Optical linewidths in an individual quantum dot

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On the basis of lattice relaxation due to the electron acoustic phonon coupling, we theoretically analyze the homogeneous optical linewidths of a single quantum dot. The calculated result based on a nonstrong-coupling approximation shows that in a considerable range of temperatures the homogeneous linewidth can be expressed in terms of $\Gamma_h = \Gamma_0 + AT$, and the size dependence is characterized by $\Gamma_0 \propto 1/R$ and $A \propto 1/R^2$, where T is the temperature and R is the quantum dot radius. While this result qualitatively agrees well with the available experiments on exciton dephasing, it straightforwardly implies the infrared optical properties due to intraband transitions with transition energy around 100 meV. [S0163-1829(99)02627-2]

I. INTRODUCTION

Semiconductor quantum dots (QD's) promise important advantages in device applications such as higher performance for lasers,[1,2] strongly enhanced oscillator strengths,[3] optical nonlinearities,[4] etc. All these optical properties are related to the three-dimensional confinement that gives rise to a sharply discrete electron energy spectrum, thus understanding of optical spectrum broadening is an essential issue. Without knowledge of the broadening, for example, we can make no useful estimate for the size of the nonlinear absorption, since it depends on the width of the linear absorption line.[4]

In practice the broadening is dominated by inhomogeneities in the size and shape of QD's. However, the intrinsic mechanisms, predominantly the phonon broadening, would set the limit to the properties of QD's that cannot be eliminated by better fabrication. This stimulated considerable interest in investigations on the homogeneous spectral linewidths with certain indirect methods on large inhomogeneous semiconductor microcrystal ensembles.[5–13] More recently, local spectroscopic techniques, such as near-field optical microscopy, make it possible to probe an individual quantum dot,[14–16] and therefore measure directly the homogeneous linewidths.[17] The essential experimental evidence can be summarized as follows:[13] (i) the linewidth has commonly linear temperature behavior in the relatively high-temperature regime, and (ii) the linear coefficient is roughly inverse-square-size dependent.

The aim of this work is twofold. First, we intend to clarify the general features of the homogeneous linewidths based on the lattice relaxation (LR) mechanism by considering the coupling of electron to the acoustic phonons during the optical transitions (for more details in the motivation on this point, see the analysis of the next paragraph). In this respect, the intraband and interband transitions are methodologically equivalent (except for certain differences in estimating the electron phonon coupling strength). This fact ensures that if we restrict the calculation to the intraband optical transitions, the obtained result can be qualitatively compared with the available experiments on interband transitions. Second, we wish to get some insight into the new subject, say, the QD infrared intraband optical properties. Very recently, stimulated by the possibility to realize the infrared light devices (e.g., infrared light emitters and infrared photodetectors), and the optical nonlinearities associated with the QD intraband transitions, there has been growing interest in the optical studies based on the intraband transitions.[25] In this context, the homogeneous linewidth is an important issue as discussed in Ref. 4 for the case of interband optical transitions. We hope that in the near future this intraband homogeneous linewidth in a single quantum dot can be probed with local spectroscopic techniques in addition to the progress of growing high quality QD samples.

As is well known, in semiconductors the electron phonon scattering is the dominant contribution to line broadening. In this mechanism, the experimental homogeneous linewidths in QD's were fitted from the phonon-scattering rate in bulk semiconductors,[8] or phenomenologically.[9,10,17] Note that, in higher dimensional systems such as in quantum wells, the lifetime broadening mechanism can account for the homogeneous linewidths, in which the electron state under consideration is scattered rapidly into other energy states by phonons, thus the finite lifetime results in a level broadening. For example, in Ref. 17, this mechanism was employed to explain the experimental result in a large size QD, where the level spacing is of several meV, which implies an efficient electron LA phonon scattering. However, this lifetime broadening mechanism breaks down in very small QD because of the absence of the exact detuning with the optical phonons,[18] and the extremely reduced scattering rate due to the acoustic phonons.[19] In this case, LR is likely the only and most reasonable mechanism to account for the homogeneous broadening in single dot optical transitions.[20–22] The main motivation of this work is aimed to provide an appropriate treatment to the homogeneous linewidth based on the LR in the present not strongly coupled electron-lattice system. More specifically, due to the localized nature of the electron states in QD, upon the optical transition, the lattice configuration changes according to the different electronic states, and this lattice relaxation effect causes the broadening of the optical lines. For the coupling of electron to a single phonon mode, or more generally, single frequency multiphonon modes, exact analytical solution exists; but for the coupling

