Phonon bottleneck in quantum dots: Role of lifetime of the confined optical phonons

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The phonon bottleneck in quantum dots is reexamined theoretically within the intrinsic phonon scattering mechanism. In the coupled-mode-equation formalism, an analytical solution is derived for the carrier relaxation. The result shows that, due to the anharmonic coupling of the confined LO phonon to the bulk acoustic phonons, the carrier relaxation rate is higher than $10^{10}\,s^{-1}$ in a wide detuning range of tens of meV around the LO phonon energy, which differs remarkably from the original phonon bottleneck prediction.
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Due to the sound potential for device applications, the semiconductor quantum dots (QD's) have been receiving increasing attention in recent years. Among the most important advantages compared to the conventional semiconductor lasers, there are such properties as less temperature sensitive threshold current,[1] lower threshold current density,[2] enhanced modulation dynamics, and improved noise properties[3] for laser actions, due to the three-dimensional confinement, which gives rise to a sharply discrete electron energy spectrum. However, the realization of these device advantages relies in large extent on the efficient carrier relaxation to the ground state. Thus, the study of energy relaxation mechanism in quantum dots is of central importance. In quantum dots, due to the discrete nature of the energy levels together with the very weak energy dispersion of the LO phonons, a simple consideration on the basis of energy conservation predicted that a strongly reduced energy relaxation rate could not be avoided within the intrinsic phonon scattering mechanism,[4–7] unless the electron level spacing equals the LO-phonon energy, or smaller than a few meV in favor of the LA-phonon scattering. This largely reduced relaxation rate from the inefficient phonon scattering in quantum dots is referred to as phonon bottleneck in literatures.

We noticed that this bottleneck, until now, is still discussed controversially. On the one hand, some experiments showed the poor luminescence from the ground state of quantum dot,[8–11] which implied the existence of phonon bottleneck. On the other hand, recently there have been a large number of publications where the bottleneck effect does not exist.[12–19] At the same time, in view of the tendency that the intrinsic phonon scattering cannot remove the problem of phonon bottleneck, some theoretical efforts have been proposed in specific situations, in an attempt to predict a rapid energy relaxation in quantum dots, such as the combined LO$\pm$LA two-phonon mechanism,[20] the defect-assisted multiphonon emission mechanism,[21,22] and the Auger-like scattering mechanism.[23,24] The LO$\pm$LA mechanism is an intrinsic phonon scattering mechanism, but it predicted only a quite narrow relaxation window of several meV around the LO-phonon energy; the other mechanisms[21–24] predicted a much wider relaxation window, but they are extrinsic or nonphonon scattering mechanisms, which do not work in some situations, e.g., in the absence of defects in the nearby barriers, or in the absence of dense electron-hole plasma as stated in some experiments.[14,19]

In this paper, restricted in the intrinsic phonon scattering mechanism, we reexamine the phonon bottleneck by including the anharmonic decay of LO phonons into bulk acoustic phonons, which is well known to be of crucial importance to large number of physical properties in semiconductors, such as the carrier relaxation dynamics and the carrier-lattice thermalization. Very recently, we performed a theoretical calculation for the anharmonic decay lifetime of the confined LO phonons in quantum dot,[25] which has value of 2.5–7 ps for a GaAs dot with size larger than 15 nm, depending on the temperature from zero to 300 K. Considering the confinement and decay feature of the LO phonons in quantum dot, the present coupled electron-LO-phonon system is quite similar to the coupled atom-photon system in an optical microcavity, where the enhanced spontaneous emission has been discussed, due to the confinement and dissipation of the photons. Accordingly, we are ready to develop a Wigner-Weisskopf description for the carrier relaxation in quantum dot through LO-phonon scattering: the electron couples directly to the confined LO-phonon modes, quantum transition would result in a repeated energy exchange between the electron and phonon modes, which is known as Rabi oscillation; however, due to the decay of the confined LO phonons, this oscillation will decay rapidly, thus the electron's energy is dissipated away through the LO phonons.

