Anharmonic decay of confined optical phonons in quantum dots

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Due to the anharmonicity of lattice forces, the lifetime of confined optical phonons in semiconductor quantum dots is studied by considering the typical channel of decay into two bulk acoustic phonons. The model in which the three-phonon coupling strength is expressed in terms of the Grüneisen constant enables us to obtain a compact expression for the lifetime from standard perturbation theory. The lifetime is shown to be weakly size dependent, which increases with decreasing dot size. For GaAs quantum dots, the results are found to be completely consistent with other theoretical approaches and experimental data in bulk GaAs. Furthermore, we present a brief discussion for the carrier relaxation in quantum dots. Due to the decay of the confined phonons, we find that the carrier relaxation time can be as short as tens of picoseconds in a wide energy detuning of tens of meV; thus the so-called phonon-bottleneck effect is not a serious problem in quantum-dot-based device applications. [S0163-1829(98)08919-X]

I. INTRODUCTION

It is well known that lattice anharmonicities influence greatly such properties as carrier-lattice thermalization, thermal expansion and conductivity, velocity of sound, and Raman and neutron scattering.[1,2] The anharmonicity force, typically expanded to the cubic term, causes the decay of an optical phonon into two acoustic phonons and consequently a finite lifetime of the optical phonon. Because of the dominant role of electron optical-phonon coupling in polar semiconductors, detailed knowledge of the optical-phonon decay is of particular importance in the understanding of the carrier dynamics. Furthermore, the finite lifetime of optical phonons determines directly the linewidth of the optical spectrum of the defects, the infrared absorption, and the Raman scattering. For these reasons, in bulk semiconductors the lattice anharmonic effects have been studied extensively for a long time; see Refs. 1 and 2 and references quoted therein; for more recent papers see, for example, Refs. 3–5. However, much less attention has been given to semiconductor microstructures, although it has been demonstrated that in the context of semiconductor lasers the optical-phonon emission alone does not dominate determinatively the carrier energy relaxation rates since the subsequent optical-phonon decay events influence the overall relaxation process.[6,7]

In this paper, applying a phenomenological approach developed by Klemens,[1] in which the various anharmonic contributions were replaced with a single parameter, i.e., the Grüneisen constant, which characterizes the average of the third-order elastic constants, we study the lifetime of the confined optical phonons in quantum dots due to the decay into two bulk acoustic phonons. The same approach has been used to study the lifetime of the imperfection-induced localized phonons[8] and the optical phonons in bulk systems.[9,10] We will derive an expression that can be conveniently applied to practical calculations for the decay rates of optical-phonon modes confined in spherical quantum dots, where the spatial overlap integral of the three coupled phonons is simplified greatly. For the zero-angular-momentum modes, the expression is simplified further and the final result can be carried out easily. As an example of application we consider the GaAs quantum dot. The analysis of the lifetime of the zero-angular-momentum mode shows that the lifetime of the confined mode is weakly size dependent, increases with decreasing dot size, and approaches a constant value (~ 7.5 ps at zero temperature) after the dot radius is larger than 5 nm. To compare with the available measured data in bulk GaAs (7 ps at 77 K and 3.5 ps at 300 K) we consider a dot with a radius of 8 nm to study the temperature dependence of the lifetime. The obtained results are in excellent agreement with the experimental data and completely consistent with the theoretical results in Ref. 4.

There is growing activity 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 problem.[11,12] However, to date, this bottleneck effect is very unlikely in experiments. Recently, a mechanism based on multiphonon emission has been proposed both theoretically and experimentally.[13–17] More interestingly, in Ref. 18, a problem of the simple use of Fermi's golden rule for the analysis was pointed out and it was shown that the carrier energy relaxation rate can be larger than 10^{10} s^{-1} in a wide energy range of tens of meV by using the coupled mode equation derived from the time-dependent Schrödinger equation and thus the phonon bottleneck is not a serious problem in device application. In that calculation, a phonon lifetime of 2.5 ps is used, which is the present theoretical result at a temperature of 300 K. In this paper we present a more straightforward understanding of this issue, on the basis of a simplified two-level picture. We conclude that, due to the decay of the confined optical phonons, as well as the enhanced electron-phonon coupling strength in quantum dots, the carrier relaxation time can be as short as tens of picoseconds in a wide energy detuning of tens of meV; thus the well-known theoretical prediction of the phonon bottleneck effect is quite questionable, even considering only the phonon mechanism.

