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Electron relaxation in a quantum dot: Significance of multiphonon processes

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Electron relaxation in a GaAs quantum dot is investigated to second order in electron-phonon interactions. Calculation of relaxation rate, as a function of level separation, indicates the significant contribution of LO $\pm$ LA processes, which create a window of rapid (subnanosecond) relaxation around the longitudinal-optical phonon energy. This result may provide a possible solution to the problem of photoluminescence degradation in small quantum dots.

The physics of two-dimensional (2D) semiconductor structures has been the subject of intense investigation for over 20 years and has already become an essential part of condensed matter physics. Thanks to the recent progress in microfabrication technology, there is now growing interest in systems with more confinement dimensions, i.e., 1D (quantum wire) and 0D (quantum dot) structures. A wealth of new quantum effects has already been predicted or discovered in such structures.[1-3]

Turning to the applications of these lower-dimensional systems, a major stimulus came with the prediction that lasers with high efficiency may be achieved by the use of 1D and, in particular, 0D structures.[4,5] (This is mainly because of their δ-function–like density of states.) Experimental efforts to confirm this prediction have been unsuccessful so far. In fact, experiments have shown evidence of photoluminescence degradation for dot size below $\sim$1000 Å.[6] The suspected reason was damage during the fabrication process. Recently, Benisty, Sotomayor-Torres, and Weisbuch[7] ruled out this explanation and ascribed the degradation to the slowing down of carrier relaxation with decreasing dot size, and increasing level separation.

In the process of light emission, electrons and holes (1) are first created in higher-energy continuum states, (2) relax down to the ground sublevel (assuming no recombination at excited sublevels) in cascade via phonon emission, and (3) finally recombine to emit light. Thus any realistic discussion of light emission efficiency should entail a discussion of the relaxation process. In higher-dimensional systems, e.g., quantum wells, the dominant relaxation process is longitudinal-optical (LO) phonon emission via Fröhlich interaction, with subpicosecond relaxation time. In a dot, however, this process is forbidden due to the very discrete nature of the levels, unless the level separation equals the LO phonon energy $\hbar\omega_{LO}$. (We define $\hbar\omega_{LO}$ to be the energy of the $q=0$ LO phonon.) Deformation potential interaction with longitudinal-acoustic (LA) phonons, which is already weak in the bulk compared to the Fröhlich interaction, becomes even weaker as the dot size is reduced, due to decreasing form factor. (The form factor decreases rapidly with q and, therefore, with the energy of the LA phonon emitted.) Thus relaxation via LA phonon emission in a dot is slower than in the bulk by many orders of magnitude.[8] This slowing down is more significant for electrons than holes, because the electron effective mass is usually $\sim\frac{1}{10}$ of the hole mass, and the energy separation is correspondingly larger.

This argument, though based on several simplifying assumptions, seems reasonable and convincing. However, in view of the importance of the subject, a more detailed study of the relaxation process is called for. An important element, which has so far been neglected or overlooked in the discussion of carrier relaxation in low-dimensional systems, is the multiphonon processes. The primary aim of the present paper is to investigate the electron relaxation in a semiconductor quantum dot with particular emphasis on the role of the two-phonon processes. Since hole relaxation is expected to be much faster than that of electrons, for the reason stated above, we confine ourselves to *electron* relaxation.

We take a GaAs dot as a model system and the electron confinement (barrier) potential is assumed to be isotropic and parabolic $\propto x^2+y^2+z^2$. The latter assumption of parabolic confinement is not essential, but it facilitates the calculation of matrix elements. The electronic spectrum is therefore of the harmonic-oscillator form $E_0(l+m+n)$, where $l,m,n=0,1,2,\ldots$. We consider transition between the lowest two electronic levels. Thus the initial state $|i\rangle$ and the final state $|f\rangle$ correspond to $(l,m,n)=(1,0,0)$ and $(0,0,0)$, respectively. As for the phonons, we take into account bulk LA and LO phonons of GaAs. Their dispersion relations, assumed to be isotropic, were chosen so as to fit the result of Ref. 9. Thus dispersion is included not only for the LA but also for the LO mode. Each of these modes was assumed to interact with electrons through deformation potential and Frölich interactions, respectively. The validity of this bulk phonon approximation will be briefly discussed later.

