Size dependence of polaronic effects on an exciton in a spherical quantum dot

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(Received 3 December 1998)

The size dependence of polaronic effects (i.e., LO-phonon effects) on an exciton in a spherical quantum dot is studied by using a variational method. Exciton energy, exciton-phonon interaction energy, and virtual phonon number are calculated by taking into account the interaction between an exciton and both bulk-type and interface-type LO phonons in the system. The numerical results for GaAs and CdSe quantum dots clearly show that (i) in the limit of the small dot, polaron effects on an exciton vanish, i.e., an exciton becomes the bare exciton receiving no LO-phonon effect and (ii) with the increase in the dot radius, the polaronic effects increase and then gradually approach the bulk values. The decrease of the polaronic effects for the small dot is due to the cancellation of the polaronic effects owing to the opposite charge of an electron and a hole. [S0163-1829(99)04715-3]

I. INTRODUCTION

In recent years quantum dot (QD) systems have attracted much attention because of the physical phenomena and their potential applications to devices. In these systems electronic states receive a strong confinement effect. Also LO-phonon states have very different features from those in the bulk, such as the confinement of the bulk-type phonons and the existence of the interface-type phonons.[1–3] Effects of LO phonons on an exciton play an important role in optical properties of QD and have been investigated experimentally and theoretically.[4–12]

In these studies the size dependence of the exciton–LO-phonon interaction effect in a QD system is one of the main issues and has been studied by many authors.[4–7,9,10,12] However, the problem has not been understood well so far. Schmitt-Rink, Miller, and Chemla pointed out that the exciton–LO-phonon coupling mediated by the Fröhlich interaction should vanish in small spherical nanocrystals.[4] Klein *et al.* discussed the problem, using (i) a donorlike exciton model in which a hole is treated as a point charge at the center of a QD and (ii) the adiabatic approximation to treat the exciton-phonon interaction. They found that the strength of the exciton–LO-phonon coupling is independent of the dot radius.[5] Marini, Strebe, and Kartheuser also used the donorlike exciton model and the adiabatic approximation and then obtained quite different results, i.e., an increase of polaronic effects with decreasing dot size.[7] More recently Fedorov, Baranov, and Inoue discussed the weak confinement case of an exciton and obtained that the LO-phonon effect for the cross section of resonant Raman scattering increases with the decrease of the dot size.[10]

The experimental situation is also confusing. Some experiments on resonant Raman scattering for II-VI compound nanocrystals have led to quite different (opposite) results for the size dependence of the strength of the exciton–LO-phonon coupling.[9,12]

In order to clarify this confusing situation for the size dependence of the exciton-phonon coupling, we pay attention to the most basic physical quantities for the exciton-phonon interaction, i.e., we calculate exciton energy, exciton-phonon interaction energy, and virtual phonon number involved in the exciton state. An exciton in a polar spherical quantum dot, embedded in a nonpolar matrix, is treated as in the previous studies.[4–12] Rather than the donorlike exciton model we start with the usual exciton model, where both an electron and a hole can move within the dot. The dielectric continuum model is used to describe the bulk-type and the interface-type LO phonons and their interaction with an exciton in the system.[3,5,13] In order to discuss effects of the exciton-phonon interaction a theory based on the variational intermediate coupling method is developed. In the present work we confine ourselves to the strong confinement case of an exciton, where an exciton arises from the quantized electron and hole states due to the individual confinement in the dot.[14] The method developed is applied to an exciton in GaAs and CdSe QD's in order to clarify the nature of polaronic effects on an exciton in the QD system.

II. METHOD

Let us consider an exciton which is confined perfectly in the spherical QD with radius R and high (low) frequency dielectric constant $\varepsilon_\infty(\varepsilon_0)$. The sphere is surrounded by the nonpolar matrix whose dielectric constant is ε_d. In the QD an exciton interacts with LO phonons. Then the Hamiltonian of the system is expressed as[3,5,7]

$$H = H_{\rm ex} + H_{\rm ph} + H_{\text{ex-ph}}. \tag{2.1}$$

Here the excitonic part $H_{\rm ex}$ takes the form of

$$H_{\rm ex} = \sum_{j=e,h} \left(\frac{\mathbf{p}_j^2}{2m_j} + V_{\rm conf}^{(j)} \right) - \frac{e^2}{\varepsilon_\infty |\mathbf{r}_e - \mathbf{r}_h|}, \tag{2.2}$$

where $j=e$ and $j=h$ denote an electron and a hole, respectively. The first and the second terms of the right hand side of Eq. (2.2) describe the kinetic energy of an electron and a hole with the confinement potential of the particle j, written as

$$V_{\rm conf}^{(j)}=\begin{cases}0 & (r_j<R)\\ \infty & (r_j>R).\end{cases} \tag{2.3}$$

