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# Density of states and phonon-induced relaxation of electrons in semiconductor quantum dots

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To understand the energy relaxation of electrons interacting with both longitudinal-optical (LO) and longitudinal-acoustic (LA) phonons in a semiconductor quantum dot, the electron density-of-states $D(E)$ is calculated using the Green's-function method taking into account interactions of all orders and self-consistent level broadening. The $D(E)$ calculated for a GaAs dot exhibits sharp peaks (width $\lesssim 0.1$ meV ) at $T=77$ K, indicating the absence of fast relaxation in the usual sense. For level separation near, but not necessarily too close to, the LO-phonon energy, the peaks are equally narrow but split by the coherent mixing of electron levels (Rabi splitting). The LA phonons are much too weak to destroy this coherence. In the time domain, the electron undergoes rapid ($<$ ps) Rabi flopping between levels. [S0163-1829(97)51432-9]

Investigation into the physics of semiconductor quantum dots has received considerable impetus from the prediction that the use of quantum dots would produce semiconductor lasers with high efficiency due to their discrete density of states.[1] Recently, however, Benisty *et al.*[2] expressed suspicion on this view. Their argument, based on an earlier calculation of acoustic phonon-induced relaxation rate of electrons,[3] can be summarized as follows: In the process of light emission, electrons and holes initially trapped into the excited states of the active region (a well, dot, etc.), relax in cascade to the band bottoms, emitting phonons, and finally recombine to produce light. Thus, the energy relaxation of the carriers is an important process that governs light-emission efficiency. In a quantum dot, longitudinal-optical (LO) phonon emission, which is the dominant relaxation path in higher-dimensional systems (wires and wells), is forbidden due to the very discrete nature of the density of states, unless the level separation exactly matches the zone-center LO-phonon energy $\hbar\omega_{\mathrm{LO}}$. Deformation potential interaction with the longitudinal-acoustic (LA) phonons, which is already weak in the bulk, weakens further as the dot size is reduced due to decreasing form factor.[2] Thus the electrons are compelled to remain at excited levels. Hole relaxation is expected to be much faster due to the smaller level separation, and, therefore, we end up with electrons at excited levels and holes at the ground levels. Orthogonality prevents radiative recombination between an excited-level electron and a ground-level hole, resulting in poor emission efficiency, i.e., phonon bottleneck.[4,5]

The above argument is based on several simplifying assumptions and needs more careful treatment. It is important to note at this point that an electron in a dot couples only with long-wavelength phonons. In the effective-mass approximation, the electron-phonon (e-p) matrix element, for electron scattering from dot level $i$ to $j$ with absorption of $\alpha$ phonon ($\alpha=$ LO or LA) of wave vector $\boldsymbol{q}$, can be written as $M_{\mathbf{q}ij}^{(\alpha)}=\beta_q^{(\alpha)}\langle i|e^{i\mathbf{q}\cdot\mathbf{r}}|j\rangle$, where $\beta_q^{(\alpha)}$ is the usual matrix element between plane-wave states[6] (momentum transfer$=\hbar\boldsymbol{q}$), and $|i\rangle$ and $|j\rangle$ are the envelope functions for levels $i$ and $j$. Since these envelope functions typically have wavelengths on the order of the dot size, the form factor $\langle i|e^{i\mathbf{q}\cdot\mathbf{r}}|j\rangle$ is negligibly small unless $q<2\pi/(\mathrm{dot\ size})$. So the LO phonons may be considered monochromatic, and the LA phonons of energy below a cutoff $E_{\mathrm{co}}$, which is inversely proportional to the dot size, are important. (Even for a small dot of 100 Å, $E_{\mathrm{co}}\lesssim 1$ meV.) In short, we have here an interesting, nontrivial situation in which a discrete electron system interacts with (a) quasimonochromatic LO phonons (strong interaction) and (b) dispersive LA phonons with low-energy cutoff (weak interaction). The LO phonons tend to couple coherently with the electron, but the LA phonons would destroy this coherence. It is also obvious that multiple-phonon interactions are essential in this interplay between the LO and LA phonons.

