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Electron self-energy in quantum dots

K. Král* and Z. Khás†

Institute of Physics, Academy of Sciences of Czech Republic, Na Slovance 2, 180 40 Prague 8, Czech Republic

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The self-energy of an electron in a quantum dot with two electronic energy levels is calculated in the self-consistent Tamm-Dancoff approximation. The electron is coupled to dispersionless bulk modes of optical phonons of GaAs. The joint spectral density, characterizing the relaxation rate of the electron between the excited and the ground state, is presented. The relation of the results to the issue of the so-called phonon bottleneck, to the width of the spectral lines of the optical transitions, and to the hot-electron relaxation in small quantum dots, is discussed. [S0163-1829(98)51404-X]

Semiconductor quantum dots are significant solid-state objects due to their discrete structure of the electronic density of states. This property makes them attractive for applications in semiconductor optoelectronics. Recent observations of the hot-electron relaxation in quantum dots lead to a discussion of a phonon bottleneck effect,[1–7] due to which the carriers are expected to stay a longer time at the excited states after being excited by a light pulse, than is usual in structures having higher dimensionality. From the simple perturbation calculation point of view, the existence of the phonon bottleneck is regarded as closely connected with the law of energy conservation in the process of the electron-phonon scattering from an excited state to a lower-energy state. In an aggregate of quantum dots with a spread of dot sizes the electronic energy-level separations are expected to only rarely match the phonon energy, especially in cases when the electrons interact primarily with the practically dispersionless longitudinal-optical (LO) phonons.

In quantum dots the electron-LO-phonon relaxation channel is very intensive[7] in a narrow interval of the electronic energy given by the energy conservation law. The hot-electron relaxation rate may then be as high as $10^{15}\ \mathrm{s}^{-1}$.[7] One can thus expect a significant role to be played by the electron-energy collision broadening due to the electron-LO-phonon coupling.[8,9]

Recently, Inoshita and Sakaki[10] calculated the energy dependence of the electronic spectral density in the ‘‘ladder’’ approximation and reported a negligible collisional broadening of the electronic energy levels. In their work[10] the Dyson equation for the electronic self-energy was solved numerically in a model of the parabolic quantum dot, considering the four lowest electronic energy levels. It is the aim of the present paper to calculate the electronic self-energy in the self-consistent Tamm-Dancoff[11] approximation, confining the electron coupling to the interaction with the dispersionless longitudinal-optical phonons of a bulk GaAs crystal, and to find an approximation under which an analytical solution of the Dyson equation can be obtained. Also, it is the aim of this work to point out some important details of the electronic self-energy and to discuss the relation of the characteristic features of the self-energy to the optical properties of quantum dots and, in comparison with the work in Ref. 10, to pay more attention to the hot-electron relaxation in small quantum dots.

In the model of the quantum dot only the electrons will be considered, neglecting the dynamics of the holes in the valence-band states. The electrons are assumed to be coupled to dispersionless longitudinal-optical phonons of the GaAs bulk crystal, neglecting other electronic couplings like the electron-electron interaction and electron-acoustic-phonon coupling. For simplicity, a quantum dot of cubic shape is considered, with GaAs inside the dot and with infinite electronic potential outside the dot. The Hamiltonian of the system then reads

$$H=\sum_{n,\sigma} E_n c^{\dagger}_{n,\sigma}c_{n,\sigma}+\sum_{\mathbf{q}} E_{\mathrm{LO}}b^{\dagger}_{\mathbf{q}}b_{\mathbf{q}}+\sum_{m,n,\sigma,\mathbf{q}} A_q\Phi(n,m,\mathbf{q}) \times(b_{\mathbf{q}}-b^{\dagger}_{-\mathbf{q}})c^{\dagger}_{n,\sigma}c_{m,\sigma}, \tag{1}$$