More specifically, our calculated result illustrates an efficient relaxation rate higher than $10^{10}\,s^{-1}$ in a wide detuning energy range of tens of meV around the LO-phonon energy, which may lead to the following implications: (i) It extends the efficient relaxation range to tens of meV from the electron level spacing smaller than a few meV ($\sim$3 meV) predicted originally by the LA phonon scattering,[4–7] or several meV ($\sim$6 meV at 300 K) detuning around the LO-phonon energy predicted from the LO$\pm$LA second-order scattering calculation.[20] (ii) Note that the LA-phonon scattering calculations[4–7] predicted an intrinsic limitation to the device application associated with the interband optical transitions, since the QD-based device applications require an efficient relaxation of the carriers to the ground state where the level spacing should be larger than 25 meV in order to realize the advantages of the discrete levels. Favorably, our present result indicates that the carrier relaxation will be efficient for level spacing approximately from 15 to 65 meV at room temperature, which improves the possibility of device appli-

cations. (iii) The present study might be helpful to clarify some confusions in literatures.

Below, considering the discrete nature of the present system (both the electron and the confined LO-phonon states in quantum dots), instead of the Fermi golden rule, we employ the Wigner-Weisskopf coupled-mode-equation approach to calculate the electron relaxation rate. In this formalism, we are able to derive an analytical solution for the time-dependent transition probability, from which the relaxation rate can be readily deduced. Specifically, consider the electron relaxation from the first excited state $|\Psi_e;\{n_k\}\rangle$ to the lowest ground state $|\Psi_g;\{(n+1)_k\}\rangle$, where $(n+1)_k$ indicates the emission of a k-mode LO phonon. Evaluating from the initial state $|\Psi_e;\{n_k\}\rangle$, the time-dependent state can be expressed as

$$\Psi(t)=a(t)e^{-iE_e t}|\Psi_e;\{n_k\}\rangle + \sum_k b_k(t)e^{-iE_g t}|\Psi_g;\{(n+1)_k\}\rangle, \quad (1)$$

where $a(t)$ and $b_k(t)$ are the corresponding amplitudes, having initial conditions $a(0)=1$ and $b_k(0)=0$, respectively. In Eq. (1), E_e and E_g are, respectively, the (total) energies of the first excited state and the ground state. We define the energy detuning $\Delta=E_e-E_g=\Delta E-\hbar\omega_0$, where ΔE is the electron energy level spacing, and $\hbar\omega_0$ is the LO-phonon energy, which is assumed to be constant for each LO-phonon mode. Substituting Eq. (1) into the time-dependent Schrödinger equation, the following coupled mode equations are obtained:

$$\frac{da(t)}{dt}=-i\sum_k g_k e^{i\Delta t}b_k(t), \quad (2)$$

$$\frac{db_k(t)}{dt}=-ig_k^* e^{-i\Delta t}a(t)-\Gamma_k b_k(t), \quad (3)$$

where Γ_k describes the anharmonic decay of the k-mode LO phonon into bulk acoustic phonons, and g_k is the coupling strength between the electron and the k-mode LO phonon. Below we present more detailed discussions for them.

Note that at thermal equilibrium the detailed balance principle requires an equal rate of the decay of LO phonons into acoustic phonons and the inverse process. Thus, Eq. (2) has no dissipation term corresponding to the function $a(t)$. On the contrary, Eq. (3) consists of a decay term corresponding to the function $b_k(t)$, which is resulted from the destruction of the thermal equilibrium condition of the LO phonons, after a new LO phonon is generated accompanying the electron transition. Recently, in Ref. 25, the lifetime of confined optical phonons in semiconductor quantum dots has been calculated by considering the typical channel of the anharmonic decaying into two bulk acoustic phonons. The calculated results show that the lifetime of the confined LO phonons is weakly size dependent, which decreases with increasing the dot size, and each mode approaches a constant value (for GaAs dot, ~7 ps at zero temperature, and ~2.5 ps at temperature 300 K) after the dot radius is larger than 8 nm. These results are in good agreement with the experimental data in bulk GaAs.[26] On the basis of Ref. 25, in this work we assume a constant lifetime τ_{ph} $(=\Gamma_k^{-1})$ for each confined LO mode, i.e., 2.5 ps at room temperature and 7 ps at zero temperature, since the concerned quantum dot has size around 20 nm.