to arbitrary multiphonon modes, it is impossible to derive an exact analytical solution. In the latter case, the strong-coupling approximation (SCA) is a popular and widely accepted approximation,[20,23,24] which worked well in most strongly coupled electron-lattice systems. However, since the coupling strength of the QD electron-phonon system is weak in regard to the LR criterion for quantum dot with size larger than, say, 4 nm, the SCA is unsuitable to be employed to calculate the optical linewidths. We shall show below that the SCA would give rise to a temperature dependence $\Gamma_h \propto \sqrt{T}$, where Γ_h is the homogeneous linewidth and T the temperature, and the size dependence $\Gamma_h \propto 1/R^{1.5}$, where R is the radius of a spherical QD. We attribute this deviation from the central experimental features to the breakdown of the strong coupling approximation. In this work, we shall further develop an effective single-phonon-mode approach to calculate Γ_h beyond the SCA. The calculated result implies that in a considerable range of temperatures the homogeneous linewidth can be expressed in terms of $\Gamma_h = \Gamma_0 + AT$, and the size dependence is characterized by $\Gamma_0 \propto 1/R$ and $A \propto 1/R^2$. The qualitative agreement of our result with the available experiments is good, but it highly suggests both a more careful analysis of the experimental data of exciton dephasing, and performing new experiments on the QD intraband optical transitions.

II. ELECTRON AND PHONON STATES

As mentioned above, we are going to focus our attention mainly to the intraband optical transitions in QD. As a result of confinement, the electronic states are localized. For simplicity, we assume a spherical QD and a rigidly confining potential. Under this consideration, the (conduction) electron wave function is

$$\psi_{lm}(k,\mathbf{r}) = B_l^e j_l(kr) Y_{lm}(\theta,\phi), \tag{1}$$

where j_l and Y_{lm} are, respectively, the spherical Bessel function and the spherical harmonics, and B_l^e is a normalization constant with a form as $B_l^e = [(R^3/2) j_{l+1}^2(kR)]^{-1/2}$, where R is the QD radius. The order of the electronic states counting from the lowest energy level to the higher ones is $1s, 1p, 1d, 2s, 1f, \ldots$, where *s, p, d,* and *f* denote the angular momentum as in atomic physics. In this work, we shall consider the optical transition between the two lowest states, namely, the ground $1s$ state $\psi_{00}(k_1,\mathbf{r})$, and the first excited $1p$ state $\psi_{10}(k_2,\mathbf{r})$. Note that due to the selection rule based on the dipole approximation for the optical transitions, only the $1p$ state with magnetic quantum number $m=0$ is involved in the transition between $1s$ and $1p$. Further, we can determine the wave number $k_1 = \pi/R$, and $k_2 = 4.4934/R$, from the rigid boundary condition $j_l(kR) = 0$.

To estimate the broadening of the optical transitions, we need to specify the acoustic-phonon modes in QD. It is clear that the optical phonons are confined in QD,[26] due to the obvious discontinuity of band-edge optical-phonon frequencies of the inside and outside materials. For the acoustic phonons in QD, the size quantized acoustic-phonon modes were observed by the low-frequency Raman scattering.[27] Theoretically, these acoustic modes can be described in terms of the elastic vibration of homogeneous particle with pressure free boundary condition; and the outside material plays the role of coupling anharmonically to these modes and dissipating the energy, consequently resulting in a finite lifetime on these localized phonon modes.[28,29] Further, the dominant contribution to the electron-phonon interaction is from the longitudinal acoustic (LA) phonons through the deformation potential coupling. The mechanical potential associated with the LA vibration can be derived as

$$\Phi_{lm}(q,\mathbf{r}) = B_l^p j_l(qr) Y_{lm}(\theta,\phi) \equiv B_l^p G_{q,lm}(\mathbf{r}). \tag{2}$$

The associated LA vibrating displacement is given by $\mathbf{u}_m = \nabla \Phi_{tm}$. With the help of this relation, we can specify the normalization constant for a single LA mode as

$$B_l^p = \left[\frac{\rho R^3}{2} q^2 j_{l+1}^2(qR) \right]^{-1/2}. \tag{3}$$

Using these LA modes as the representation basis, the electron LA-phonon interaction Hamiltonian that resulted from the deformation-potential model can be written as

$$H_{ep} = \sum_{q,lm} V_{q,l} G_{q,lm}(\mathbf{r}) Q_{q,lm} \equiv \sum_{q,lm} u_{lm}(q,\mathbf{r}) Q_{q,lm}, \tag{4}$$

where $Q_{q,lm}$ is the canonical coordinate of the normal LA mode, the coupling strength has the form $V_{q,l} = D\omega_q / \sqrt{(\rho R^3/2) q^2 j_{l+1}^2(qR)}$, D is the deformation potential constant, and $\omega_q = qv$ is the LA phonon frequency with wave number q and sound velocity v.

