Ultrafast energy relaxation in quantum dots through defect states: A lattice-relaxation approach

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We theoretically discuss the impact of defect-related rapid energy relaxation in quantum dots, by applying a lattice relaxation approach based on a multimode description for the electron phonon interaction. Our calculation for the Huang-Rhys parameters is able to show explicitly the dependence of lattice relaxation on the spatial extent of electron states. The calculated result indicates that a relaxation rate faster than picosecond can be obtained in a wide energy range of tens of meV. [S0163-1829(97)04140-4]

In late years there has been extensive interest in the physics of semiconductor quantum dots (QD's), due to the important advantages in device applications, such as the higher performance from lasers,[1,2] the strongly enhanced oscillator strengths,[3] and the optical nonlinearities,[4] etc. In particular, growing activities are in the search for energy relaxation mechanisms in quantum dots, where the discrete nature of energy levels implies a strongly reduced energy relaxation unless the level separation equals the LO phonon energy, which is the so called phonon bottleneck effect.[5,6] However, to date this bottleneck effect is very unlikely in experiments.[7–13] Some theoretical effects have been contributed to understand the rapid energy relaxation in a quantum dot, such as the Auger-like mechanism[14,15] that could allow the relaxation on picosecond time scales in the presence of a dense electron-hole plasma, and the combined LO+LA two phonon mechanism[16] which could open a rapid but narrow relaxation window of several meV around the energy separation of $\hbar\omega_{\rm LO}\pm\hbar\omega_{\rm LA}$. On the other hand, in recent experiments,[7–13] the rapid relaxation has also occurred in quantum dot with energy-level separation as large as several LO phonon energies, and at low electron-hole pair density (less than one per dot). These findings suggest strongly that a multiphonon emission (MPE) mechanism is responsible for the rapid energy relaxation in quantum dots.

Recently, Sercel and co-workers presented a phenomenological description for a possible defect-related extrinsic relaxation mechanism,[17] in terms of a single configuration coordinate to describe the lattice, and a phenomenological Huang-Rhys factor to describe the relaxation strength. The basic idea is that in quantum dot the defect states may exist widely in the nearby barrier region during practical material growth processes, thus an electron makes a transition from the higher quantum dot states or the extended states over the barrier to the defect, sequentially the defect relaxes by MPE, and the electron makes a second transition to a lower energy level of the quantum dot. In this way, the defect provides a rapid channel for the relaxation of states in quantum dot. This picture is very similar as most situations in bulk semiconductors,[18] where an electron in the conduction band is captured first into a bound impurity state, then the bound electron recombines with a hole in the valence band.

In this work, we apply the lattice relaxation (LR) theory developed by Huang and others in 1950s,[19–21] to discuss further this defect-related MPE relaxation mechanism. By using originally the deformation potential model for the electron LA phonon coupling, and the Fröhlich model for the electron LO phonon coupling, as well as adopting the parameters in GaAs material, we employ the multimode LR approach to set our study on certain fundamental level. To our knowledge, the study of detailed models in LR theory has always been a weak point. However, this kind of study is obviously important, since the detailed nature of the lattice wave function and localized electronic state is sensitively relevant to an understanding for the intensity distribution. In the present work, we will take into account the full information of coupling between electrons and phonons, as well as the localized nature of the electron states. Accordingly, we are able to show that, besides the electron phonon coupling strength, the localization extent of electronic state is crucial to cause strong lattice relaxation, which allows a large energy relaxation by MPE. By numerical calculation, we find that, even for small quantum dot with radius about 5 nm, the LR between two bound QD states is very weak, but strong LR exists between the QD states and a defect state in the nearby barrier region. Our results support favorably the phenomenological description of Sercel and co-workers in Ref. 17. In our opinion, the present study is basically interesting and valuable, since it adds qualitative insight to the lattice relaxation effects in semiconductors.

In the following, we first present a theoretical formalism for the transition rate between QD and defect states, where the electron LO-phonon coupling is treated exactly, while the electron LA-phonon interaction is treated in a strong-coupling limit. By numerical calculation, we will show the strong LR between the QD and defect states as a function of the localization extent of the defect state, and point out that the direct LR between two QD states is too weak to cause MPE. After that, the relaxation rate between the QD and defect states will be presented and discussed in detail.