In a previous paper,[7] we performed a second-order perturbation calculation of the relaxation rate and concluded that two-phonon processes LO$\pm$LA ($+$ and $-$ denote phonon emission and absorption, respectively) give rise to rapid ($<10$ ps) relaxation if the level separation is within a few meV of $\hbar\omega_{\mathrm{LO}}$, significantly alleviating the stringent energy matching required by the one-phonon (LO emission) process. The electron levels were assumed to have vanishing width: The inclusion of broadening requires a self-consistent theory and was not pursued. Furthermore, coherence effects were neglected altogether. The present paper investigates the effects of both the coherence and multiple-phonon interactions (which were neglected in Refs. 2 and 3) taking into account self-consistent level broadening. Since a direct calculation of the time evolution requires a heavy numerical calculation, we take an alternative route and calculate first the electron density-of-states $D(E)$. The $D(E)$ obtained is subsequently used to discuss electron relaxation.[8]

Let us consider an electron in GaAs confined by an isotropic and parabolic potential. (We assume parabolic confinement merely to facilitate the evaluation of matrix elements. The results are quite general and not necessarily limited parabolic dots.[9]) The electronic spectrum is therefore of the harmonic oscillator form $E_{n_x,n_y,n_z}=\Delta(n_x+n_y+n_z+3/2)$, where $n_x,n_y,n_z=0,1,2...$, and $\Delta$ is the level separation. Hereafter, for simplicity, we use a single index $n$ to represent $(n_x,n_y,n_z)$ and write $E_n$ instead of

 

$E_{n_x,n_y,n_z}$. Electron-electron and electron-hole interactions are neglected. As for the phonons, we take into account the LA and LO modes, which interact with electrons via deformation potential and Fröhlich interactions, respectively. We assume bulk GaAs phonons.[10] Only long-wavelength phonons being important, the LO mode is taken to be dispersionless and the LA modes are assumed to have linear dispersion. We calculate $D(E)$ by the Green's-function method[11] with the e-p interactions taken into account to all orders by the ladder approximation. This leads to the following Dyson equation for the retarded self-energy of the electron:

$$\Sigma_i(E)=\frac{V}{2\pi^4}\sum_{\mathscr{l}}\int_{-\infty}^{\infty}ds D_{\mathscr{l}}(s)\sum_{\alpha}\int_0^{\infty}q^2dq R_{qi\mathscr{l}}^{(\alpha)} \times\left(\frac{N_q^{(\alpha)}}{E+\hbar\omega_q^{(\alpha)}-s+i0^+}+\frac{N_q^{(\alpha)}+1}{E-\hbar\omega_q^{(\alpha)}-s+i0^+}\right), \tag{1}$$

where $V$ is the system volume, $i$ and $\mathscr{l}$ electron-level indexes, $R_{qi\mathscr{l}}^{(\alpha)}=\int d\Omega_q|M_{\boldsymbol{q}i\mathscr{l}}^{(\alpha)}|^2$ ($\int d\Omega_q$ denotes integration over the solid angle of $\boldsymbol{q}$), $\alpha$ the phonon branch index ($\alpha=$ LA or LO), $\omega_q^{(\alpha)}$ the frequency of the phonon with wave vector $q$ ($=\omega_{\rm LO}$ for LO and $cq$ for LA, $c$ being the sound velocity), phonon distribution function $N_q^{(\alpha)}=1/[\exp(\hbar\omega_q^{(\alpha)}/kT)-1]$, and the density of states (spectral function) $D_{\mathscr{l}}(E)$ for level $\mathscr{l}$ is given by

$$D_{\mathscr{l}}(E)=-2\,\mathrm{Im}[E-E_{\mathscr{l}}-\Sigma_{\mathscr{l}}(E)]^{-1}. \tag{2}$$

