPORT EQUATION

Similarly, as it was done in the previous works, we restrict ourselves to a simple model of the electronic and phonon structure of the quantum dot. First of all, we ignore completely the presence of the holes in the valence band states of the dot. Although the holes may be simply assumed as being in thermal equilibrium with the surrounding, we simplify our model assuming the holes to be heavy enough, so that they do not influence considerably the motion of the system under consideration and contribute only to the static potential of the quantum dot.

We consider the presence of a single electron only. The quantum dot electronic orbital states, and the corresponding energies, are approximated by the eigenstates of the infinitely deep cubic quantum dot, with the lateral size \( d \) and with the material properties of GaAs crystal. We consider again the two-energy level model, assuming that the electronic ground state \( \psi_0(\mathbf{r}) \), with the index \( n = 0 \) and with the energy \( E_0 \), is given by the electronic ground state in the infinitely deep cubic quantum dot. The excited state, with \( \psi_1(\mathbf{r}) \) being the wave function of the state \( n = 1 \) with the energy \( E_1 \), is chosen to be one of the triple degenerate lowest energy excited states in this cubic dot. We put \( E_0 = 0 \) in this work.

The electron is assumed to interact with the bulk modes of the longitudinal optical phonons of lattice vibrations, neglecting in this way any influence which the interfaces in the quantum dot may have on the structure of the optical phonons. It was shown earlier that the details of the optical phonons can be neglected in heterostructures with not too small characteristic dimensions. We confine ourselves to this simple model, neglecting all other interaction, like the electronic coupling to the acoustic phonons, and other electronic interactions. Neglecting the electronic spin, the Hamiltonian of the electron-phonon system under consideration reads:

\[
H = H_0 + H_1,
\]

where \( H_0 \) is Hamiltonian of free electrons and free optical phonons,

\[
H_0 = \sum_{n=0,1} E_n c_n^+ c_n + \sum_{\mathbf{q}} b_\mathbf{q}^+ b_\mathbf{q},
\]

\( c_n \) is annihilation operator of electron in state \( n \), \( b_\mathbf{q} \) is annihilation operator of optical phonon with wave vector \( \mathbf{q} \). The electron-phonon interaction operator (Fröhlich's coupling) is

\[
H_1 = \sum_{m,n=0,1} \sum_{\mathbf{q}} A_{nm,\mathbf{q}} \Phi(n,m,\mathbf{q})(b_\mathbf{q} - b_\mathbf{q}^+) c_m^+ c_n.
\]

The relaxation rate \( dN_1/dt \) can be obtained using the nonequilibrium Green's functions method. Assuming the self-consistent Born approximation for the electronic self-energy, the instantaneous collisions approximation, the diagonal approximation for the single-particle Green's functions and distribution functions, and the Kadanoff-Baym ansatz, with the use of the equilibrium spectral densities for the interacting system, we finally get:

\[
\frac{dN_1}{dt} = -\frac{2\pi}{\hbar} \alpha_0 \left[ N_1(1 - N_0) \right. \left. \left( 1 + \nu_{LO} \right) \int_{-\infty}^{\infty} dE \sigma_1(E) \sigma_0(E - E_{LO}) + \nu_{LO} \int_{-\infty}^{\infty} dE \sigma_1(E) \sigma_0(E + E_{LO}) \right. \\
- N_0(1 - N_1) \left( 1 + \nu_{LO} \right) \int_{-\infty}^{\infty} dE \sigma_0(E) \sigma_1(E - E_{LO}) + \nu_{LO} \int_{-\infty}^{\infty} dE \sigma_0(E) \sigma_1(E + E_{LO}) \right]
\]

Einstein distribution, giving the population of the optical phonon modes at the given temperature \( T \) of the lattice.
The constant $\alpha_{mn}$ is given by the definition

$$
\alpha_{mn} = \sum_q |A_q|^2 |\Phi(n, m, q)|^2, \quad \alpha_{mn} = \alpha_{nm}. \quad (6)
$$