We start from the standard electron phonon interaction Hamiltonian

$$H=H_e+H_L+H_{eL}, \tag{1}$$

where H_e and H_L are the uncoupled electron and lattice Hamiltonians, and H_{eL} is their linear interaction

$$H_{eL}=\sum_q V_q e^{i\mathbf{q}\cdot\mathbf{r}}Q_q. \tag{2}$$

Here and in the following, we treat the coupling of electron to LO and LA phonons in a unified form. For electron LA-phonon interaction, $V_q^2 = D^2 q \omega_q / \rho c \Omega$, where Ω is the system volume, D the deformation potential, ρ the density, and c the sound velocity. For the Fröhlich electron LO-phonon interaction $V_q^2 = 4\pi\alpha\sqrt{\hbar/2m\omega_{\rm LO}}(2\hbar\omega_{\rm LO}^3/\Omega q^2)$, with the dimensionless coupling constant $\alpha = (e^2/2\hbar\omega_{\rm LO})\sqrt{2m\omega_{\rm LO}/\hbar}(1/\epsilon_\infty - 1/\epsilon_0)$, where $\hbar\omega_{\rm LO}$ is the LO-phonon energy, ϵ_∞ and ϵ_0 are the high-frequency and static dielectric constants. Note that in Eq. (2), Q_q is a complex coordinate describing the normal mode of lattice vibration. To apply the standard LR theory straightforwardly, we separate its real and imaginary parts as $Q_q = Q_{1q} + iQ_{2q}$. In the form of $Q_{\lambda q}$ ($\lambda = 1,2$), the interaction Hamiltonian can be rewritten as

$$H_{eL} = \sum_{\lambda,q} V_q f_\lambda(\mathbf{q}\cdot\mathbf{r}) Q_{\lambda q} \equiv \sum_{\lambda,q} u_{\lambda q} Q_{\lambda q}, \tag{3}$$

with $f_1(\mathbf{q}\cdot\mathbf{r}) = \sin(\mathbf{q}\cdot\mathbf{r})$ and $f_2(\mathbf{q}\cdot\mathbf{r}) = \cos(\mathbf{q}\cdot\mathbf{r})$. Correspondingly, the lattice Hamiltonian $H_L = 1/2\Sigma_q[\dot{Q}_q^*\dot{Q}_q + \omega_q^2 Q_q^* Q_q]$ becomes $H_L = \frac{1}{2}\Sigma_{\lambda q}[\dot{Q}_{\lambda q}^2 + \omega_q^2 Q_{\lambda q}^2]$.

For the coupled electron lattice system, in the Born-Oppenheimer approximation the state can be decomposed as $\Psi_i(\mathbf{r},\{Q_{\lambda q}\}) = \psi_i(\mathbf{r},\{Q_{\lambda q}\})\Phi_i(\{Q_{\lambda q}\})$, where ψ_i and Φ_i are the electron and lattice wave functions, respectively. In the electron wave function, $Q_{\lambda q}$ only plays a role of parameter. The lattice wave function Φ_i is a direct product of states of harmonic oscillators whose oscillating equilibrium origins are influenced by the electronic state. According to Fermi's golden rule, the decay rate of the initial state Ψ_i is given by

$$W = \frac{2\pi}{\hbar}{\rm Av}_i \sum_f |\langle f|H'|i\rangle|^2 \delta\left[E_f - E_i - \sum_{\lambda q} \hbar\omega_q (n_{\lambda q}^f - n_{\lambda q}^i)\right], \tag{4}$$