The e-p matrix element $M_{\boldsymbol{q}ij}^{(\alpha)}=\beta_q^{(\alpha)}\langle i|e^{i\boldsymbol{q}\cdot\boldsymbol{r}}|j\rangle$ is calculated using $\beta_q^{(\rm LA)}=A\sqrt{\hbar q/2\rho cV}$ and $\beta_q^{(\rm LO)}=M/q\sqrt{V}$,[6] where $A$ is the acoustic deformation potential $=6.8$ eV, density $\rho=5.36$ g/cm$^3$, and $c=5.15\times10^5$ cm/s. The Fröhlich coupling constant $M$ was obtained from $M=\sqrt{2\pi e^2\hbar\omega_{\rm LO}(1/\varepsilon_\infty-1/\varepsilon_0)}$ with $\hbar\omega_{\rm LO}=35.9$ meV, $\varepsilon_\infty=10.9$, and $\varepsilon_0=12.9$.

In deriving Eq. (1), the phonon system is assumed to be always in thermal equilibrium with temperature $T$, and an average is taken over the canonical ensemble of phonon baths. Thus the equation describes $D(E)$ *averaged over a collection of dots*, each being connected to a separate phonon bath. Also, the nondiagonal elements of the self-energy as to level index are found to be negligible and therefore neglected.

The coupled Eqs. (1) and (2) are solved numerically in an iterative manner. Because the LO phonons are monochromatic, we retained the full energy dependence of $\Sigma_i(E)$. The numerical calculation is facilitated by noting that the integral over $s$ in Eq. (1) is a convolution integral of the form $\int ds D(s)C(E-s)$, which can be computed by using the Fourier convolution theorem. We first solve for the imaginary part of $\Sigma_i(E)$ from Eq. (1) by Fourier transforming each of the two factors in the $s$ integral and then inverse transforming their product. Next, the real part of $\Sigma_i(E)$ was obtained by the Kramers-Krönig transformation (also a convolution integral), which can be computed in a similar way. The $\Sigma_i(E)$ thus obtained is used in Eq. (2) to get $D_i(E)$, which, in turn, is used in Eq. (1) to get an improved $\Sigma_i(E)$. This process is iterated until convergence is achieved.

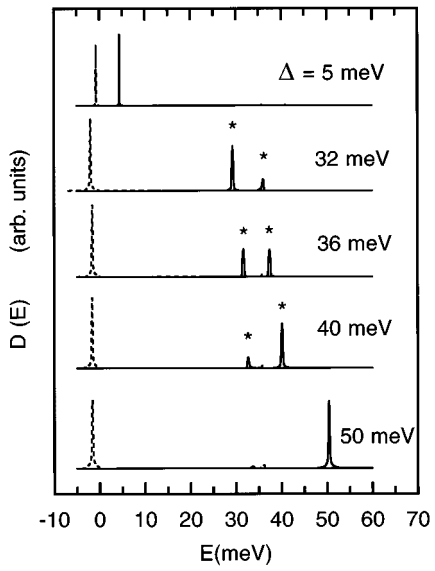


FIG. 1. Calculated density of states at 77 K for different level separations $\Delta$. The dashed and solid lines denote $D_{(0,0,0)}(E)$ and $D_{(1,0,0)}(E)=D_{(0,1,0)}(E)=D_{(0,0,1)}(E)$, respectively. Rabi splitting is indicated by asterisks.

We have included the lowest four electronic levels, i.e., the ground level (0,0,0) and the first excited levels (1,0,0),(0,1,0),(0,0,1) which are degenerate. Figure 1 shows $D_i(E)$ calculated at $T=77$ K for different values of $\Delta\propto(\text{dot size})^{-2}$. The dashed line and the solid line denote $D_{(0,0,0)}(E)$ and $D_{(1,0,0)}(E)=D_{(0,1,0)}(E)=D_{(0,0,1)}(E)$, respectively. (The e-p interactions preserve the degeneracy.) The energy $E$ is measured relative to the unperturbed ground-level energy. Excepting the case of $\Delta\approx\hbar\omega_{\rm LO}=36$ meV, $D(E)$ resembles the noninteracting density of states: The main effects of the interactions are broadening and self-energy shift. LO sidebands, seen, for example, at $\approx35$ meV for $\Delta=50$ meV, are generally weak. The peaks are invariably very sharp with the full width at half maximum on the order of 0.1 meV; it is slightly larger for the lower level, and increases gradually as $\Delta$ increases. The sharpness of the peaks indicates long electron lifetime. It is to be noted in Fig. 1 that the upper-level density of states splits when $\Delta\approx\hbar\omega_{\rm LO}$ *without broadening*.