II. FORMALISM

To calculate the lifetime, the vibration function of the confined mode is needed. In Ref. 19 we found its solution based on the macroscopic hydrodynamic (HD) approach. To satisfy both the HD mechanical and the electromagnetic boundary conditions, say, the continuities of the normal component of vibrating velocity, the HD pressure, the electric potential, and the normal component of electric displacement vector, we included an interface polariton (IP) component in addition to the LO vibration. For both the LO and IP vibration components, we are able to introduce the mechanical potentials $\widetilde{\Phi}$ and G, respectively, which satisfy the wave equation $\nabla^2\widetilde{\Phi}+k^2\widetilde{\Phi}=0$ and the Laplace equation $\nabla^2 G=0$, where k^2 is defined by the dispersion relation $\omega^2=\omega_L^2-v^2 k^2$, with ω_L the limiting LO frequency and v the sound velocity. The vibrating displacement field $\mathbf{u}$ is related to the mechanical potential by

$$\mathbf{u}=A\nabla[\widetilde{\Phi}+BG]\equiv A\nabla\Phi,\tag{1}$$

where A and B are two constants determined by the boundary conditions and the amplitude normalization.

First we normalize the amplitudes of the confined modes in quantum dots. From the virial theorem we know the kinetic energy

$$\sum_{j=1,2}\left[\frac{M_j\omega^2}{2}\frac{1}{b_j^3}\int_{V_j}d^3\mathbf{x}(\nabla\Phi)^2\right]A^2=\frac{E_p}{2},\tag{2}$$

where V_j characterizes the inner ($j=1$) and the outer ($j=2$) volume of the dot, M_j is the cell atomic mass, b_j^3 is the cell volume, and $\omega(E_p)$ is the frequency (energy) of the mode. With the help of the vector-field-theory formula $\int_V d^3\mathbf{x}[\phi\nabla^2\psi+\nabla\phi\cdot\nabla\psi]=\oint_\Sigma d\sigma\,\phi\hat{\mathbf{n}}\cdot\nabla\psi$, where Σ is a closed surface surrounding the volume V and $\hat{\mathbf{n}}$ is the normal unit vector of the integral element $d\sigma$, the normalized constant A can be expressed as

$$A=\left[\frac{E_p}{(M_1 S_k+M_2 S_\kappa+\Delta_{12})\omega^2}\right]^{1/2}\equiv\left[\frac{E_p}{\widetilde{M}\omega^2}\right]^{1/2},\tag{3}$$

where

$$S_k=\frac{k^2}{b_1^3}\int_{V_1}d^3\mathbf{x}\,\Phi_1^2,$$

$$S_\kappa=-\frac{\kappa^2}{b_2^3}\int_{V_2}d^3\mathbf{x}\,\Phi_2^2,$$

$$\Delta_{12}=\oint_\Sigma d\sigma\,u_r\left[\frac{M_1\Phi_1}{b_1^3}-\frac{M_2\Phi_2}{b_2^3}\right].$$

Here Φ_1 and Φ_2 are the dot-inside and -outside mechanical potentials, which satisfy the wave equations $\nabla^2\Phi_1+k^2\Phi_1=0$ and $\nabla^2\Phi_2-\kappa^2\Phi_2=0$, respectively, where $-\kappa^2$ is due to the decay feature of the mode outside the dot. The radial component of the vibrating displacement at the interface is continuous, i.e., $u_r=(\partial_r\Phi_1)|_\Sigma=(\partial_r\Phi_2)|_\Sigma$. However, the mechanical potentials Φ_1 and Φ_2 can be different from each other at the interface in the HD treatment (see Ref. 19), where the boundary condition of the continuity of the electric

potential instead of the mechanical potential is applied. The discontinuity of the mechanical potential implies that the slightly tangential slide of the two materials at the interface is allowed, which is in fact an intrinsic feature of the hydrodynamic approach.