where $n_{\lambda q}^i$ and $n_{\lambda q}^f$ are the initial and final phonon number, and the average over the initial phonon states and summation over the final phonon states are explicitly shown in Eq. (4). E_f (E_i) is the initial (final) electronic-state energy, including the renormalization due to lattice relaxation, i.e., $E_j = E_j^0 - \Sigma_{\lambda q}\omega_q^2/2\Delta_{j\lambda q}^2$, wehre E_j^0 is the bare-electronic-state energy, and $\Delta_{j\lambda q} = \langle\psi_j|u_{\lambda q}(\mathbf{q}\cdot\mathbf{r})|\psi_j\rangle/\omega_q^2$. Note that, in this work, we are in fact considering the transition between the QD state and the defect state in the vicinity of barrier. As a tight-binding treatment,[17] H' in Eq. (4) is the defect potential if the initial (final) state is the QD (defect) state, and H' is the QD potential vice versa. In the Condon approximation, the electronic matrix in Eq. (4), $H'_{fi} = \langle\psi_f|H'|\psi_i\rangle$, can be regarded as lattice coordinate independent. Following the standard procedure of LR to carry out the average (summation) over initial (final) phonon states,[19–21] we have

$$W = \frac{|H'_{fi}|^2}{\hbar}\int_{-\infty}^{\infty} d\mu\ e^{F(\mu)}, \tag{5}$$

with

$$F(\mu) = -i\mu(E_f - E_i) + \sum_{\lambda q}\left(\frac{\omega_q}{2\hbar}\right)\Delta_{fi\lambda q}^2 \times\left[\coth\frac{\beta\hbar\omega_q}{2}(\cos\mu\hbar\omega_q - 1) + i\sin\mu\hbar\omega_q\right], \tag{6}$$

where $\Delta_{fi\lambda q}$ is equal to $\Delta_{f\lambda q} - \Delta_{i\lambda q}$, describing the shift of the lattice normal oscillator origin before and after the transition. As mentioned previously, we have treated the electron LA-phonon and LO-phonon couplings in a unified form. Equation (6) contains the contribution of both electron LA-phonon and LO-phonon interactions. We decompose these two parts as

$$F(\mu) = F_{\rm LA}(\mu) + S_{\rm LO}\left[\coth\frac{\beta\hbar\omega_{\rm LO}}{2}(\cos\mu\hbar\omega_{\rm LO} - 1) + i\sin\mu\hbar\omega_{\rm LO}\right], \tag{7}$$

with $S_{\rm LO} = \Sigma'_{\lambda q}(\omega_{\rm LO}/2\hbar)\Delta_{fi\lambda q}^2$, where the prime means a summation only over LO-phonon modes. $F_{\rm LA}(\mu)$ has the same form of Eq. (6), except that the summation is restricted only over LA-phonon modes, which we denote by $\Sigma''_{\lambda q}$. For the LO-phonon modes with single frequency, we can treat them exactly by applying the well-known formula $e^{z\cos\theta} = \Sigma_p I_p(z)e^{ip\theta}$, where $I_p(z)$ is the imaginary argument Bessel function. For the LA-phonon modes, in the strong-coupling limit, the function $F_{\rm LA}(\mu)$ can be expanded approximately up to the second order of μ. Consequently, the integral of Eq. (5) can be carried out as

$$W = \frac{|H'_{fi}|^2}{\hbar}\sum_p e^{-S_{\rm LO}(2N_0+1)} I_p[2S_{\rm LO}\sqrt{N_0(N_0+1)}] \times\left(\frac{N_0+1}{N_0}\right)^{p/2}\left\{\left[\frac{2\pi}{S_T\overline{(\hbar\omega_{\rm LA})_T^2}}\right]^{1/2} \times\exp\left[-\frac{(E_f - E_i - p\hbar\omega_{\rm LO} - S\overline{\hbar\omega_{\rm LA}})^2}{2S_T\overline{((\hbar\omega_{\rm LA})_T^2}}\right]\right\}, \tag{8}$$