III. LR APPROACH: STRONG-COUPLING APPROXIMATION

For the coupled electron-lattice system, in the Born-Oppenheimer approximation the state can be decomposed as $\Psi_i(\mathbf{r},\{Q_{q,lm}\}) = \psi_i(\mathbf{r},\{Q_{q,lm}\}) \chi_i(\{Q_{q,lm}\})$, where ψ_i and χ_i are the electron and lattice wave function respectively. In the electron wave function, $Q_{q,lm}$ only plays the role of parameter. The lattice wave function χ_i is a direct product of states of harmonic oscillators whose oscillating equilibrium origins are influenced by the electronic state.[20–24]

Consider the optical transition between the two lowest conduction electron states in QD. The transition probability is determined by the following *spectral shape* function:

$$W(E) = A v_i \sum_f |\langle f|M|i\rangle|^2 \delta[E - \Delta E_{fi}]. \tag{5}$$

Here M is the electric dipole moment of the electron, E is the photon energy, and ΔE_{fi} is the energy difference of the whole electron-lattice system before and after the optical transition. The average over the initial phonon states and summation over the final phonon states have been explicitly shown in the equation.

In the Condon approximation, the electronic matrix $M_{fi} = \langle \psi_f | M | \psi_i \rangle$ can be regarded as lattice coordinate independent. Following the standard procedure of LR to carry out the average (summation) over initial (final) phonon states, we have

$$W(E) = |M_{fi}|^2 \int_{-\infty}^{\infty} d\mu e^{F(\mu,E)}, \tag{6}$$

with

$$F(\mu,E)=-i\mu(E-\Delta E_{fi})+\sum_{lmq}\left(\frac{\omega_q}{2\hbar}\right)\Delta_{filmq}^2 \times\left[\coth\frac{\beta\hbar\omega_q}{2}(\cos\mu\hbar\omega_q-1)+i\sin\mu\hbar\omega_q\right], \quad (7)$$

where β is the inverse temperature, and $\Delta_{filmq}=\Delta_{flmq}-\Delta_{ilmq}$ with

$$\Delta_{jlmq}=\langle\psi_j|u_{lm}(q,\mathbf{r})|\psi_j\rangle/\omega_q^2, \quad (8)$$

describing the shift of the lattice normal oscillator origin before and after the electron transition.

For the case of single phonon mode or multimodes with single frequency, the integral of Eq. (6) can be carried out exactly. Otherwise, this integral is analytically intractable for the arbitrary multimodes. In such a case, the strong-coupling approximation is a popular and widely accepted approximation, i.e., by expanding the function $F(\mu,E)$ to the second order of μ, and using the steepest descent method, Eq. (6) yields

$$W(E)=|M_{fi}|^2\left[\frac{2\pi}{\overline{S_T(\hbar\omega_q)_T^2}}\right]^{1/2}\exp\left[-\frac{(E-\Delta E_{fi}-\overline{S\hbar\omega_q})^2}{2\overline{S_T(\hbar\omega_q)_T^2}}\right], \quad (9)$$

where

$$\overline{S\hbar\omega_q}=\sum_{lmq}\left(\frac{\omega_q}{2\hbar}\right)\Delta_{filmq}^2\hbar\omega_q, \quad (10)$$

$$\overline{S_T(\hbar\omega_q)_T^2}=\sum_{lmq}\left(\frac{\omega_q}{2\hbar}\right)\Delta_{filmq}^2(\hbar\omega_q)^2\coth\left(\frac{\beta\hbar\omega_q}{2}\right). \quad (11)$$