The various one- and two-phonon processes that contribute to relaxation are shown schematically in Fig. 1. Due to energy conservation, each of these processes is possible in a certain limited range of level spacing E_0, which is taken as the abscissa in the figure. The total relaxation time τ is obtained simply by the inverse summation law $1/\tau=\sum_j 1/\tau_j$, where the summation is over the various phonon processes. Of the eight possible (one-phonon and two-phonon) processes shown in Fig. 1, the first five (LO, LA, LO $\pm$ LA, and 2LA) are important and are taken into account in our calculation.

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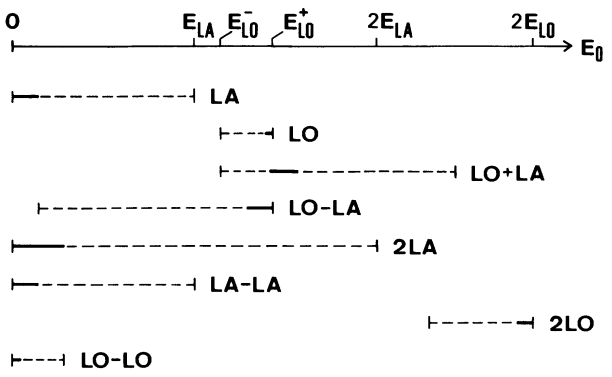

FIG. 1. Schematic diagram showing the possible phonon processes, indicated by bars, against level spacing E_0. Here, E_{LA} denotes the maximum LA phonon energy, and $E_{\overline{LO}}$ and $E_{LO}^{+}=\hbar\omega_{LO}$ are the minimum (zone edge) and maximum (zone center) LO phonon energy, respectively. Efficient carrier relaxation takes place only for energies indicated by thick solid lines.

A first-order contribution (LO or LA emission) is given simply by the golden rule as

$$\frac{1}{\tau}=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}|M_{\mathbf{q}}^{if}|^2(N_q+1)\delta(E_0-\hbar\omega_q)\,, \tag{1}$$

where N_q is the Bose distribution function $1/(e^{\hbar\omega_q/kT}-1)$. The matrix elements $M_{\mathbf{q}}^{if}$ can be written as

$$M_{\mathbf{q}}^{if}=a_q\langle i|e^{i\mathbf{q}\cdot\mathbf{r}}|f\rangle\,, \tag{2}$$

with

$$a_q=\begin{cases}D\sqrt{\hbar q/2\rho c\Omega} & \text{(LA mode)}\,,\\ M/q\sqrt{\Omega} & \text{(LO mode)}\,.\end{cases} \tag{3}$$

Here, Ω is the system volume, the deformation potential $D=6.8$ eV, density $\rho=5.36$ g/cm^3, and the sound velocity $c=5.15\times10^5$ cm/s. The Fröhlich coupling constant M was obtained from $M=[2\pi e^2\hbar\omega_{LO}(1/\epsilon_\infty-1/\epsilon_0)]^{1/2}$ with $\hbar\omega_{LO}=35.9$ meV, $\epsilon_\infty=10.9$, and $\epsilon_0=12.9$. The mode indices to $M_{\mathbf{q}}^{if}$, N_q, and ω_q are suppressed to avoid unnecessary complication. By converting the summation into an integral, Eq. (1) can be reduced to an analytic form.