Before going into the discussion of this splitting, a word is in order about the level width. One may be tempted to ascribe the width to lifetime broadening due to LA-phonon emission. This is not correct. We found that the broadening is caused by the LO phonons alone. The contribution of the LA phonons is orders-of-magnitude smaller. As is clear from the derivation of Eq. (1), $D(E)$ calculated here is an ensemble-averaged quantity. The dispersionless LO modes can be considered as a collection of local Einstein oscillators located at various distances from the dot: distant (nearby) oscillators cause a small (large) self-energy shift, and ensemble averaging over the oscillators results in broadening. The LO phonons interact so strongly with the electron that they overshadow the small lifetime broadening by the LA modes.

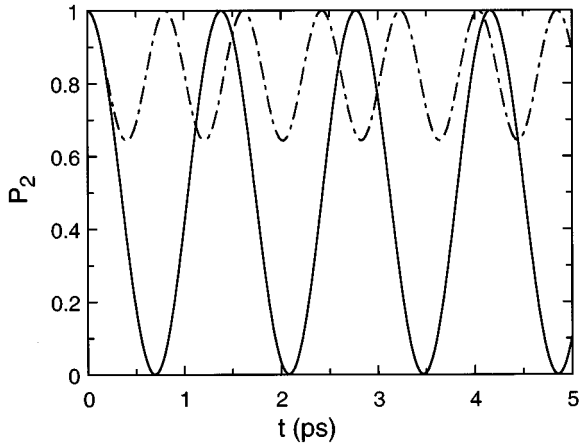


FIG. 2. Time evolution of the upper-level occupation $P_2(t)$ at 77 K for detuning $\Delta-\hbar\omega_{\rm LO}=0$ (solid line) and 4 meV (dash-dotted line) in the two-level approximation. The LA phonons are ignored, and the levels are assumed to be infinitely sharp.

The splitting of $D(E)$ when $\Delta\approx\hbar\omega_{\rm LO}$ is caused by the mixing (anticrossing) by the e-p interactions of the main (zero-phonon) upper-level peak and an LO sideband of the ground level, and known as polaron splitting in magnetodonor spectra.[12] It is also a phonon analog of the Rabi splitting familiar in the interaction of monochromatic light with an atom.[13] The absence of any additional broadening near resonance indicates the absence of additional energy relaxation contrary to the perturbation calculation,[4,7] which predicted rapid relaxation through LO$\pm$LA processes near resonance. The different conclusions derive from the neglect of coherent e-p interactions in the perturbative calculation. Ignoring the broadening, the magnitude of the Rabi splitting on exact resonance is equal to[13]

$$E_{\rm Rabi}=\sqrt{\sum_q M_q^2/V(N^{(LO)}+1)}. \tag{3}$$

($M_q$ here denotes $M_q^{\rm LO}$ taken between the pertinent levels.) Since $N^{(LO)}\ll 1$ below room temperature, $E_{\rm Rabi}$ is nearly independent of temperature.

In the time domain, the electron undergoes coherent flopping between the two levels (Rabi oscillation) with period $2\pi/E_{\rm Rabi}$.[14] This is illustrated in Fig. 2, both on resonance (detuning $=0$) and off resonance (detuning $=4$ meV), which shows the time evolution of the probability $P_2(t)$ that an electron, placed initially at the upper level at $t=0$, is found in the same level. This calculation is done in the simple two-level approximation taking into account the (0,0,0) and the (1,0,0) levels. The LA modes are neglected and the levels are assumed to be infinitely sharp. Thus the ground-level occupation $P_g$ is simply $P_g=1-P_2$. For zero detuning ($\Delta=\hbar\omega_{\rm LO}$), one has full flopping between 0 and 1. The time $\tau_{\rm flop}$ required for the electron to transfer to the lower level ($=$ half the flopping period) is less than 1 ps. Although this transition is not relaxation in the usual sense of being incoherent and irreversible, it may still be termed as ''generalized relaxation'' with relaxation time $\tau_{\rm flop}$, since radiative recombination from the ground level is possible when $P_g\neq 0$.