To analyze the dispersion relation of the phonons, the crystal potential is expanded only to the quadratic term, which is the harmonic approximation. However, to describe the decay of phonons, the cubic and other higher-order terms known as the anharmonic terms in the crystal potential should be taken into account. It is widely accepted that the cubic anharmonic term will dominate the phonon decay and thus the matrix element of the perturbation Hamiltonian due to the three-phonon interaction has the form[1]

$$|H'_\pm|^2=\left[\frac{\hbar^3}{\widetilde{M}M_2^2 V^2}\right]\frac{|c(\omega,\omega',\omega'')I(\mathbf{k}',\mathbf{k}'')|^2}{\omega\omega'\omega''}$$
$$\times\left[\begin{array}{c}(N+1)N'N''\\N(N'+1)(N''+1)\end{array}\right],\tag{4}$$

where N, N', and N'' are the Bose occupation numbers of the confined mode and the bulk acoustic phonons, with frequencies ω, ω', and ω'' respectively; the upper line of the matrix describes the decay process of the confined mode into the bulk acoustic phonons, whereas the lower line describes the microscopic inverse process. Other quantities in Eq. (4) are explained as follows. Since for LO phonons the decay occurs mainly through the creation of two LA phonons, the overlap integral of the three phonons with the polarization summation can be treated as

$$I(k',k'')=\sum_\alpha\int d^3\mathbf{x}\,e^{-i(\mathbf{k}'+\mathbf{k}'')\cdot\mathbf{x}}[\nabla\Phi]_\alpha,\tag{5}$$

where the index α characterizes the vibrating projection component of the confined modes. To estimate the three-phonon coupling strength, two approaches can be classified: the highly complex microscopic model with parameters that are very difficult to measure and the simple macroscopic model where various anharmonic contributions are replaced by a single parameter related to the averaged third-order elastic constants. In this work we adopt the latter approach for simplicity, in which the coefficient $c(\omega,\omega',\omega'')$ is approximately[1]

$$c(\omega,\omega',\omega'')=-i\frac{2M}{\sqrt{3}}\frac{\gamma}{v}\omega\omega'\omega'',\tag{6}$$

where γ is the Grüneisen constant, v is the sound velocity, and M is the cell atomic mass. Due to the strong confinement of the optical mode, v and M are approximately the dot-inside material values. In deriving Eq. (4) the normalized amplitudes of the phonons have been used, i.e., $(\hbar/\widetilde{M}\omega)^{1/2}\nabla\Phi$ for the confined mode and $(\hbar/M_2\omega')^{1/2}e^{-i\mathbf{k}'\cdot\mathbf{x}}$ and $(\hbar/M_2\omega'')^{1/2}e^{-i\mathbf{k}''\cdot\mathbf{x}}$ for the acoustic phonons. Because of the relatively small size of the dot, the normalization for the acoustic phonons can be regarded mainly over the outer material of the quantum dot.

Following Fermi's golden rule, the transition rate is obtained as

$$\Gamma^{\pm} = \frac{2\pi}{\hbar} \sum_{\mathbf{k}',\mathbf{k}''} |H'_{\pm}|^2 \delta(\hbar\omega - \hbar\omega' - \hbar\omega''). \qquad (7)$$

The lifetime of the confined phonons is related to both the rate Γ^+ at which they decay into two acoustic phonons and the rate Γ^- at which they are generated by the annihilation of two acoustic phonons. At thermal equilibrium, the detailed balance principle requires $\Gamma^+ - \Gamma^- \sim (N+1)N'N'' - N(N'+1)(N''+1) = 0$. We assume that the derivation from equilibrium is caused by the change of occupation of the confined phonons, i.e., $N \rightarrow N + \delta N$; hence we have

$$\Gamma^+ - \Gamma^- \sim - \delta N(N' + N'' + 1). \qquad (8)$$

Consequently, the lifetime τ can be defined as

$$\frac{1}{\tau} = -\frac{\Gamma^+ - \Gamma^-}{\delta N}, \qquad (9)$$

which gives

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} \frac{\hbar^3}{\widetilde{M} M_2^2 V^2} \sum_{\mathbf{k}',\mathbf{k}''} \frac{|c(\omega,\omega',\omega'') I(\mathbf{k}',\mathbf{k}'')|^2}{\omega \omega' \omega''}$$

$$\times (N' + N'' + 1) \delta(\hbar\omega - \hbar\omega' - \hbar\omega''). \qquad (10)$$

After replacing the summation over $\mathbf{k}'$ and $\mathbf{k}''$ by an integral and integrating out the δ function, we obtain

$$\frac{1}{\tau} = \left[\frac{4}{3} \frac{1}{(2\pi)^5} \left(\frac{M}{M_2} \right)^2 \frac{\hbar\omega}{\widetilde{M} v^2} \frac{\gamma^2}{v} \right] \int d^3\mathbf{k}'$$

$$\times \int d\Omega'' k''^2 \omega' \omega'' |I(\mathbf{k}',\mathbf{k}'')|^2 (N' + N'' + 1), \quad (11)$$

where $d\Omega''$ is the solid angle of $\mathbf{k}''$ and ω'' is related to ω' by $\omega' + \omega'' = k'v + k''v = \omega$.