where Φ is the form factor

$$\Phi(n,m,\mathbf{q})=\int d^3\mathbf{r}\,\psi^{*}_n(\mathbf{r})e^{i\mathbf{q}\cdot\mathbf{r}}\psi_m(\mathbf{r}), \tag{2}$$

in which the integration spreads over the volume of the dot. $\psi_n(\mathbf{r})$, with $\mathbf{r}=(r_1,r_2,r_3)$, are unperturbed electronic eigenfunctions of the spatial motion in the quantum dot. The operator $c_{n,\sigma}$ annihilates an electron in the state with the spatial motion index n, spin σ, and energy E_n. $b_{\mathbf{q}}$ is the annihilation operator of bulk LO phonon with energy $E_{\mathrm{LO}}=\hbar\omega_{\mathrm{LO}}$.

The electron-phonon coupling constant is[12]

$$A_q=-\frac{ie}{q}\sqrt{\frac{E_{\mathrm{LO}}}{2\varepsilon_0 V}}\sqrt{\kappa_\infty^{-1}-\kappa_0^{-1}}, \tag{3}$$

where $q=|\mathbf{q}|$ is the magnitude of the phonon wave vector, and V is the volume of the sample. $-e$ is the electronic charge, ε_0 is permittivity of free space, and κ_∞ and κ_0 are high-frequency and static dielectric constants. It should be emphasized that in the heterostructures of the type of GaAs/Al$_x$Ga$_{1-x}$As the approximation of the optical lattice vibrations of the structure by the bulk modes of GaAs crystal may be very appropriate.[13] In other cases, especially those of nanocrystals dispersed in a glass matrix, or in a liquid or gas, the effects brought about by the presence of the interfaces, into the coupling of electrons with the lattice vibrations, have to be given more detailed attention.

The first term on the right-hand side of Eq. (1) gives the Hamiltonian of unperturbed electrons, with the unperturbed electronic energies E_n. In the basis of the electronic eigenstates in the quantum dot the electronic Green's function can generally be considered as a matrix. Neglecting in this work some higher-order effects that the electron-phonon coupling may have on the electronic energies in the interacting system, we shall confine ourselves to considering only diagonal terms corresponding to the spatial quantum number n. Using then Matsubara's Green's functions, as they are defined in Ref. 14, the self-energy correction $M_{n,\sigma}$ to the unperturbed energy E_n, in the self-consistent Tamm-Dancoff approximation,[11] is at the temperature T of the lattice

$$M_{n,\sigma}(i\hbar\omega_p)=k_BT\sum_{m,\mathbf{q}}|A_q|^2|\Phi(m,n,\mathbf{q})|^2\sum_r\{i\hbar(\omega_p-\omega_r)-E_m+\mu-M_{m,\sigma}[i\hbar(\omega_p-\omega_r)]\}^{-1}\times\frac{2\omega_{\mathrm{LO}}}{\hbar(\omega_{\mathrm{LO}}^2+\omega_r^2)}, \tag{4}$$

μ being the chemical potential. Here $\hbar\omega_p=(2p+1)\pi k_BT$, while the summations are performed over the frequencies $\hbar\omega_\tau=2r\pi k_BT$. p and r are integers. The self-energy is then continued analytically to the real axis of the electronic energy and the sum over r is performed.[15]

The expression under the sum over r in Eq. (4) can be regarded as a function $\phi(\hbar\omega_r)$. The corresponding function $\phi(z)$ of the complex variable z has obviously three poles at $z_1=i\hbar\omega_{\mathrm{LO}}$, $z_2=-i\hbar\omega_{\mathrm{LO}}$, and

$$z_3=\hbar\omega_p+i\{E_m-\mu+M_{m,\sigma}[i(\hbar\omega_p-z_3)]\}. \tag{5}$$

Performing the sum over r, the self-energy $M_{n,\sigma}$ appears to consist of several terms, two of which depend on z_3. The z_3-dependent terms are simplified, namely, z_3 is substituted by such a value, at which the two terms in question give their maximum contribution to the self-energy expression $M_{n,\sigma}$. This approximation has a character of making a single-pole approximation. The implications of such a simplification of the self-consistent equation for the self-energy are not studied in this paper. The electronic distribution function is approximated by Fermi-Dirac distribution with unperturbed electronic energies. Thus, in the following, $n_{m,\sigma}$ is the number of electrons at the energy level E_m, with spin σ, while ν_{LO} is the Bose-Einstein distribution function of unperturbed phonons at the temperature T of the lattice.