From the spectral shape function Eq. (9), it is straightforward to identify the full width at half maximum (FWHM) as

$$\Gamma_h=[(8\ln 2)\overline{S_T(\hbar\omega_q)_T^2}]^{1/2}. \quad (12)$$

To carry out these LR quantities, we need to specify the oscillator displacement parameter Δ_{jlmq}. In the dipole approximation for the optical transition between $1s$ and $1p$ states, we know from group theory that only the $1p$ state with zero magnetic quantum number is involved. Thus, we have

$$\Delta_{jl0q}=\frac{1}{\omega_q^2}\langle Y_{j0}|Y_{l0}|Y_{j0}\rangle\int_0^R drr^2|j_j(kr)|^2j_l(qr). \quad (13)$$

For convenience, we denote the angular matrix element by $Z_{jlj}\equiv\langle Y_{j0}|Y_{l0}|Y_{j0}\rangle$. From the angular momentum coupling theory, we know that the nonzero matrices are $Z_{000}=1/\sqrt{4\pi}$, $Z_{101}=1/\sqrt{4\pi}$, and $Z_{121}=-1/\sqrt{5\pi}$. Based on this analysis, we see that only the phonon modes with angular quantum number $l=0$ and 2 respond to the electron-lattice coupling in terms of lattice relaxation.

Figure 1(a) shows the homogeneous linewidth Γ_h under the strong-coupling approximation, where the adopted parameters are from the CdSe quantum dot (in the following numerical calculations the same material parameters will be used). The solid line is directly calculated from Eq. (12), and the dashed line is from a further approximation, say, the high-temperature approximation, which gives rise to

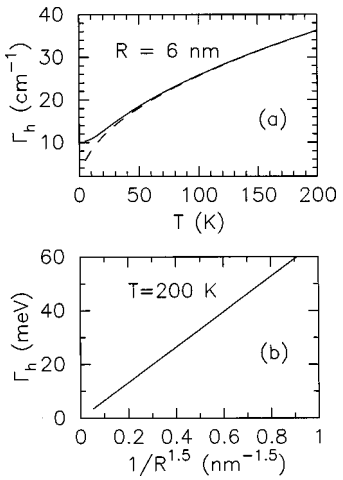

FIG. 1. Homogeneous linewidth Γ_h under the strong-coupling approximation shown in (a) for the temperature dependence ($\Gamma_h\propto\sqrt{T}$), and in (b) for the size dependence ($\Gamma_h\propto 1/R^{1.5}$). The dashed curve in (a) is from the high-temperature approximation.

$$\Gamma_h^2=(16\ln 2)kT\overline{S\hbar\omega_q}. \quad (14)$$

In Fig. 1(a) we see that in a considerably wide range of temperatures this high-temperature approximation works very well. Consequently, Eq. (14) implies that the temperature dependence of the linewidth is $\Gamma_h\propto\sqrt{T}$ rather than $\Gamma_h\propto T$ as indicated by experiments[5,8,13,14] in the high-temperature regime due to acoustic-phonon scattering. From Eqs. (14) and (10) we can further know that for a given temperature the homogeneous linewidth depends on the QD size approximately as $\Gamma_h\propto 1/R^{1.5}$ from a simple scaling analysis by noting the insensitive feature of the overlap integral to the QD size. This conclusion is demonstrated by the numerical calculation shown in Fig. 1(b). Note that this feature differs qualitatively from the experimental evidence that indicated an inverse-square-size dependence behavior (i.e., $\Gamma_h\propto 1/R^2$).

We attribute the above discrepancies to the application of the strong-coupling approximation. As a matter of fact, in respect to the lattice relaxation criterion, the electron-lattice coupling strength is quite weak if the dot size is larger than several nanometers (e.g., 4 nm).[23] Below we present an alternative treatment beyond the strong-coupling approximation, which can provide a better description for the homogeneous linewidths.

IV. LR APPROACH: EFFECTIVE SINGLE MODE APPROXIMATION

In Ref. 5, by phenomenologically approximating the quasicontinuum of acoustic phonons by a single mode, the temperature dependence was discussed on the basis of a shifted oscillator model. In what follows, from a direct multimode calculation for the LR quantities, we first define an effective single phonon mode, then present an exact analysis for the linewidth based on the single phonon mode. The basic idea is that we let the effective single mode have the same lattice

relaxation feature as the multimodes, thus we have a definition for the single phonon mode as

$$\hbar\Omega = \overline{S\hbar\omega_q}/S, \tag{15}$$

where

$$S = \sum_{lmq} \left(\frac{\omega_q}{2\hbar}\right) \Delta_{filmq}^2 \tag{16}$$