The second-order contributions to τ can be obtained similarly. For the LO $\pm$ LA processes [one LO phonon is emitted and one LA phonon is emitted (+) or absorbed (−)],

$$\frac{1}{\tau}=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}\sum_{\mathbf{k}}\left|\sum_{s}{}'\left(\frac{M_{\mathbf{q}}^{is}M_{\mathbf{k}}^{sf}}{E_i-E_s-\hbar\omega_q}+\frac{M_{\mathbf{k}}^{is}M_{\mathbf{q}}^{sf}}{E_i-E_s\mp\hbar\omega_k}\right)\right|^2$$
$$\times(N_q+1)(N_k+\tfrac{1}{2}\pm\tfrac{1}{2})\delta(E_0-\hbar\omega_q\mp\hbar\omega_k)\,, \tag{4}$$

where the upper (lower) sign corresponds to LO+LA (LO−LA). In Eq. (4), quantities suffixed with $\mathbf{q}$ and $\mathbf{k}$ refer to LO and LA modes, respectively; E_s denotes energy of level s, where s represents a set of quantum numbers (l,m,n); and the prime denotes that the summation excludes the initial and final states. After some algebra, Eq. (4) can be reduced to a one-dimensional integral, which was evaluated numerically. As for the summation over intermediate states s, good convergence was achieved by taking about 40 lowest states.

If we look at the $\mathbf{q}$ dependence of the matrix elements $M_{\mathbf{q}}^{ss'}$, their behavior as $q\to0$ is governed by a_q, whereas their large q behavior is governed by the form factor $\langle s|e^{i\mathbf{q}\cdot\mathbf{r}}|s'\rangle$, which rapidly decays to zero as q goes beyond $Q=\pi/L$, L being the dot diameter. For a typical dot, L is more than an order of magnitude larger than the lattice constants, and therefore only phonons with very small q ($q<Q\ll10^{-1}$ times the zone edge wave vector) contribute to relaxation. Combining this with energy conservation ($\hbar\omega_q=E_0$ or $\hbar\omega_q\pm\hbar\omega_k=E_0$), we immediately see that rapid electron relaxation is possible only in limited ranges of E_0, which are indicated by thick solid lines in Fig. 1. Thus, as far as a perturbation expansion works for this system, efficient photoluminescence is possible only when E_0 is very small (less than a few millielectron volts), or near integral multiples of $\hbar\omega_{LO}$.

Let us now discuss the result of calculations for $E_0\simeq\hbar\omega_{LO}=35.9$ meV. Figure 2(a) shows the relaxation rate $1/\tau$ at $T=0$ K calculated as a function of E_0. The LO (one-phonon) contribution has a sharp peak immediately below $\hbar\omega_{LO}$. This peak decreases exponentially on the low-energy side, while it drops more steeply (but con-

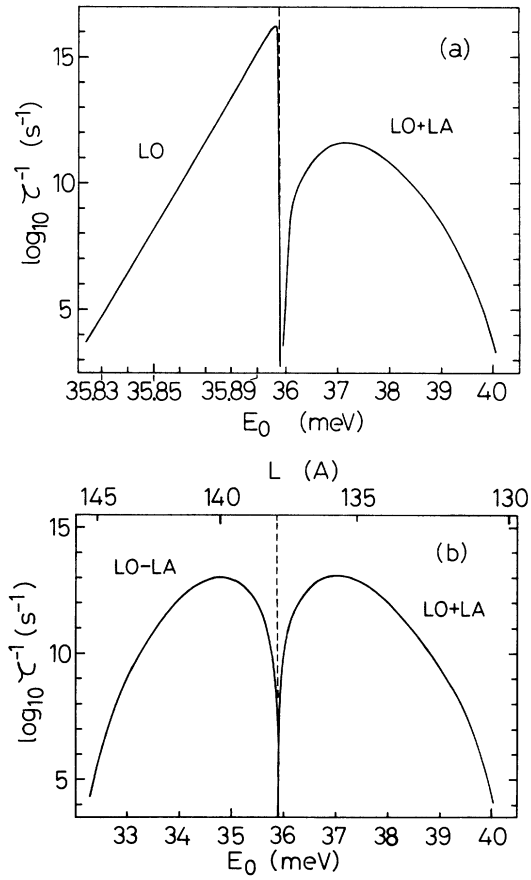

FIG. 2. Calculated electron relaxation rate $1/\tau$ for E_0 in the vicinity of $\hbar\omega_{LO}$. (a) $T=0$ K; (b) $T=300$ K. The LO peak is not shown in (b), since it is nearly the same as in (a), in this logarithmic sale. In (a), the scale in abscissa is different above and below $\hbar\omega_{LO}$, which is indicated by the vertical dashed line. Dot diameter L corresponding to E_0 is shown across the upper part.