After continuing the result analytically to the upper half plane of energy E the equation for $M_{n,\sigma}(E)$ is obtained. Eliminating the chemical potential by a simple transformation of the real energy variable E, it is obtained that

$$M_{n,\sigma}(E)=\sum_{m,\mathbf{q}}|A_q|^2|\Phi(m,n,\mathbf{q})|^2\times\left[\frac{1-n_{m,\sigma}+\nu_{\mathrm{LO}}}{E-E_m-\hbar\omega_{\mathrm{LO}}-M_{m,\sigma}(E-\hbar\omega_{\mathrm{LO}})+i\delta}+\frac{n_{m,\sigma}+\nu_{\mathrm{LO}}}{E-E_m+\hbar\omega_{\mathrm{LO}}-M_{m,\sigma}(E+\hbar\omega_{\mathrm{LO}})+i\delta}\right],$$

$$\delta=0^{\dagger}. \tag{6}$$

Let us note that this resulting formula can be compared to the expression for self-energy studied extensively earlier in a different context of molecular excitons interacting with crystal phonons.[16] Equation (6) is also similar to the Dyson equation considered in Ref. 10.

In the present calculation the real part of the self-energy, $\mathrm{Re}\,M_{n,\sigma}(E)$ will be neglected in the expression $M_{n,\sigma}(E)=\mathrm{Re}\,M_{n,\sigma}(E)-i\,\mathrm{Im}\,M_{n,\sigma}(E)$, $\mathrm{Im}\,M_{n,\sigma}(E)\geqslant 0$. Only the imaginary part

$$\Gamma_{n,\sigma}(E)=-\mathrm{Im}\,M_{n,\sigma}(E) \tag{7}$$

will be kept. Neglecting the real part of the electronic self-energy is done on the grounds of the intuitive expectation based on the knowledge of the rather small magnitude of the polaron constant in bulk GaAs,[13] due to which the influence of the Fröhlich coupling on the electron energies in the interacting system of the quantum dot is expected to not be significant. The validity of this approximation is not tested here.

The two lowest lying (nondegenerate) energy levels of electrons in the quantum dot will be taken into consideration. The existence of other nonperturbed electronic states will be ignored. We shall denote the electronic ground state as $n=0$ and the excited state as $n=1$. We take into consideration only one state corresponding to the triply degenerate lowest excited energy level in the cubic quantum dot. Considering only one electron in the dot, the spin index in the quantity $\Gamma_{n,\sigma}$ will be dropped. In Eq. (6) only those terms will be included that give the most significant contribution in cases close to the resonance between the electronic energy level separations and the optical-phonon energy. This simplification allows one to obtain an explicit solution of the equations for the self-energy. One arrives then at the formulas

$$\Gamma_1(E)=-\alpha_{01}(1-\eta_{0,\sigma}+\nu_{\mathrm{LO}})\times\mathrm{Im}\left\{\frac{1}{E-E_0-E_{\mathrm{LO}}+i\Gamma_0(E-E_{\mathrm{LO}})+i\delta}\right\} \tag{8}$$

and

$$\Gamma_0(E)=-\alpha_{01}(n_{1,\sigma}+\nu_{\mathrm{LO}})\times\mathrm{Im}\left\{\frac{1}{E-E_1+E_{\mathrm{LO}}+i\Gamma_1(E+E_{\mathrm{LO}})+i\delta}\right\}, \tag{9}$$

with $\alpha_{01}=\Sigma_{\mathbf{q}}|A_q|^2|\Phi(0,1,\mathbf{q})|^2$. The spin index is kept in $n_{m,\sigma}$ in order to keep record of the fact that this quantity is not larger than 1.