To estimate the coupling strength g_k in Eqs. (2) and (3), the knowledge of the confined LO modes is necessary. In Ref. 27, a continuum hydrodynamic theory was employed to study the confinement of optical phonons in a spherical quantum dot, where the material-related realistic boundary conditions were treated in a rather serious sense. To satisfy both the hydrodynamic mechanical and the electromagnetic boundary conditions, say, the continuities of the normal component of vibrating velocity, the hydrodynamic pressure, the electric potential, and the normal component of electric displacement vector, we included an interface polariton component in addition to the LO vibration. Although it was found that the interface polariton component has effect on the phonon associated electrical potential, for our present phonon bottleneck problem a simplified model description for the confined optical phonons will be precise enough, since the final result of the relaxation time does not so sensitively depend on the coupling strength g_k.

For mathematical simplicity but not losing any physical generality, we assume a pressure-free boundary conditions for the LO-phonon confinement in a cubic quantum box with size a. Accordingly, the LO vibration eigenmode has the displacement function $\mathbf{u}(x,y,z)=\nabla\Phi(x,y,z)$, and $\Phi(x,y,z)\sim\sin(k_1x)\sin(k_2y)\sin(k_3z)$, where $k_j=n_j\pi/a$, $n_j=1,2,3,\ldots$. It is easy to check that this solution satisfies the mechanical pressure-free and the associated electrical potential continuity boundary conditions. Consequently, the Fröhlich electron LO-phonon interaction is given by

$$H'=\sum_k \sqrt{\frac{\hbar}{2\omega_0}}V_k\sin(k_1x)\sin(k_2y)\sin(k_3z)(\hat{a}_k+\hat{a}_k^\dagger), \quad (4)$$

where $\hat{a}_k$ $(\hat{a}_k^\dagger)$ is the annihilation (creation) operator of the k-mode LO phonon, the Fröhlich coupling strength $V_k^2=4\pi\alpha\sqrt{\hbar/2m\omega_0}(16\hbar\omega_0^3/a^3k^2)$ with $k^2=k_1^2+k_2^2+k_3^2$ and the dimensionless polaron parameter $\alpha=(e^2/2\hbar\omega_0)\sqrt{2m\omega_0/\hbar}(1/\epsilon_\infty-1/\epsilon_0)$, where ϵ_∞ and ϵ_0 are the high frequency and static dielectric constants. Now, we are ready to express the coupling strength in Eqs. (2) and (3) as $g_k=\langle\Psi_g;\{(n+1)_k\}|H'|\Psi_e;\{n_k\}\rangle$. Considering a GaAs quantum box, and assuming an infinite deep confining potential for the conduction-band electrons, we estimate the total coupling strength of the electron to all the LO modes as

$$g=\frac{0.35\hbar\omega_0}{\sqrt{a}}\sqrt{N_B+1}, \quad (5)$$

from $g^2\equiv\Sigma_k|g_k|^2$, where the dot size a is in unit of nm, and N_B is the LO-phonon number at the concerned temperature. The numerical factor 0.35 is obtained by a convergent summation over more than 10^3 confined LO-phonon modes.

To obtain an explicit solution of $a(t)$, substituting the formal solution of $b_k(t)$ from Eq. (3) into Eq. (2), we get

$$\frac{da(t)}{dt}=-\sum_k |g_k|^2\int_0^t dt' e^{i\Delta(t-t')}e^{-\Gamma(t-t')}a(t'), \quad (6)$$