Below we analyze the overlap integral $I(\mathbf{k}',\mathbf{k}'')$, see Eq. (5). Again, applying the vector-field-theory formula $\int_V \boldsymbol{\nabla} \psi \, d^3\mathbf{x} = \oint_\Sigma \psi \hat{n} \, d\sigma$, we can reexpress the overlap integral as

$$I(\mathbf{k}',\mathbf{k}'') = \sum_{\alpha} \hat{\mathbf{e}}_\alpha \cdot \left[\oint_\Sigma d\sigma \, \hat{\mathbf{n}} e^{-i(\mathbf{k}'+\mathbf{k}'') \cdot \mathbf{x}} (\Phi_1 - \Phi_2) \right.$$

$$\left. + i(\mathbf{k}' + \mathbf{k}'') \int d^3\mathbf{x} \, e^{-i(\mathbf{k}'+\mathbf{k}'') \cdot \mathbf{x}} \Phi \right]. \qquad (12)$$

As mentioned previously, the mechanical potential can be discontinuous at the interface within the hydrodynamic treatment. However, this discontinuity is very small if the electromagnetic properties of the two materials are not very different since the electric potential is in fact continuous. Furthermore, due to the approximately rigid confinement, which implies that both the electric potential and the mechanical potential are almost zero at the interface, the first term on the right-hand side of Eq. (12) can be neglected. Let us denote $\mathbf{q} \equiv \mathbf{k}' + \mathbf{k}''$ and choose $\mathbf{q}$ to be the reference direction for the projection of the vibrating amplitude of the confined mode. Thus the summation for the polarization of the confined mode yields

$$I(\mathbf{k}',\mathbf{k}'') = iq \int d^3\mathbf{x} \, e^{-i\mathbf{q} \cdot \mathbf{x}} \Phi, \qquad (13)$$

where $q = |\mathbf{q}| \equiv |\mathbf{k}' + \mathbf{k}''|$. The specific function form of Φ due to the spherical confinement

$$\Phi(\mathbf{x}) = f_l(r) Y_{lm}(\theta,\phi), \qquad (14)$$

where Y_{lm} is the spherical harmonics and $f_l(r)$ is the radial part of the vibration potential, enables us to simplify further the integral of Eq. (13). We expand the plane wave as

$$e^{-i\mathbf{q} \cdot \mathbf{x}} = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^{l} (-i)^l j_l(qr) Y_{l,m}(\theta_q,\phi_q) Y_{lm}^*(\theta,\phi), \qquad (15)$$

where (θ_q,ϕ_q) are the direction angles of $\mathbf{q}$. Inserting Eqs. (14) and (15) into Eq. (13) and utilizing the property of the spherical harmonics, we obtain

$$I(k',k'') = 4\pi(-i)^{l-1} q Y_{lm}(\theta_q,\phi_q) \int dr \, r^2 j_l(qr) f_l(r). \qquad (16)$$

To carry out the integral of Eq. (11), we need the expressions of q and θ_q in terms of $\mathbf{k}'$ and $\mathbf{k}''$. Let (θ',ϕ') and (θ'',ϕ'') be the direction angles of $\mathbf{k}'$ and $\mathbf{k}''$. It is easy to show that

$$q = [k'^2 + k''^2 + 2k'k'' \cos \Gamma]^{1/2}, \qquad (17)$$

where Γ is the angle between $\mathbf{k}'$ and $\mathbf{k}''$, which can be obtained from $\cos \Gamma = \cos \theta' \cos \theta'' + \sin \theta' \sin \theta'' \cos(\phi' - \phi'')$. The angle θ_q between $\mathbf{q}$ and $\mathbf{e}_z$ is given simply by

$$\cos \theta_q = (k' \cos\theta' + k'' \theta'')/q. \qquad (18)$$

Combining Eqs. (16)–(18), from Eq. (11) we can calculate straightforwardly the lifetime for arbitrarily confined optical phonons in spherical quantum dots.