Equations (7) and (8) for $\Gamma_1(E)$ and $\Gamma_0(E)$ can be solved analytically providing α_{01}, ν_{LO}, and $n_{m,\sigma}$ are given. In the case of a single electron in the dot $n_{0,\sigma}+n_{1,\sigma}=1$. Taking the zero of energy at E_0 and denoting $\alpha=\alpha_{01}(1-n_{0,\sigma}+\nu_{\mathrm{LO}})$, Eqs. (8) and (9) read

$$\Gamma_1(E_1+\Delta E)=\alpha\,\frac{\Gamma_0(\Delta E-x)}{(\Delta E-x)^2+\Gamma_0^2(\Delta E-x)}, \tag{10}$$

$$\Gamma_0(\Delta E-x)=\alpha\,\frac{\Gamma_1(E_1+\Delta E)}{(\Delta E)^2+\Gamma_1^2(E_1+\Delta E)}, \tag{11}$$

where $x=E_{\mathrm{LO}}-E_1$ and $\Delta E=E-E_1$. Equations (10) and (11) have the solution

$$\Gamma_1(E)=\mathrm{Re}\left\{\sqrt{\alpha\,\frac{|E-E_1|}{|E-E_{\mathrm{LO}}|}-(E-E_1)^2}\right\} \tag{12}$$

and

$$\Gamma_0(E)=\mathrm{Re}\left\{\sqrt{\alpha\,\frac{|E|}{|E-(E_1-E_{\mathrm{LO}})|}-E^2}\right\}. \tag{13}$$

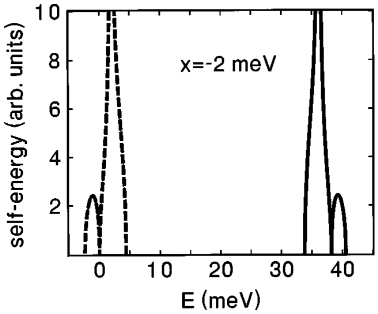

FIG. 1. Imaginary part of electronic self-energy $\Gamma_0(E)$ (dashed line) and $\Gamma_1(E)$ (full line) in a quantum dot at 77 K.

Formulas (12) and (13) give the E dependence of the imaginary part of the electronic self-energy for the case of the two electronic levels.

The numerical evaluations will be made for the above-specified simple model of the quantum dot. The electron state with $n=0$ will be $\psi_0(r_1)\psi_0(r_2)\psi_0(r_3)$, while the state with $n=1$ will be $\psi_1(r_1)\psi_0(r_2)\psi_0(r_3)$, with ψ_n being the electron wave function in a one-dimensional quantum well with infinite walls. The energy of the excited state will play the role of an independent parameter of the dot, measuring in this way the size of the dot. Besides the imaginary part of the self-energy of the states $n=0,1$ the corresponding spectral densities $D_m(E)$ of these two states will be plotted also. We define here

$$D_m(E)=\frac{\Gamma_m(E)}{(E-E_m)^2+\Gamma_m^2(E)}, \tag{14}$$

$m=0,1$.

An important quantity characterizing the electronic transitions between the excited- and the ground-state levels, with emission or absorption of the LO phonon, is the convolution of the spectral densities of the two electronic states (joint spectral density):

$$C(x)=\int_{-\infty}^{\infty} dE\, D_1(E) D_0(E-\hbar\omega_{\mathrm{LO}}). \tag{15}$$

The transition rate of electron from the excited state to the ground state would be given by a similar integral containing a statistical factor.[17] We believe that for the purpose of this work this statistical factor can be ignored. We shall regard the above formula as a characterization of the dependence of the transition rate of the electron in the excited state, due to the electron-LO-phonon coupling, on the detuning $x=E_{\mathrm{LO}}-E_1$, in other words, on the difference between the optical-phonon energy and the energy of the first electronic excited state.