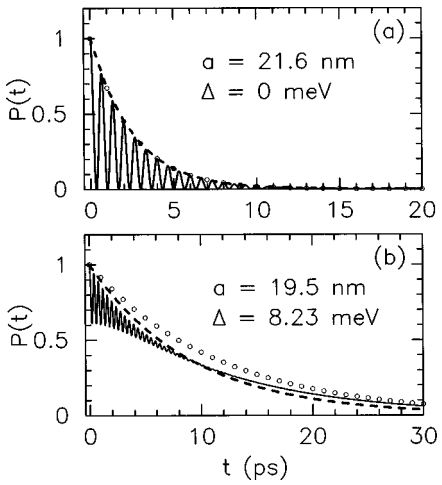

FIG. 1. Probability of the initial electron state, where the solid curves plot the exact solution Eq. (9), the dashed curves represent the $1/e$ fitting to determine the carrier relaxation time. The small circles are plotted using $e^{-(\Gamma-2\beta_2)t}$ to underestimate the entire envelope function of $P(t)$, which would set a lower bound to the relaxation rate. The LO-phonon lifetime is 2.5 ps at temperature 300 K.

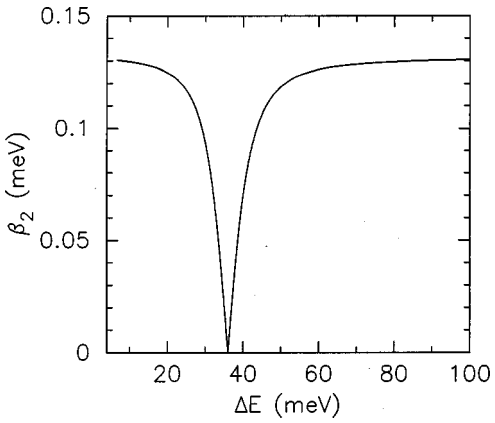

FIG. 2. β_2 (see text for its definition) as a function of the electron level spacing ΔE.

where we have neglected the dispersion of the LO phonons ($\Delta=\Delta_k$), and assumed an identical lifetime for each LO-phonon mode ($\Gamma=\Gamma_k$). In terms of Laplace transform, the solution of Eq. (6) can be derived as

$$A(s)\equiv\mathcal{L}[a(t)]=\frac{s-\gamma}{s^2-\gamma s+g^2}, \tag{7}$$

where $\gamma=-\Gamma+i\Delta$. Straightforwardly, the inverse Laplace transform yields

$$a(t)=\mathcal{L}^{-1}[A(s)]=e^{\gamma t/2}\left[\cos\beta t-\frac{\gamma}{2\beta}\sin\beta t\right], \tag{8}$$

where β is defined from $\beta^2=g^2-\gamma^2/4$. The validity of Eq. (8) can be examined in two limiting cases: (i) If $g=0$ (there is no coupling between the electron and LO phonons), solution (8) reduces to $a(t)=1$, which means that the electron remains constantly in the initial state. (ii) If $\Gamma=0$ (the LO phonon has infinite lifetime), $|a(t)|^2=\cos^2(\beta t)+\Delta^2/4\beta^2\sin^2(\beta t)$. This solution can be alternatively derived from $|a(t)|^2=|\langle i|U(t,0)|i\rangle|^2$, where $U(t,0)$ is the quantum evolution operator, and $|i\rangle$ is the initial state at $t=0$. Generally, for $g\neq0$ and $\Gamma\neq0$, we express explicitly the probability of the initial state as

$$\begin{aligned}P(t)\equiv|a(t)|^2=&\frac{e^{-\Gamma t}}{16|\beta|^2}[|2\beta-i\gamma|^2e^{2\beta_2 t}+|2\beta+i\gamma|^2e^{-2\beta_2 t}\\&+2(4|\beta|^2-|\gamma|^2)\cos(2\beta_1 t)\\&+8(\Gamma\beta_1-\Delta\beta_2)\sin(2\beta_1 t)],\end{aligned} \tag{9}$$

where β_1 and β_2 are, respectively, the real and imaginary part of β.