is the Huang-Rhys parameter. In this way, the single mode description keeps the same Huang-Rhys parameter and lattice relaxation energy (i.e., $\overline{S\hbar\omega_q}$) as the multimodes do. Replacing $\hbar\omega_q$ in Eq. (7) by the just obtained average phonon energy $\hbar\Omega$, a precise expression follows directly as

$$W(E) = |M_{fi}|^2 e^{-(1+2N)S} \sum_{p=-\infty}^{\infty} I_p[2S\sqrt{N(N+1)}] \times \left(\frac{1+N}{N}\right)^{p/2} \delta(E - \Delta\epsilon_{fi} - p\hbar\Omega), \tag{17}$$

where $N = [\exp(\hbar\Omega/k_BT) - 1]^{-1}$ is the Bose function, $I_p(x)$ is the imaginary argument Bessel function, and $\Delta\epsilon_{fi}$ is the electron level spacing. Equation (17) gives rise to a number of discrete phonon peaks by the δ function. Due to the small energy of the low-frequency acoustic phonon and its rapid decay to the bulk acoustic phonons (i.e., the heat bath), conventionally we can regard the envelope of the discrete phonon peaks given by Eq. (17) as the practical observed spectral shape function of the optical transition. Therefore, we make a continuity of p from the discrete integer to a continuous variable $p = \Delta E/\hbar\Omega$, where ΔE is the optical detuning energy $\Delta E = E - \Delta\epsilon_{fi}$. Accordingly, the homogeneous linewidth is obtained from the following envelope function:

$$f(\Delta E) = I_p[2S\sqrt{N(N+1)}] \left(\frac{1+N}{N}\right)^{p/2} \Bigg|_{p=\Delta E/\hbar\Omega}. \tag{18}$$

Based on this line-shape function, we can determine the FWHM.

We noted that, in the context of bulk semiconductor lasers and based on the carrier-LO phonon and carrier-carrier scatterings, Ref. 30 addressed the non-Markovian relaxation processes and obtained a non-Lorentzian line-shape function, which has an asymmetry and stronger convergent characteristics in comparison with the Lorentzian function. In our case, despite the different microscopic mechanism, Eq. (18) shows a similar asymmetry non-Lorentzian line-shape feature. More specifically, at zero temperature the envelope function Eq. (18) corresponds to a Poissonian distribution, while it evolves gradually to a Gaussian function with increase of temperature. The underlying physics for this non-Lorentzian behavior is owing to the fact that the present LR treatment for the coupled QD electron-lattice system goes beyond the conventional Markovian approximation by involving a certain non-Markovian relaxation effect on the acoustic-phonon bath. For example, in LR the effect of the electron subsystem on the lattice subsystem would result in an origin shift of the bath oscillators (i.e., the acoustic phonons). This lattice relaxation effect is essential in understanding the present homogeneous broadening, but it would be ignored completely in the Markovian approximation.

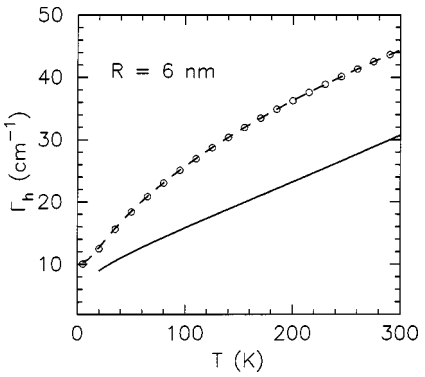

FIG. 2. Linear temperature dependence (solid line) calculated exactly from the effective single mode model. Self-consistency between the effective single mode and the multimodes is examined under the strong-coupling approximation by the dashed curve and the small circles, which, having a $\sqrt{T}$ behavior, also show an obvious deviation from the linear temperature dependence.

Figure 2 shows the calculated linewidth on the basis of Eq. (18) as a function of temperature by the solid line, where, as a reference, we also plot the result of the strong-coupling approximation by the dashed curve (from multiphonon modes), and the small circles (from the effective single phonon mode), which in a certain sense demonstrate the self-consistency between the descriptions of the single mode and multimodes. Resulting from the exact solution from the effective single mode, the solid line in Fig. 2 clearly shows a linear temperature-dependent behavior of the linewidth in a wide range of temperatures, which can be characterized by

$$\Gamma_h = \Gamma_0 + AT. \tag{19}$$

This T linear behavior is in good agreement with experiments. For example, in Ref. 12, the origin of this behavior is attributed to the significantly enhanced coupling to the low-frequency acoustic phonons, since the strong linear temperature dependence found there cannot be accounted for by the increasing LO phonon population.