In the numerical calculations, the electronic effective mass of the conduction-band Γ minimum, the values of the dielectric constants, and the energy of the optical phonon (E_{LO}=36.2 meV) of bulk GaAs are used.[18] The temperature of the lattice is 77 K. The energy dependence of the imaginary part of the self-energy, $\Gamma_0(E)$ and $\Gamma_1(E)$, for $x=-2$ meV is displayed in Fig. 1. The curve of $\Gamma_0(E)$ goes to zero at the energy $E=E_0=0$ of the unperturbed electron ground state. $\Gamma_0(E)$ diverges at energy E_1-E_{LO}. This behavior shows singular properties of the electronic self-energy in a quantum dot. In particular, a pole approximation to the self-energy cannot be introduced in a straightforward way. The curve giving $\Gamma_1(E)$ goes to zero at the energy of the unperturbed excited state $E=E_1$ and diverges at $E=E_0+E_{\mathrm{LO}}$. The features at $E=E_0+E_{\mathrm{LO}}$ and $E=E_1-E_{\mathrm{LO}}$ are single-phonon satellites, which emerge due to the coupling of a given electron state to states with a different number of phonons. Such effects have recently been discussed in quantum dots in a similar context.[19] Let us remark that the presence of these satellites in the self-energies has in fact an implication, which is the absence of the LO-phonon bottleneck effect, as it is discussed below in connection with the joint spectral density. The effect of Rabi splitting, observed in the results of Ref. 10, is not obtained here. This effect is probably lost in the course of making the approximations leading to Eqs. (8) and (9).

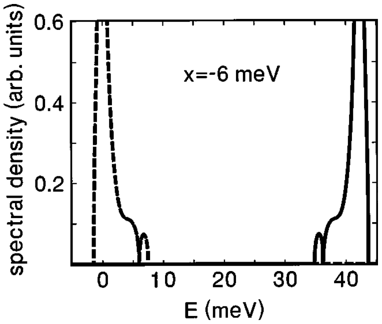

FIG. 2. Spectral density of electron interacting with LO phonons in a quantum dot. Dashed line is $D_0(E)$, full line is $D_1(E)$.

The spectral densities $D_0(E)$ and $D_1(E)$, displayed in Fig. 2 for the special case of the detuning $x=-6$ meV, show graphically the general overall property of the spectral density, namely, that the spectral density of the state 1 (or 0) always contains a feature, which is placed by E_{LO} apart from the energy E_0 (or E_1).

In Fig. 3 the spectral density $D_1(E)$ of the excited state $n=1$ is demonstrated in dependence on the detuning $x=E_{\mathrm{LO}}-E_1$. The spectral density of the ground state ($n=0$) and the spectral densities taken at negative values of x behave in an analogical manner and are not displayed in detail here. In all sections of Fig. 3 (and in Fig. 2) the maximum (a divergence) of $D_1(E)$ is found at $E=E_1$, while at E_0+E_{LO} a satellite feature of the spectral density is present, having the property of going to zero at this energy. At large x a separate satellite feature develops at E_0+E_{LO} as shown in Figs. 3(e) and 3(f). The divergencies in all the figures behave as $|E|^{-1/2}$ and they are therefore integrable.