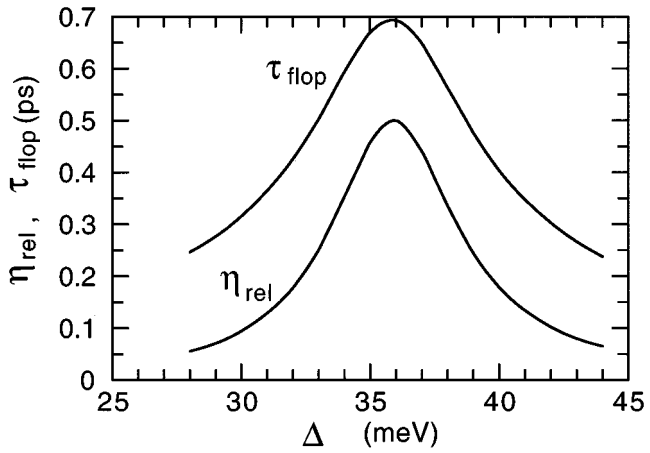


FIG. 3. Rabi flopping time $\tau_{\rm flop}$ and relative emission efficiency $\eta_{\rm rel}$ at 77 K as a function of $\Delta$.

For finite detuning, the flopping is partial, i.e., $P_g$ never reaches unity (dash-dotted line in Fig. 2): The ''relaxation time'' $\tau_{\rm flop}$, which we continue to define as half the flopping period, decreases as detuning increases. Light-emission efficiency $\eta$, however, also decreases. If we define relative light-emission efficiency $\eta_{\rm rel}$ as the ratio of $\eta$ to the ''ideal'' $\eta$ obtained if the electron relaxed infinitely rapidly (in the usual incoherent manner) to the lower level [i.e., $P_g(t>0)=1$]. $\eta_{\rm rel}=\overline{P_g}$, where $\overline{P_g}$ is the time-averaged ground-level occupation. Thus, for zero detuning, $\eta_{\rm rel}=\overline{P_g}=1/2$, and for detuning of 4 meV (Fig. 3), $\eta_{\rm rel}=\overline{P_g}\approx 1-0.82=0.18$. Figure 3 presents $\eta_{\rm rel}$ as a function of detuning, indicating that although $\eta_{\rm rel}$ decreases with detuning, the decrease is rather slow.

Figure 4 compares $D(E)$ for $\Delta=\hbar\omega_{\rm LO}$ at $T=77$ and 300 K. In spite of the broadening, the Rabi splitting is still visible at room temperature. The peaks at 300 K are strongly non-Lorentzian, reflecting the monochromaticity of the LO phonons.

Although real LO phonons are affected by confinement,

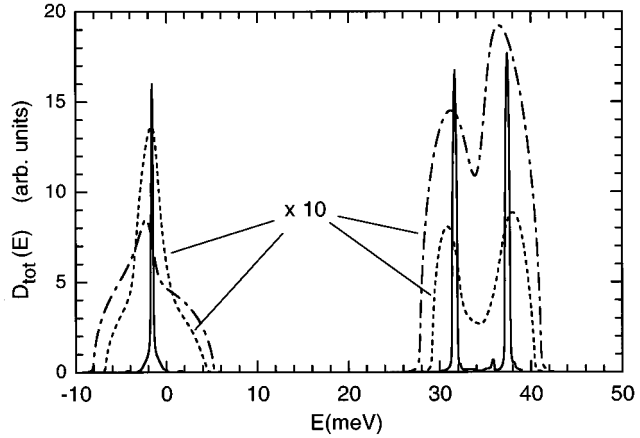


FIG. 4. Total density of states, $D_{\rm tot}=\Sigma_i D_i(E)$ for $\Delta=\hbar\omega_{\rm LO}$ calculated at $T=77$ K (solid curve) and 300 K (dash-dotted curve) with the upper-level degeneracy included [i.e., $i=(000)$, (100), (010), (001)]. For comparison, the result at 300 K *without* degeneracy [$i=(000)$, (100)] is plotted by the dashed curve.