The spectral densities, obeying the condition $\int_{-\infty}^{\infty} \sigma_{q}(E)dE = 1$, are determined from the corresponding Green’s functions. The retarded electronic self-energy $M_n(E)$ of the state $n$ is obtained, in the self-consistent Born approximation, from the self-consistent equation

$$
M_n(E) = \sum_m \alpha_{nm} \times \left( \frac{1 - N_m + \nu_{LO}}{E - E_m - E_{LO} - M_m(E - E_{LO}) + i0^+} + \frac{N_m + \nu_{LO}}{E - E_m - E_{LO} - M_m(E + E_{LO}) + i0^+} \right) \quad (7)
$$

In the formula for the relaxation rate the spectral densities depend on the electronic density matrix by means of the equation for the self-energy. This dependence of the relaxation rate on the state of the electronic system is not easily seen in a quantitative manner from the formula and will be therefore demonstrated numerically below. Let us remark, that in the case of approximating the electronic spectral densities by delta-functions, which would be a suitable approximation in the case of very weak electron-phonon interaction, the integrals on the right hand side of would be performed and the dependence of the relaxation rate on the population of the unperturbed electronic states would be simple. This approximation would give nonzero relaxation rate only when the inter-level energy separation equals the optical phonon energy. We shall therefore not pay any further attention to this simple approximation.

III. ELECTRONIC RELAXATION

In the present approximation, in which we confine ourselves to the diagonal terms of the electronic density matrix, given by $N_n$, $n = 0, 1$, the electronic density matrix is determined by one of the terms $N_n$, because $N_0 + N_1 = 1$. This fact allows us to plot the electronic relaxation rate $-dN_1/dt$ simply as a function of $N_0$. The dependence of the relaxation rate on $N_0$ is shown in the Fig. The plot is shown for three values of the quantum dot size, in which the corresponding inter-level separation, or the detuning, is set up to be such, that $E_1 = \xi E_{LO}$, with $\xi$ being 0.7, 1 and 1.7. The value of $\xi = 1$ corresponds to the resonance between $E_1$ and $E_{LO}$. The relaxation rate is not constant as a function of $N_0$. Also, the relaxation rate depends on the magnitude of the detuning $\xi$. The relaxation rate changes sign at some values of $N_0$, which are different from 0 and 1. Another numerical result says that the dependence of the rate on the population is monotoneous. We see in Fig. that at all three cases of the detuning $\xi$ the relaxation rate becomes zero at the population, which is about between 0.1 and 0.2 at the lattice temperature of 10 K. Realizing that the optical phonon energy in GaAs is about 36.2 meV, the electronic inter-level energy separations considered in this figure are of the same order of magnitude. The value of $N_0$ in Fig. at which the rate is zero and the relaxation stops, is therefore not expected to be simply due to the equilibrium effect of the lattice temperature electronic level population.

Fig. 1 shows that the final state of the electronic relaxation from the state with the population $N_1 = 1$, at low temperatures, cannot be the state with $N_1 = 0$, but it should be another state of the system, in which the population of the ground state $n = 0$ is different from one. In other words, the electron does not relax to the ground state even in the limit of low temperatures. In Fig. 2 we plot the final population $N_{1f}$ of the excited state, which is defined as such a value of the population $N_1$, at which the relaxation rate becomes zero. The quantity $N_{1f}$ is plotted as a function of the detuning and lattice temperature. We expect that due to the population dependence of the relaxation rate, shown in Fig. the population of the state $n = 1$ will be always either decreasing at $t > 0$, if we start with $N_1 = 1$ at $t = 0$, or increasing, if we start with the population $N_1 = 0$ at $t = 0$. The population of the states will be developing towards the limiting population $N_{1f}$ plotted in Fig. 2.