The shape of the spectral density near the unperturbed electron energy has the form of a quite narrow peak, the overall spread of which is not larger than several meV. Such narrow peaks of the spectral density appear to be in rather good agreement with measurements of optical transitions between the conduction- and valence-band states in the quantum dots.[20]

The hot-electron relaxation rate, represented here approximately by the joint spectral density $C(x)$, does not display any abrupt behavior as a function of the detuning x. This is given in Fig. 4. Let us realize that in Born approximation the curve $C(x)$ would reduce to the function $\delta(x)$, being zero at $x\neq 0$. In the present case, the relaxation of the electron from

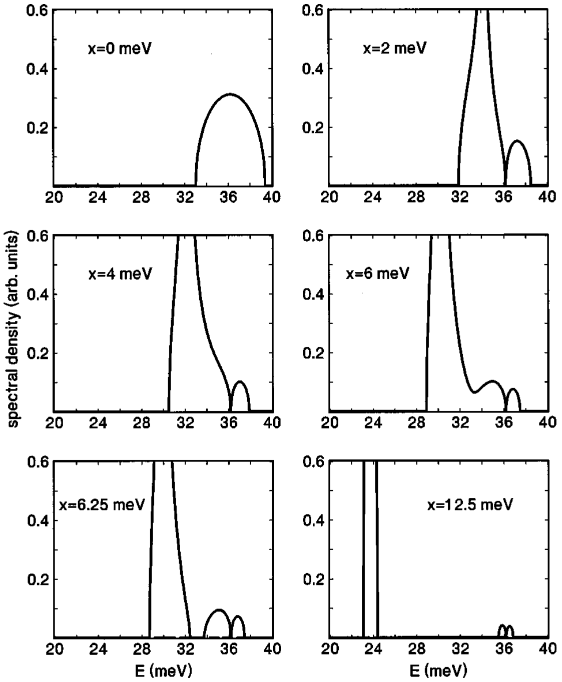

FIG. 3. Dependence of the spectral density $D_1(E)$ on the detuning x.

the excited state to the ground state, with emission of a phonon, has a form of a double maximum near the resonance case of $E_1 = E_{\mathrm{LO}}$ and decays slowly with increasing $|x|$. The nonzero values of $C(x)$ at $x \neq 0$ mean that even in quantum dots, in which the electron energy-level separations are different from E_{LO}, there may be a rapid hot-electron relaxation and the LO-phonon bottleneck may not be observed. Such a property of the hot-electron relaxation rate can be interpreted as an absence of the optical-phonon bottleneck. This finding, speaking in favor of the nonexistence of the phonon bottleneck effect, is closely connected to the presence of the satellite features in the self-energy, or spectral density, and thus to a multiphonon character of the electron states.

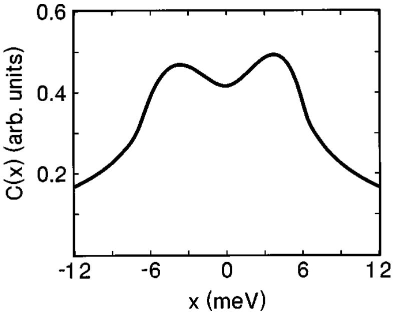

FIG. 4. Joint spectral density $C(E)$ as a function of the detuning x.

The approximation leading to Eqs. (8) and (9) suppress some multiphonon[21] features in the self-energy, which would correspond to satellites shifted by a multiple of the phonon energy from the principal features. These details of the self-energy deserve additional attention.

Besides the question of the dependence of the above results on the shape of the quantum dot, being not treated here, there is also an important problem of the dependence of the hot-electron relaxation rate, represented here by the function $C(x)$, on the choice of the material inside the quantum dot. The nonzero values of $C(x)$ at nonzero x are obviously due to the existence of the phonon satellites in the electronic spectral densities. These satellites are due to the electron-phonon coupling. The effect of the hot-electron relaxation at nonzero detuning should thus be expected to strengthen with the increase of the polaron constant of the material.

Summing up, the electronic self-energy was studied numerically in quantum dots, together with electronic spectral density and the joint spectral density. In agreement with some optical experiments, the spectral density consists of narrow peaks. On the other hand, the hot-electron relaxation rate, due to the emission of the LO phonon, appears to display a rather broad dependence on the detuning between the electron-energy-level separations and the energy of the optical phonon.

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*Electronic address: kral@fzu.cz

†Electronic address: khas@fzu.cz

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