where $N_0 = (e^{\beta\hbar\omega_{\rm LO}} - 1)^{-1}$ is the Bose occupation function of LO phonons. Here we defined two LA-phonon relaxation quantities: $S\overline{\hbar\omega_{\rm LA}} = \Sigma''_{\lambda q}\omega_q^2/2\Delta_{fi\lambda q}^2$, and $S_T\overline{(\hbar\omega_{\rm LA})_T^2} = \Sigma''_{\lambda q}(\omega_q/2\hbar)\Delta_{fi\lambda q}^2(\hbar\omega_q)^2\coth(\beta\hbar\omega_q/2)$. The LO-phonon relaxation quantities $S\overline{\hbar\omega_{\rm LO}}$ and $S_T\overline{(\hbar\omega_{\rm LO})_T^2}$ can be defined similarly. These LR quantities, e.g., $S\overline{\hbar\omega}$, can be interpreted physically as the difference of lattice elastic energies of different lattice configurations owing to the different electron states, which corresponds to a most probable MPE process with the electron energy difference exactly equal to the LR energy. If the electron energy difference does not equal the LR energy, quantum transition can also happen due to the flexible MPE, but with a smaller transition rate with respect to the most probable case. It is worthwhile to point out that Eq. (8) describes the arbitrary multi-LO and LA phonons involved in nonradiative transition. For each p LO phonon process, the peak weight is proportional to the Bessel function I_p, while the multi-LA phonons manifest themselves by

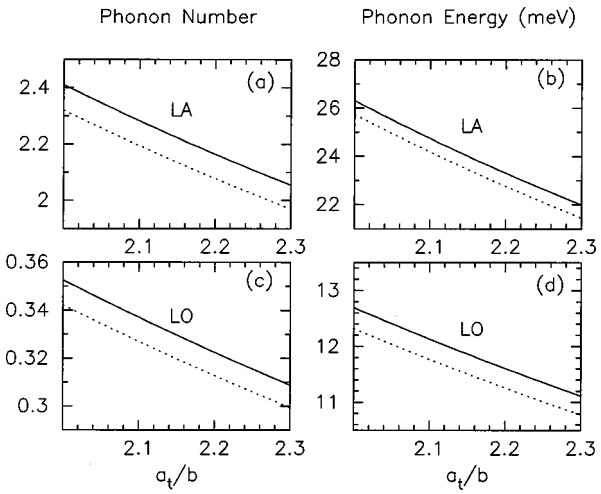

FIG. 1. LR quantities between the defect state and the QD $1s$ state (solid line) and $2s$ state (dotted line), where the average LA- and LO-phonon number and the corresponding phonon energy emitted in the relaxation process are shown as a function of spatial extent of the defect state.

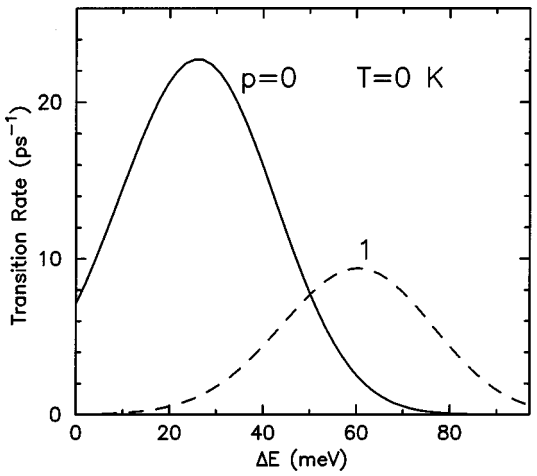

FIG. 2. Transition rate from the defect to the QD ground state as a function of their energy difference at zero temperature. The zero and one LO-phonon processes are shown by the solid and dashed curves.

shifting the p LO phonon peak by $S\overline{\hbar\omega_{\mathrm{LA}}}$ and expanding it to a width $\sqrt{2S_T(\overline{\hbar\omega_{\mathrm{LA}}})_T^2}$. At zero temperature $S_T\overline{(\hbar\omega_{\mathrm{LA}})_T^2} = S\overline{\hbar\omega_{\mathrm{LA}})^2}$, and in the high-temperature limit $S_T(\hbar\omega_{\mathrm{LA}})_T^2 \simeq 2k_BTS\overline{\hbar\omega_{\mathrm{LA}}}$. Note that the temperature dependence in Eq. (8) is more complicated than the usual second-order perturbation calculation for a LO$\pm$LA process,[16] where the temperature dependence is simply proportional to the Bose function.