In Fig. 1, the solid curves plotted from Eq. (9) show the time-dependent behavior of the relaxation of the initial state. Here we consider a GaAs dot at temperature 300 K, the corresponding LO-phonon lifetime is 2.5 ps. In Fig. 1(a), the electron relaxation from the first excited state to the ground state is shown for energy detuning $\Delta=0$, while in Fig. 1(b) we show the relaxation for the nonzero energy detuning $\Delta\neq0$. Due to the finite lifetime of the LO phonons, we observe a decaying Rabi-like oscillation, however, which decays more slowly with increasing the detuning. To determine the relaxation rate, we need a proper definition for the relaxation time. We use an exponential function $e^{-t/\tau}$ to approximate the exact envelope function, where τ is determined such that at this time the envelope function has value $1/e$. In Fig. 1 we show this $1/e$ fitting by the dashed curves. We notice that this $1/e$ fitting is exact for the zero detuning, but it overestimates slightly the relaxation for the nonzero detuning. To set a lower bound to the relaxation rate, by observing the dominant contribution of the first term in Eq. (9), we apply $e^{-(\Gamma-2\beta_2)t}$ to approximate the entire envelope function of $P(t)$, which gives a simple analytical formula for the relaxation rate

$$\tau'\equiv\frac{1}{\Gamma-2\beta_2}=\frac{1}{\Gamma-\sqrt{2(R-X)}}, \tag{10}$$

where $R=\sqrt{X^2+Y^2}$, and $X=g^2+(\Delta^2-\Gamma^2)/4$, $Y=\Gamma\Delta/2$. Obviously, this approximation underestimates the relaxation rate, and the fitting result is plotted by the small circles in Fig. 1. From the following Fig. 3 we will see these two approximations differ slightly and do not influence the physical conclusion. We here emphasize that the envelope function of $P(t)$ is not an exact exponential function, thus no *mathematical* relaxation time can be defined exactly, however, any better exponential fitting must drop between the two bounds shown in Fig. 3.

To understand more clearly the dependence of the carrier relaxation rate on the detuning, in Fig. 2 we plot β_2 as a function of the the electron level spacing ΔE illustratively. Since β_2 is an increasing function of the detuning $|\Delta|$, the dominant term of Eq. (9) is a slower exponentially decreasing function for larger $|\Delta|$, which implies a smaller relaxation rate.

In Fig. 3 the relaxation rate from the first excited state to the ground state is shown as a function of the level spacing, where the zero and room temperatures are considered (the corresponding LO-phonon lifetime are, respectively, τ_{ph}

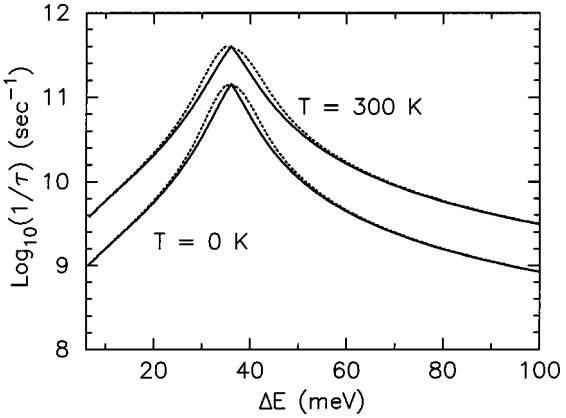

FIG. 3. Electron relaxation rate from the first excited state to the ground state as a function of the level spacing ΔE, where the solid and dotted curves are obtained from Eq. (10) and the $1/e$ fitting, respectively. Note that any better fitting for the relaxation rate in the present Wigner-Weisskopf approach will drop between these two bounds, since these two fitting approaches either overestimate or underestimate the relaxation rate. Here we see the present fittings are satisfactory, any further improvements will not change the physical conclusion.