It is well known that in higher dimensional systems the homogeneous linewidth due to the electron LA phonon scattering is proportional to T at high-temperature limit. Interestingly, we noted that in Ref. 17 the optical spectra resulting from a relatively large QD were investigated in high precision, where at respectively high temperatures (i.e., between 25 and 50 K) the linewidth also has linear temperature dependence. There, since the QD size is not too small, then the exciton level spacing is in the range of efficient LA phonon scattering, a simple lifetime broadening mechanism can account for both the extremely narrow linewidth of the order of 0.025–0.12 meV, and the temperature dependence. Here we stress that in very small QD, where the energy level spacing is considerably large thus the lifetime broadening mechanism breaks down completely, the low-frequency acoustic phonons can as well result in a linear-T linewidth in wide range of temperatures, due to the lattice relaxation effect in optical transition.

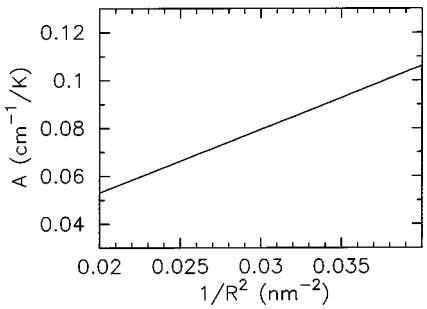

FIG. 3. Inverse-square-size dependence of the linear coefficient *A*.

The size dependence of Γ_h is characterized by A and Γ_0 in Eq. (19). Figure 3 shows the numerical result of A based on the effective-single-mode LR approach. This precise inverse-square-size dependence is in excellent agreement with the experimental fitting in Ref. 8. Other experiments also roughly support this behavior by showing that $\Gamma_h \propto R^{-2}$. However, strictly speaking, our calculation revealed a slight deviation of Γ_h from the R^{-2} behavior. In Fig. 4(a) we plot the size behavior of Γ_h for two given temperatures, from which we deduce the size dependence of Γ_0 in Fig. 4(b). Very interestingly, we see that Γ_0 is characterized well by a linear inverse size dependence, i.e., $\Gamma_0 \propto R^{-1}$. To our knowledge, this fact has not been reported in any experiments. On the contrary, in literature, the Γ_0 part in Eq. (19) was widely regarded as a contribution of nonphonon scattering, and having no size dependence. We here recognize that Γ_0 (at least part of it) share the common origin of the term AT due to the LA phonon scattering, and has clear size dependent behavior.

V. CONCLUSION

In summary, we theoretically investigated the homogeneous optical linewidth in QD by showing its temperature and size dependences. Our calculation was restricted to the conduction intraband optical transitions in the intermediate size of QD, i.e., it has energy level spacing around 100 meV, which implies that the usual lifetime broadening mechanism due to phonon scattering breaks down completely, thus the lattice relaxation in the optical transitions is likely the only and most reasonable way to account for the homogeneous linewidth, due to the significant size localization of electron states in QD. Due to the common origin of LR in small QD, we can compare the qualitative features between the present obtained results and the exciton dephasing behaviors in the available experiments, and satisfactory agreement was obtained. The present study may suggest both further careful analysis of the experimental data in exciton dephasing, and performing new experiments on the QD intraband optical transitions. Especially, in view of the sound potential being applied to infrared optical devices (e.g., infrared detectors and emitters), the study of intraband optical transitions in QD's itself is of great interest.

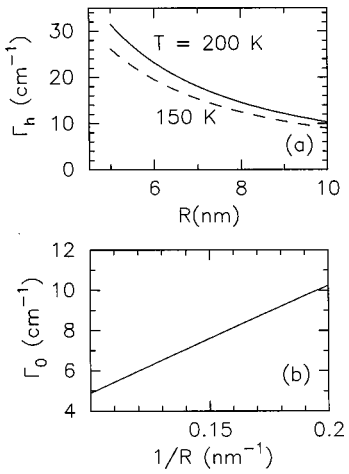

FIG. 4. (a) Size dependence of the homogeneous linewidth Γ_h and (b) the inverse size dependent behavior of the temperature independent part in Γ_h.

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