The final value $N_{1f}$ of $N_1$ depends both on lattice temperature and on the detuning $\xi$. The quantity $N_{1f}$ appears to increase with increasing the lattice temperature, which is expected. The dependence of $N_{1f}$ on the detuning $\xi$ displays a certain modulation. At the integer values of $\xi$, which correspond to the resonance between the ex-
FIG. 2: The final population $N_{1f}$, at which the relaxation rate is zero, as a function of detuning $\xi$ and lattice temperature $T$.

FIG. 3: Time dependence of population $N_1$ of the higher-energy electronic state $n=1$ for the detuning $\xi=1$, ($E_1 = E_{LO}$), at lattice temperature $T=10$ K.

FIG. 4: Time evolution of the population $N_1$, of the higher-energy state $n=1$, at the detuning $\xi=0.7$.

FIG. 5: Time evolution of the population $N_1$, of the higher-energy state $n=1$, at the detuning $\xi=1.7$.

citation energy $E_1$ and a multiple of the LO phonon energy, the value of $N_{1f}$ shows a minimum, which is rather pronounced especially in the limit of low temperature and near $\xi = 1$. At higher lattice temperatures the latter resonance structure appears to weaken. The present two-level model shows that the limiting value $N_{1f}$ is especially large, achieving the magnitude of about 0.4 to 0.5, when the detuning is either much larger, or much lower than 1.

Studying the time dependence of the electronic distribution in the process of the electronic relaxation, let us first assume that the electron is prepared in the ground state $n=0$ at $t=0$. The numerical result is displayed in Fig. 3. On the time scale of picoseconds, the population of the excited state decreases with time, reaching the limiting value of the electronic population, in agreement with the data shown in Fig. 2.

It is very interesting to observe the time development of the electronic distribution in the present two-level model in the particular case when the electron is prepared in the ground state $n=0$, namely when the population $N_1 = 0$ is prepared at time $t = 0$. This initial condition may correspond to the experiments, in which time resolved depopulation of the energy levels is measured, including the time dependence of the optical properties of quantum dots after exciting the quantum dots to the lowest energy state of the electronic subsystem in a single quantum dot 17,18. The time dependence of the population $N_1$ corresponding to this initial condition is displayed in Figs. 4 and 5, in which this time dependence is shown for two values of the detuning $\xi$. From these figures it is seen that on the time scale of picoseconds the population of the electronic excited state can...
increases to about 0.2 to 0.3, which means that the population of the lower energy state can decrease to 0.8 to 0.7. This happens in the limit of low temperatures, at which the thermal population of the higher-energy states would be unexpected. From the difference between Figs. 4 and 5 we see that the characteristic time of this population upconversion slightly varies with detuning and temperature. In agreement with the data shown in the Figs. 2 the results shown in Figs. 4 and 5 demonstrate that at temperatures below about 100 K the rapidity of the population changes does not depend significantly on temperature.

IV. DISCUSSION

The present theoretical results can find a counterpart in the recent experiments in which the time-resolved response of quantum dots to short laser pulse is studied. The experimental experience shows that when a quantum dot is excited with a short laser pulse to the lowest-energy state, the subsequent relaxation process may lead to the appearance of the electronic population of electronic states with higher energy. Another effect found in experiment is: the system of quantum dot, when prepared in an excited state, relaxes fast from this state, but the relaxation ends without depopulating completely the excited state. In these experiments the depopulation effects do not show any dependence on the resonance between the electronic inter-level energy separation and optical phonon energy. These effects were recently interpreted to be a result of a non-geminate excitation of the electrons and holes in quantum dots or as a manifestation of multiphonon scattering processes including acoustic phonons.

Besides the interpretations of the origin of these depopulation effects given in the experimental papers, we propose the mechanism studied in this paper as another possible source of these effects. This mechanism assumes the absence of the phonon bottleneck effect: The formula for the relaxation rate does not contain the energy conservation delta function. This is the way how we can understand the electronic relaxation in the case of nonresonant situation between the inter-level energy separation and the phonon energy, both in the case of the relaxation from the high energy level and in the case of the relaxation of the electron from the low-energy level to the high-energy one.