the bulk mode approximation used here works very well for calculating the overall scattering rate.[10] A more rigorous treatment leads, for example, to Rabi splitting, Eq. (3), with the $q$ summation replaced by summation over the LO modes (confined, unconfined, and interface) if the slight energy difference (dispersion) between these modes is neglected.[15] The results are nearly equal due to a sum rule,[16] justifying the simple bulk approximation.

Figure 2 was obtained for the case of vanishing widths of electrons and phonons. Nonvanishing widths (i.e., finite phase relaxation time) would change the sinusoidal oscillation into a damped oscillation ($P_g \rightarrow 1/2$ as $t \rightarrow \infty$). In particular, if the phase relaxation time is shorter than $\tau_{\text{flop}}$ (or, equivalently, if their widths are larger than $E_{\text{Rabi}}$), the Rabi flopping is destroyed. Such rapid relaxation is unlikely for phonons at least at temperatures of order 10 K, since the polaron splitting is routinely observed in magnetodonors.[12] Phase relaxation time of an electron would be more subtle because it is sensitive to electron-electron and electron-hole interactions neglected in the present calculation: In particular, under optical excitation, a large number of electrons and holes exist in and out of the dots and might destroy the e-p coherence.[17] Then we would have the usual (incoherent) relaxation instead of the Rabi flopping. In this case, the perturbative treatment,[7] with proper inclusion of the broadening due to phase relaxation, should be valid.

In summary, if the dot level separation is near, but not necessarily too close to, $\hbar\omega_{\text{LO}}$, fairly efficient light emission at the ground level is possible through coherent Rabi flopping induced by the electron-LO phonon interaction. This pertains to the situation in which energy broadening (inverse of the phase relaxation time) in the electron/phonon system arising from mechanisms other than the electron-phonon interaction, is much smaller than the Rabi splitting. If, on the other hand, the broadening is larger than the Rabi splitting, the Rabi flopping is overtaken by the conventional (irreversible) relaxation, and the perturbative calculation of Ref. 7, with broadening taken into account, should be valid.

The Rabi splitting has been routinely observed in the absorption spectrum of magnetodonors.[12] This indicates that we should be able to observe the Rabi splitting in dots as well, as long as the disturbing effect of free electrons or holes is negligible. Such observation will provide clear evidence for ''coherent relaxation.''

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[8] Some preliminary results of the present work were reported in Ref. 4.

[9] While $M_{\mathbf{q}ij}^{(\alpha)}$ varies monotonically with $q$ for parabolic confinement, it oscillates for hard wall confinement. Aside from the oscillation, the two models give almost identical results. Therefore, the results obtained in the present work apply to self-organized dots as well.

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[11] See, e.g., G. D. Mahan, *Many-Particle Physics* (Ref. 6), Chaps. 2 and 3.

[12] See, e.g., B. D. McCombe and A. Petrou, in *Handbook on Semiconductors*, edited by T. S. Moss and M. Balkanski (Elsevier, Amsterdam, 1994), Vol. 2, p. 285.

[13] See, e.g., P. Meystre and M. Sargent III, *Elements of Quantum Optics* (Springer-Verlag, Berlin, 1990), Chap. 13.

[14] The system flops between the states $A$ and $B$ where $A$=(electron at the upper level $+m$ LO phonons) and $B$=[electron at the lower level $+(m+1)$ phonons].

[15] Even for an extremely narrow quantum well of width 20 Å, the energies of the LO modes agree to within 2.5 meV [A. K. Sood *et al.*, Phys. Rev. B **54**, 2115 (1985).] Thus, the LO dispersion is insignificant.

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