$=7\,\mathrm{ps}$ and $\tau_{ph}=2.5\,\mathrm{ps}$), and the solid and dotted curves are obtained from Eq. (10) and the $1/e$ fitting (note that any better fitting of the relaxation rate in the present Wigner-Weisskopf approach will drop between these two bounds). Since the radiative recombination lifetime τ_{rad} from the conduction-band electrons to the valence-band holes is typically of $\sim$1 ns, the relaxation of the conduction electrons can be regarded as efficient if the relaxation rate is higher than $10^{10}\,\mathrm{s}^{-1}$. From Fig. 3 we achieve an efficient detuning range of $\sim$20 meV at zero temperature, and of $\sim$50 meV at room temperature. We stress that this result is qualitatively different from the earlier prediction for the extremely slowed down relaxation resulting from the LA-phonon scattering after the electron level spacing is larger than a few meV.[4–7] The present result also differs considerably from the result reported in Ref. 20, where although the LO$\pm$LA multiphonon scattering mechanism seems intuitively beneficial, the obtained window around the LO-phonon energy for rapid relaxation rate higher than $10^{10}\,\mathrm{s}^{-1}$ is only $\sim$3 meV at zero temperature, and $\sim$6 meV at room temperature. To our knowledge, this is the first time to demonstrate such a favorable detuning window for efficient carrier relaxation in quantum dots, within the pure intrinsic phonon scattering mechanism, which implies that the efficient photoluminescence and lasing from the ground state in quantum dots have no intrinsic restriction due to the discrete electron energy levels, in a relatively wide energy range, which was predicted to be a forbidden regime in the earlier studies.

It would be instructive to emphasize further the physical origin of the efficient carrier relaxation at wide detuning in the context of phonon bottleneck. In terms of the decaying Rabi oscillation, we have got a quite intuitive picture how the energy is exchanged between the electron and LO phonons, and eventually is dissipated away into the heat bath (i.e., the acoustic phonons). We would like to mention that the present definition for the efficient carrier relaxation is based on a device relevant criterion (i.e., the radiative recombination lifetime of the conduction electrons, which has the magnitude of nanoseconds), which sets a critical value of $10^{10}\,\mathrm{s}^{-1}$ for the *efficient relaxation*, thus makes us achieve a wide efficient detuning range of tens of meV. Alternatively, according to the definition of the usual full width at half maximum (FWHM) linewidth, from Fig. 3 one may get a linewidth of about 8 meV. However, this linewidth does not correspond to the efficient relaxation detuning range in the context of phonon bottleneck. Please note that the ''phonon bottleneck'' is not an intrinsic physical phenomenon. This phenomenon is closely based on some other parameters such as the radiative lifetime, which thus set the corresponding criterion in certain extrinsic sense.

One may note that the inverse lifetime of the LO phonon corresponds to energy 1.65 meV for 2.5 ps (or 0.59 meV for 7 ps), which is approximately the FWHM linewidth of the relaxation spectrum from a Fermi golden rule argument. We would like to point out that this golden rule treatment is physically invalid in the present two level system, since the Lorentzian spectrum, resulting from the unstable feature of the LO-phonon state, differs essentially from the *real* density of states in the Fermi golden rule, and in fact no golden rule exists in this situation. Therefore, it is not suitable to compare the inverse LO-phonon lifetime with the FWHM linewidth obtained from the Wigner-Weisskopf approach. In other words, we should not be surprised to the difference of these two quantities. In this context, we would like to claim that even using the Fermi golden rule one can also obtain a wide detuning range in which the relaxation rate is larger than that determined by the ''phonon bottleneck'' criterion, due to the relatively strong coupling of the electron with the LO phonons. This may help one to understand more easily the obtained wide efficient detuning range. However, it should be noted that the Fermi golden rule is a misleading argument in the present system.

In conclusion, the electron relaxation in quantum dot has been discussed within the pure phonon scattering mechanism. Due to the anharmonic coupling of the LO phonons to the acoustic phonons, we have demonstrated an efficient carrier relaxation rate higher than $>10^{10}\,\mathrm{s}^{-1}$ in wide energy detuning of tens of meV around the LO-phonon energy, which modifies remarkably the earlier phonon-bottleneck prediction within the phonon scattering mechanism.[4–7,20] Concerning the device applications, the obtained result is of interest, since it excludes the phonon bottleneck problem in an *intrinsic* sense in a relatively wide detuning range.

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