To calculate the relaxation quantities in Eq. (8), the detailed nature of electronic states and their coupling to the lattice are required. In effective-mass approximation (EMA), we model the quantum dot by a finite deep spherical potential well with radius a and potential depth U_0. The solution of this simple quantum mechanical problem is well known. The order of states from the low-energy level to the higher one is $1s,1p,1d,2s,1f,...$, where s, p, d, and f denote the angular momentum. For the defect state in the nearby barrier, as in Ref. 17, we model it by a wave function $\psi_t = \sqrt{2/a_t}e^{-R/a_t}/R$, resulted from a δ trap potential, where $R = |\mathbf{r}-\mathbf{R}_0|$, $\mathbf{R_0}$ is the position of the defect that is assumed 2.5 lattice constant away from the QD interface in our calculation. For GaAs the lattice constant b is 5.65 Å. The energy level of the defect state is at $E_t = U_0 - \hbar^2/2m^*a_t^2$, with the effective mass $m^* = 0.067m_e$. In our numerical calculation, other parameters related to GaAs material are chosen as: for the quantum dot, the conduction-band offset $U_0 = 600$ meV, the dot radius $a = 5$ nm; for the deformation potential model of the electron LA-phonon interaction, the deformation potential $D = 7.8$ eV, density $\rho = 5.32$ g/cm^3, and the sound velocity $c = 5.22\times10^5$ cm/sec; for the Fröhlich electron LO-phonon interaction, the dimensionless coupling constant $\alpha = 0.07$, the LO-phonon energy $\hbar\omega_{\mathrm{LO}} = 36$ meV.

In Fig. 1 we present our calculated LR quantities between the QD and defect states, to demonstrate that the LR depends obviously on the localization extent of the defect state, where the LR quantities decrease with increasing the extension. In our calculation, the summation of phonon modes is approximated by an integral of $\mathbf{q}$ with an upper cutoff $q_c = \pi/b$, and the linear LA-phonon dispersion $\omega_q = qc$ is assumed. In Figs. 1(a)–1(d), we show the average relaxed LA-phonon number S_{LA}, LO-phonon number S_{LO}, and corresponding relaxation energies $S\overline{\hbar\omega_{\mathrm{LA}}}$ and $S\overline{\hbar\omega_{\mathrm{LO}}}$ as a function of the localization extent of the defect state, where the solid (dotted) curve is for the lattice relaxation between the QD $1s$ ($2s$) state and the defect state. Here, we notice that the LA-phonon relaxation energy of 20–30 meV is smaller than the phenomenological Huang-Rhys parameter 100 meV used by Sercel and co-workers in Ref. 17, while the relaxation LO-phonon number $S_{\mathrm{LO}} \sim 0.3$ is considerably smaller than the typical value $S_{\mathrm{LO}} \sim 5$ in the polar ionic crystal where the electron LO-phonon coupling ($\alpha > 1$) is much stronger than the present value $\alpha = 0.07$ from the GaAs material. However, in the following we will show that this LR strength is strong enough to result in a rapid energy relaxation to overcome the phonon bottleneck in a wide energy range of tens of meV.

To our knowledge, a quantitative criterion is too widely absent in the LR approach to justify the necessary localization extent of a bound state to cause strong lattice relaxation. In bulk semiconductors, the reason that the impurity state can cause strong LR may be understood by its bound-state nature. Accordingly, one may expect strong LR between the QD bound states. Unfortunately, in the present electron-phonon coupling strength, our calculation failed to show strong LR between the QD states, mainly because the nanometer scale (e.g., 10 nm) of the QD state is not so localized, compared to the defect state.

To make a conservative estimation for the time scale of relaxation, we present here the transition rate from the defect state to the QD ground state, which is slower than the transition rate from higher QD states or extended states over the barrier to the defect state, due to the smaller overlap of wave functions. Figure 2 shows the transition rate from the defect state to the QD ground state as a function of their energy difference at zero temperature, where the defect energy level changes by varying the wave-function extension a_t, and the electronic state-dependent renormalization energy has been