The coupling of the electron to the phonon system via multiple scattering virtual transitions may cause a strong mixing of the two electronic states causes. Because of the effective lack of the energy conservation, the final state of the relaxation of the system need not to be identical with the low energy state, in the limit of zero temperature.

Summing up, we have demonstrated the time dependence of electronic relaxation in the model quantum dot with two levels and a single electron. We demonstrate the incomplete depopulation of the excited state and we also demonstrate the appearance of the population of the excited state after the resonance excitation of the ground state. We present the characteristic times of these processes. These theoretical results are discussed in relation to the recent experiments and an additional interpretation of these experiments is proposed.

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1. [kral@fzu.cz](mailto:kral@fzu.cz), corresponding author

2. A. D. Yoffe, Adv. Phys. 50, 1 (2001).

3. U. Bockelmann and G. Bastard, Phys. Rev. B 42, 8947 (1990).

4. H. Benisty, C. Sotomayor-Torres, and C. Weisbuch, Phys. Rev. B 44, 10945 (1991).

5. N. N. Ledentsov, V. M. Ustinov, V. A. Shchukin, P. S. Kop’ev, and Z. I. Alferov, Semiconductors 32, 343 (1998).

6. T. Inoshita and H. Salakai, Phys. Rev. B 56, R4355 (1997).

7. K. Král and Z. Kháš, Phys. Rev. B 57, R2061 (1998).

8. K. Král and Z. Kháš, Phys. Stat. Sol. B 204, R3 (1997).

9. K. Král and Z. Kháš, Phys. Stat. Sol. B 208, R5 (1998).

10. H. Tsuchiya and T. Miyoshi, J. Appl. Phys. 83, 2574 (1998).

11. K. Král and Z. Kháš, arXiv:cond-mat/0103061.

12. K. Král and Z. Kháš, in 24th International conference on The physics of semiconductors, edited by D. Gershoni (World Scientific, Singapore, 1999), on CD-ROM.

13. N. Terzi, in Collective excitations in solids, edited by B. DiBartolo, NATO ASI Series, Series B (Plenum Press, New York, 1983), vol. 88, pp. 149–182.

14. K. Král, Z. Kháš, C.-Y. Lin, and S. H. Lin, J. Chin. Chem. Soc. 47, 753 (2000).
15 T. Tanamoto, arXiv:quant-ph/9902031.
16 N.-J. Wu, M. Kamada, A. Natori, and H. Yasunaga, arXiv:quant-ph/9912036.
17 J. Urayama, T. B. Noris, J. Singh, and P. Bhattacharya, Phys. Rev. Lett. 86, 4930 (2001).
18 F. Quochi, M. Dinu, N. H. Bonadeo, J. Shah, L. N. Pfeiffer, K. W. West, and P. M. Platzman, Physica B 314, 263 (2002).
19 J. Urayama, T. B. Noris, H. Jiang, J. Singh, and P. Bhattacharya, Physica B 316–317, 74 (2002).
20 H. Jiang and J. Singh, Phys. Rev. B 56, 4696 (1997).
21 H. Rücker, E. Molinari, and P. Lugli, Phys. Rev. B 44, 3463 (1991).
22 K. Král and Z. Khás, in Optical properties of semiconductor nanostructures, edited by M. L. Sadowski, NATO Science Series, 3. High Technology (Kluver Academic Publishers, New York, 2000), vol. 81, pp. 149–182.
23 J. Callaway, Quantum theory of the solid state, Part B (Academic Press, New York, 1974).
24 E. M. Lifshitz and L. P. Pitaevskii, Physical kinetics (Butterworth-Heinemann, June 1981), ISBN 0750626356, reprint edition.
25 L. P. Kadanoff and G. Baym, Quantum statistical mechanics (W. A. Benjamin, Inc., New York, 1962).