Considering the special case for modes with zero angular momentum, $Y_{00}(\theta_q,\phi_q) = 1/\sqrt{4\pi}$ is a $\mathbf{k}'$- and $\mathbf{k}''$-independent constant, which implies the integral $\int d\Omega''$ in Eq. (11) being independent of the direction of $\mathbf{k}'$. Accordingly, for the $l=0$ modes, the lifetime formula has a more simplified form

$$\frac{1}{\tau} = \left[\frac{4}{3\pi^2} \left(\frac{M}{M_2} \right)^2 \frac{\hbar\omega}{\widetilde{M} v^2} \frac{\gamma^2}{v} \right] \int_0^{k_D} dk' k'^2$$

$$\times \int_0^{\pi} d\Gamma \, \sin\Gamma k''^2 \omega' \omega'' q^2 I_0^2(q)(N' + N'' + 1), \qquad (19)$$

where $I_0(q) = \int dr \, r^2 j_0(qr) f_0(r)$, $k_D = \omega_D/v$, and ω_D is the Debye frequency.

III. APPLICATION

Below we apply the above formalism to GaAs quantum dots. In the numerical calculation we adopt the GaAs parameters as the sound velocity $v = 5.22 \times 10^5$ cm/s, the zone-center LO-phonon energy $\hbar\omega_L = 36$ meV, and $M \simeq M_1 = \rho b^3$, where $\rho = 5.32$ g/cm^3 is the material density and $b = 5.65$ Å is the lattice constant. To compare with the avail-
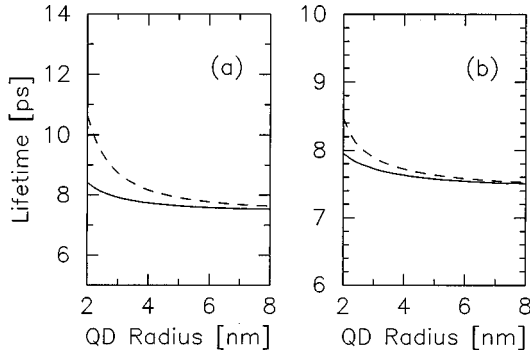
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FIG. 1. Size dependence of the lifetime at zero temperature: (a) the dispersion relation (21) is used; (b) the dispersionless phonon energy $\hbar\omega \equiv \hbar\omega_L = 36$ meV is applied to set a lower limit for the lifetime. The solid (dashed) curve is for the $n=1$ ($n=2$) mode.

able measured result in bulk GaAs, we approximately take $M_2 \simeq M_1$ and estimate the Debye wave number k_D from $(4\pi/3)k_D^3/(2\pi)^3 = 1/b^3$, which gives $k_D = [2\pi(3/4\pi)^{1/3}]/b$. We take $\gamma = 2$ as a typical value of the Grüneisen constant.[8,9] As a specific example we consider the mode with zero angular momentum $l=0$. In Ref. 19 we have shown that the $l=0$ mode is purely LO vibrational, being different from the $l \neq 0$ mode, which mixes an interface polariton component into the bulklike LO vibration. Further, as a good approximation to the realistic confinement, we assume a rigid boundary condition, which results in a simple function form for the $l=0$ mode as

$$\Phi \sim f_{0,n}(r) = \frac{\sin(k_n r)}{k_n r}, \qquad (20)$$

where $k_n = n\pi/a$ and $n=1,2,3,\dots$. Correspondingly, the phonon energy is given by

$$\hbar\omega_n = \sqrt{(\hbar\omega_L)^2 - (\hbar k_n v)^2}, \qquad (21)$$

according to the dispersive model for LO vibrations.