taken into account [see the explanation of Eq. (4)]. The $p=0$ and 1 LO-phonon-related processes are shown by the solid and dashed curves, respectively. We see that for each LO-phonon process there is a rather wide peak due to the multiple LA-phonon emission. The width of each peak is approximately $\sqrt{2S\overline{(\hbar\omega_{\mathrm{LA}})^2}}$, having the order of tens of meV. The strong relaxation rate in this wide energy range indicates a rather rapid relaxation process faster than picosecond, which means that no bottleneck effect exists if one takes into account the role of defect state during the QD state relaxing. In Fig. 2 the position of each peak is determined by the p LO-phonon emission and the average LA-phonon relaxation energy $S\overline{\hbar\omega_{\mathrm{LA}}}$, which gives rise to a separation of about $\hbar\omega_{\mathrm{LO}}$ between them. The relative height of each peak is determined by the p LO-phonon emission strength distribution, which was presented in detail in Ref. 22. At zero temperature, it satisfies the Poisson distribution, with the highest peak approximately at S_{LO}. In our case, $S_{\mathrm{LO}}\sim 0.3$, thus the zero LO-phonon peak is the highest one. Here we do not show the higher-order LO-phonon processes because of the relatively weak electron LO-phonon coupling in GaAs material.

One may have noticed an unsatisfactory feature in our zero-temperature result: for the p LO-phonon process, the transition rate is nonzero when $\Delta E<p\hbar\omega_{\mathrm{LO}}$. This unphysical problem is caused by the strong-coupling approximation in Eq. (8), when treating the LA-phonon relaxation. In Eq. (5), this kind of unphysical factor does not exist. At zero temperature, the strong-coupling approximation is valid in the energy range of $S\overline{\hbar\omega_{\mathrm{LA}}}$ around the peak, which is fortunately wide enough for the phonon bottleneck problem. At high temperatures, this approximation becomes better, where the nonzero transition rate in $\Delta E<p\hbar\omega_{\mathrm{LO}}$ is mainly due to the LA-phonon absorption. In Fig. 3 we show the transition rate from the defect to the QD ground state at room temperature $T=300$ K. We see that each p LO-phonon emission peak is further expanded as a result of the temperature effect, since the high-temperature LR quantity $S_T\overline{(\hbar\omega_{\mathrm{LA}})^2_T}$ is larger than the zero-temperature value. The height of each specific LO-phonon peak is reduced by the increased multiple LO- and LA-phonon processes at high temperatures, which can be understood easily from Eq. (8). We conclude that at high temperatures an even larger energy window is opened for the relaxation, and the phonon bottleneck effect cannot happen at all in the presence of nearby defect states in the QD barrier.

To summarize, we have described a defect-assisted rapid energy relaxation pathway for states in quantum dot, based on a multimode LR approach. Our calculation is able to show the influence of localization extent of electronic state on LR. The numerical result showed that the bound state of quantum dot with radius about 5 nm does not give rise to strong LR, but strong LR exists between the QD state and a nearby defect state in the barrier region, which results in large energy relaxation by MPE. We calculated the relaxation rate from the defect state to the QD ground state, which shows a wide energy window of tens of meV, permitting a rapid nonradiative transition faster than picosecond. Consequently, from a QD excited state to the ground state, a rapid relaxation rate exists in the presence of a nearby defect state with energy between their energy levels, since the capture rate from the QD excited state to the defect is expected to be even faster than the transition rate from the defect state to the QD ground state. In practice, this extrinsic energy relaxation mechanism may be utilized positively to overcome the phonon bottleneck problem, thus to enhance the luminescence efficiency of quantum dot structures.

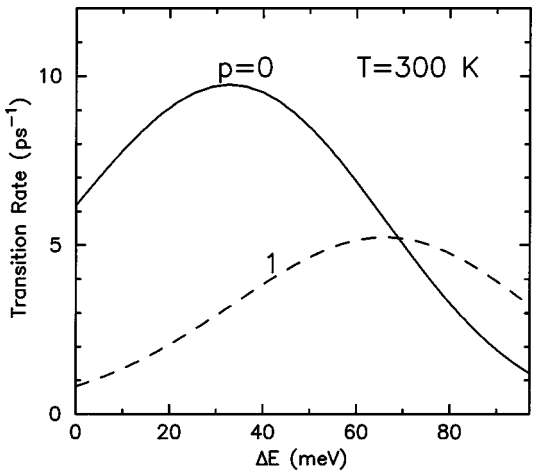

FIG. 3. Transition rate from the defect to the QD ground state as a function of their energy difference at $T=300$ K.

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