tinuously) in the high-energy side and vanishes for $E_0 \geq \hbar\omega_{LO}$. Although the peak value exceeds 10^{15} s^{-1}, a slight detuning of E_0 from the peak dramatically reduces $1/\tau$. (For instance, $1/\tau$ is only 10^8 s^{-1} for detuning $\Delta E_0 = -0.05$ meV.) This indicates that taking advantage of the LO process requires extremely precise tuning of E_0 to $\hbar\omega_{LO}$, far more precise than is possible with current microfabrication technology. The inclusion of the second-order LO+LA process significantly alters the situation. It gives rise to a rather broad peak on the high-energy side of $\hbar\omega_{LO}$ with a peak value exceeding $\simeq 10^{11}$ s^{-1}. (Note the change in the scale in the abscissa above and below $\hbar\omega_{LO}$.) This peak value (corresponding to $\tau = 10$ ps), of course, is much smaller than that of the one-phonon peak, but it is still large enough for our purpose of having efficient light emission. (The radiative recombination lifetime τ_{rad} of electrons and holes in their ground sublevels is ~ 1 ns. Thus efficient luminescence may be achieved if $\tau < 1$ ns.) Besides, the large width of the peak significantly alleviates the tuning condition. For an electron to relax within, for instance, 1 ns, the tolerable range of E_0 is a broad $36.1 < E_0 < 38.8$ meV.

The tuning condition is further alleviated by elevating the temperature. Figure 2(b) shows $1/\tau$ at $T = 300$ K. Now it is seen that the LO $-$ LA process, which is absent at $T = 0$ K, gives rise to another broad peak on the low-energy side of $\hbar\omega_{LO}$. The peak structure is nearly the mirror image of the LO+LA peak with respect to $\hbar\omega_{LO}$. This is readily understood from Eq. (4) and the fact that the Bose function N_q for the (low-energy) LA phonons contributing here are so large ($\sim 10^2$) that $N_q + 1 \approx N_q$. By comparing Figs. 2(a) and 2(b), it is seen that the LO+LA peak is enhanced by a factor of 10^2 by the temperature increase. This, again, reflects the enhanced Bose function of LA phonons. Thus, at 300 K, the large Bose function and the emergence of the LO $-$ LA peak act to increase the tolerance in detuning. For τ to satisfy $\tau < 1$ ns, the tolerable range of E_0 is now $33 < E_0 < 39.1$ meV. To be precise, there is a narrow dip in $1/\tau$ on the immediate high-energy side of $\hbar\omega_{LO}$ (which results from the decreasing LO+LA peak and the absence of LO peak), and E_0 should avoid this dip to attain efficient relaxation. In real systems, however, this dip, being so narrow, would be easily smeared out by phonon broadening, which is neglected in the present calculation. In Fig. 2(b), dot diameter L is shown across the top. This L was obtained from the relation $L = 2\langle g|x^2+y^2+z^2|g\rangle^{1/2}$ where g denotes the ground state $(l,m,n = 0)$. (Due to the parabolic confinement, the definition of L is rather arbitrary.)