Figure 1 shows the size dependence of the lifetime at zero temperature, where the solid and dashed lines represent the results of the $n=1$ and 2 modes, respectively. The slightly longer lifetime of the $n=2$ mode is due to its weaker coupling to the two bulk LA phonons, which is manifested mathematically in the overlap integral in Eq. (19), i.e., $|I_{0,1}(q)|^2 > |I_{0,2}(q)|^2$. In Fig. 1(a) the dispersion relation (21) is used. This relation is reasonable at the long-wavelength limit. For ultrasmall quantum dots, which implies the breakdown of the long-wavelength approximation, we know from Eq. (19) that the use of this dispersion relation would overestimate the lifetime since the realistic dispersion of LO phonons is weaker than this relation. To set a lower limit for the lifetime within the present formalism, we use alternatively a dispersionless LO-phonon energy of $\hbar\omega = \hbar\omega_L = 36$ meV in Eq. (19). The result is shown in Fig. 1(b). We see that except for the region $a < 4$ nm, the results in Figs. 1(a) and 1(b) are almost the same. At this stage, we arrive at the conclusion that the lifetime of the confined mode is weakly size dependent in the range of ultrasmall sizes and it eventually approaches the bulk value after $a > 5$ nm. The size-dependent behavior of the lifetime in Fig. 1 can be understood from the general overlap integral in Eq. (11) as
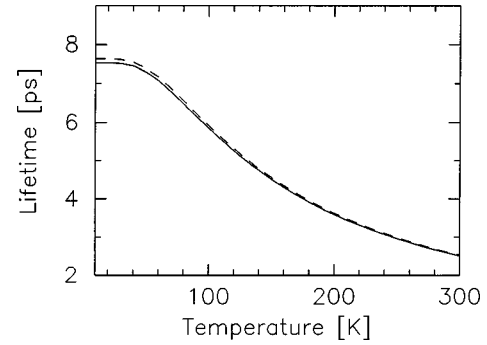

FIG. 2. Temperature dependence of the lifetime for a dot with radius $a = 8$ nm. The curves are the same as in Fig. 1.

follows. Since $I(\mathbf{q})$ is in fact the Fourier component of the confined phonon potential, for a more localized mode, the Fourier $\mathbf{q}$ spectrum will extend to a wider range. On the other hand, the final momentum summation of the two LA phonons is physically restricted to a finite range. Thus a longer lifetime is expected for a more localized mode. This simple argument seems to be generally valid in the three-phonon interaction model, which leads us to the following two conjectures: (i) The lifetime of confined phonons in other microstructures may also be weakly size dependent, just like the behavior of the confined modes in quantum dots, and (ii) for the $l \neq 0$ mode in quantum dots, despite its more complex functional form due to the mixture of an IP component, a size dependence similar to that of the $l=0$ mode is expected. The detailed study of the $l \neq 0$ mode is left to our future work.

In Fig. 2, for a given size of dot with radius $a = 8$ nm, we show the temperature dependence of the lifetime of the confined mode with zero angular momentum, where the curves are the same as in Fig. 1. We see that the results in Fig. 2 are in excellent agreement with the measured data (7 ps at 77 K and 3.5 ps at 300 K) in bulk GaAs, especially at low temperatures. At room temperature, our calculated lifetime (2.5 ps) is slightly shorter than the measured value (3.5 ps), which can be understood from the neglect of the temperature dependence of the three-phonon coupling constant, say, the Grüneisen constant. Note that in Ref. 4, where a more complex microscopic treatment was presented for bulk GaAs, better agreement with the measured data is obtained at room temperature, but poorer agreement at low temperatures, which is attributed to the overall use of the third-order elastic constant estimated at 300 K. Therefore, if a temperature-dependent coupling constant were available, our results would be completely consistent with that in Ref. 4 and with the experimental result.

IV. CARRIER ENERGY RELAXATION

In this section we address the impact of the finite lifetime of phonons on the phonon bottleneck effect, in a slightly different way from the direct numerical calculation in Ref. 18, which is expected to be instructive in the understanding of this issue more physically and intuitively. For simplicity, consider the electron in a quantum dot coupling to a single phonon mode and denote the electron ground and excited states by $|g\rangle$ and $|e\rangle$, respectively. As we consider the elec-
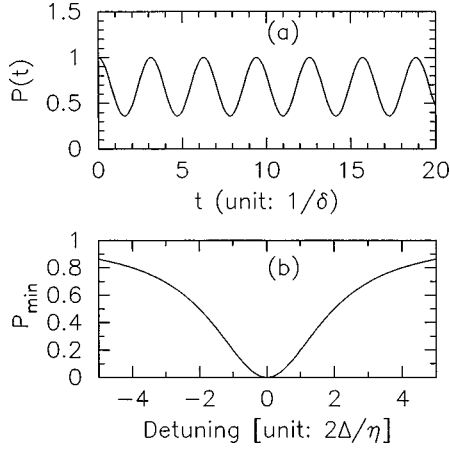
FIG. 3. (a) Rabi oscillation for a two-level system with no dissipation, where $P(t)$ is the probability of finding the initial state; (b) the dependence of the minimum of $P(t)$ on the detuning of the two levels.

tron transition from state $|e\rangle$ to $|g\rangle$ by emitting an optical phonon, the two relevant states are $|e,0\rangle$ and $|g,1\rangle$, where 0 and 1 mean no phonon and one phonon states. This is in fact a two-level description.