Let us now turn to the discussion of the case of small E_0. Figure 3 shows calculated $1/\tau$ at $T = 0$ and 300 K. At $T = 0$ K, the LA process gives rise to a peak, with peak height of 4×10^6 s^{-1} and peak position at $E_0 = 0.07$ meV (corresponding to $L \approx 3000$ Å). Its large width, which is in marked contrast to the extremely narrow LO peak, is mainly a result of the larger dispersion of LA mode. Its small peak value, more than 2 orders of magnitude smaller than $1/\tau_{rad} = 10^9$ s^{-1}, indicates that this relaxation path can induce only weak photoluminescence. The 2LA process produces much broader but even weaker peak towards higher energy, whose peak height is less than 10^4 s^{-1}. The situation improves significantly as T is raised to 300 K. The LA peak is enhanced by more than 2 orders of magnitude (through the Bose function), and its height is over 10^9 s^{-1}, or $\tau < \tau_{rad} = 1$ ns. More impressive is the change in the 2LA peak, which is now enhanced by nearly 5 orders of magnitude and its peak reaches 10^8 s^{-1}, only an order of magnitude smaller than $1/\tau_{rad}$. Figure 3 also suggests that phonon processes of higher order (≥ 3) may be important for larger E_0 (smaller dot size), but this situation may be more adequately treated by a different, perhaps nonperturbative, method.

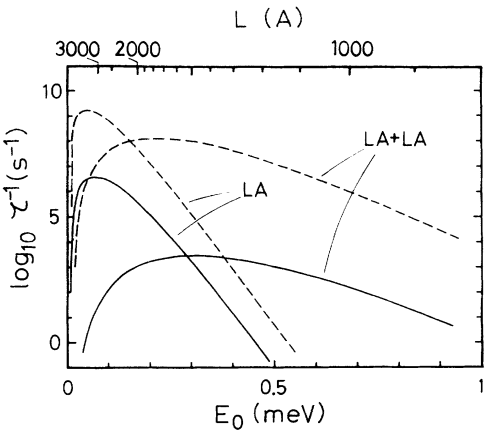

FIG. 3. Calculated electron relaxation rate $1/\tau$ for small E_0. The solid curves are for $T = 0$ K, and the dashed curves are for $T = 300$ K.

Throughout our calculation, phonons were treated as bulk modes. This is known to be quite legitimate for acoustic modes.[10] However, optical modes tend to localize in heterostructures and form confined modes and interface modes.[10] The localization effect of LO phonons on electron relaxation has been a matter of controversy for a long time. A number of macroscopic theories, with different assumptions about boundary conditions, have been advanced with conflicting results.[11] This confusion seems to have been resolved by a recent fully microscopic theory by Rücker, Molinari, and Lugli[12] which concluded that electron-LO-phonon coupling in a quantum well does not differ significantly from that obtained in the bulk phonon model. (To be precise, the above authors found that in a GaAs/AlAs quantum well, the total electron relaxation rate always falls between that for bulk GaAs LO mode and that for bulk AlAs LO mode.) We believe that this conclusion for 2D applies equally well to our 0D situation.

To summarize, the electron relaxation time in a GaAs quantum dot is calculated with emphasis on the role played by two-phonon processes. For interlevel spacing $E_0 > 0.2$ meV (or dot diameter $L > 2000$ Å), multiphonon processes are generally found to provide the dominant relaxation path, with the exception of the narrow energy range $\hbar\omega_{LO} - 0.03$ meV $< E_0 < \hbar\omega_{LO}$, where LO (one-phonon) emission dominates. This range is so narrow that it would be hopelessly difficult to tune the interlevel spacing to this energy. An alternative and more realistic way

to achieve rapid relaxation is the use of the LO $\pm$ LA processes, which are effective in a wider range of E_0 near $\hbar\omega_{LO}$. To give a numerical example, relaxation time <1 ns can be achieved for E_0 within the window $|E_0-\hbar\omega_{LO}|<3$ meV (at $T=300$ K). This suggests that efficient photoluminescence and lasing from a quantum dot will be possible if a dot can be designed to have a nearly harmonic (equal-spaced) level structure, where the level spacings fall into the above window. The use of mixed crystals of multi-LO-mode type, such as $Al_xGa_{1-x}As$, will bring additional freedom into designing, since now the level spacings are required to fall into *any* of the energy windows centered at the (more than one) LO phonon frequencies.

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