First we neglect the dissipation of the phonon mode. In the absence of electron-phonon coupling, the energy levels of the two product states are $E_1 = E_e \equiv E_0 + \Delta$ and $E_2 = E_g + \hbar\omega_L \equiv E_0 - \Delta$, where E_e and E_g are the energy levels of electron states $|e\rangle$ and $|g\rangle$, and 2Δ is the detuning of the two states. In the presence of electron-phonon coupling, the two energy levels separate further as

$$E_\pm = E_0 \pm \sqrt{\eta^2 + \Delta^2} \equiv E_0 \pm \delta, \qquad (22)$$

where η is the electron-phonon coupling strength. Assuming that the initial state is $|e,0\rangle$, a simple derivation gives for the probability of the electron to stay on the initial state $|e,0\rangle$,

$$P(t) = 1 + \frac{1 - (\Delta/\delta)^2}{2} [\cos(2\delta t) - 1]. \qquad (23)$$

In Fig. 3(a) we schematically show this oscillating behavior, which is known as the Rabi oscillation. Note that if the detuning 2Δ is zero, the minimum of $P(t)$ can reach zero; otherwise $P(t)$ oscillates between 1 and nonzero minimum $P_{min} = (\Delta/\delta)^2$, which depends on both the electron-phonon coupling strength η and the detuning 2Δ. In Fig. 3(b) we show the dependence of the minimum of $P(t)$ on the detuning.

Now we take into account the dissipation of the phonon mode, i.e., its decay to bulk acoustic phonons. In the presence of dissipation, $P(t)$ is a decaying oscillating function. Approximately, the maximum value of $P(t)$ will reduce a factor P_{min} after each period of the phonon lifetime τ. Consequently, if the detuning is zero, the carrier lifetime τ_c is equal to the phonon lifetime τ. For the nonresonant case, we can reasonably regard the carrier energy relaxation to be ful-

filled if the maximum of $P(t)$ is smaller than $1/e$. Thus, for the detuning that satisfies the condition $P_{min} < 1/e$, the carrier relaxation time is also about the phonon lifetime. For the strong detuning with $P_{min} > 1/e$, the carrier relaxation time can be estimated as $\tau_c = N\tau$, where N is obtained from $(P_{min})^N \sim 1/e$, which gives, for the carrier relaxation time,

$$\tau_c \sim \frac{\tau}{\ln(1 + \eta^2/\Delta^2)}. \qquad (24)$$

For GaAs quantum dots, based on the Fröhlich interaction model, we estimate the coupling strength between the electron and the *most-efficient* single phonon mode to be approximately $\eta \sim 0.3\hbar\omega_0/\sqrt{a}$, where a is the dot size in nanometers. For a typical dot with $a = 10$–25 nm, the coupling strength η is about 2–3.5 meV. From Fig. 3 and the analysis giving rise to Eq. (24), the detuning can be as large as about 8η, which corresponds to an energy range of tens of meV; in this detuning range the carrier relaxation time is only several times the phonon lifetime, which is fast enough with respect to the phonon bottleneck problem, since the phonon lifetime is only a few picoseconds. Although the above discussion is restricted to a two-level analysis, the qualitative result will remain the same when one employs a full treatment to the coupling to multimodes. For more details, see Ref. 18.

V. CONCLUSION

In this paper, on the basis of a simple phenomenological approach treating the phonon decay into two acoustic phonons, we have derived an expression for the lifetime of the confined optical phonons in spherical quantum dots. For GaAs dots, we have shown the size and temperature dependences of the lifetime by taking the zero-angular-momentum mode as an illustrative example. The obtained results are in excellent agreement with other theoretical and experimental results in bulk GaAs systems. Considering the wide lack of knowledge of the confined optical phonons in semiconductor quantum dots, we feel that the present study is of particular interest. It will be helpful to understand the optical spectrum linewidths associated with the emission and absorption of optical phonons in a large number of experiments. Furthermore, it leads us to the important conclusion that due to the decay of the confined optical phonons, the carrier relaxation time can be as short as tens of picoseconds in a wide energy detuning of tens of meV. Thus the so-called phonon bottleneck effect is not a serious problem for the energy relaxation in quantum dots.

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