The last term of the right hand side of Eq. (2.2) represents the Coulomb interaction between an electron and a hole.

The LO-phonon Hamiltonian $H_{\rm ph}$ can be written as

$$H_{\rm ph}=\sum_{s,\sigma}\hbar\omega_{s\sigma}a_{s\sigma}^{\dagger}a_{s\sigma}. \tag{2.4}$$

Here $\sigma=1$ (2) denotes the bulk- (interface-) type LO phonon. Another index s describes the quantum numbers $s=(n=1,2,3,...;l=0,1,2,...;m=0,\pm1,\pm2,...,\pm l)$ for the bulk-type phonon and $s=(l=1,2,3,...;m=0,\pm1,\pm2,...,\pm l)$ for the interface-type phonon. The creation (annihilation) operator of the $s\sigma$ mode is denoted by $a_{s\sigma}^{\dagger}(a_{s\sigma})$. The energy for the bulk-type LO phonon $\hbar\omega_{s1}$ is equal to just the bulk LO-phonon energy $\hbar\omega_{\rm LO}$, being independent of the index s. For the interface-type LO phonon energy $\hbar\omega_{s2}$ we have

$$\hbar\omega_{s2}\equiv\hbar\omega_l=\left[\frac{\epsilon_\infty[(l+1)\epsilon_d+l\epsilon_0]}{\epsilon_0[(l+1)\epsilon_d+l\epsilon_\infty]}\right]^{1/2}\hbar\omega_{\rm LO}\quad(\text{for any } m). \tag{2.5}$$

Exciton–LO-phonon interaction term $H_{\text{ex-ph}}$ is given by

$$H_{\text{ex-ph}}=\sum_{s,\sigma}\hbar\omega_{s\sigma}v_{s\sigma}[\{S_{s\sigma}(\mathbf{r}_e)-S_{s\sigma}(\mathbf{r}_h)\}a_{s\sigma}+\text{H.c.}], \tag{2.6}$$

where

$$S_{s1}(\mathbf{r})=j_l(k_{nl}r)Y_l^m(\theta,\varphi)\quad(r\leqslant R), \tag{2.7}$$

$$S_{s2}(\mathbf{r})=(r/R)^lY_l^m(\theta,\varphi)\quad(r\leqslant R). \tag{2.8}$$

Here the electron-bulk-type phonon coupling constant v_{s1} is written as

$$v_{s1}=\sqrt{\frac{8\pi\alpha_bR_p^{(e)}}{\mu_{ln}^2j_{l+1}^2(\mu_{ln})R}}, \tag{2.9}$$

where $R_p^{(e)}\equiv\sqrt{\hbar/2m_e\omega_{\rm LO}}$ is the polaron radius for an electron and μ_{ln} denotes the nth zero of the spherical Bessel function of the order l, i.e., $j_l(\mu_{ln})=0$. The well-known Fröhlich electron–LO-phonon coupling constant α_b is defined as

$$\alpha_b=\frac{e^2}{2R_p^{(e)}\hbar\omega_{\rm LO}}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right).$$

For the interface-type phonon, we have the coupling constant v_{s2} as follows:

$$v_{s2}=\sqrt{\frac{4\pi\alpha_sR_p^{(e)}}{R}}, \tag{2.10}$$

where α_s is defined as

$$\alpha_s=\alpha_b\left(\frac{\sqrt{l}\varepsilon_\infty}{l\varepsilon_0+(l+1)\varepsilon_d}\right)^2\left(\frac{\hbar\omega_{\rm LO}}{\hbar\omega_l}\right)^3.$$

Typical values of α_b, $\hbar\omega_{Il}$, and α_s for several semiconductor dots are summarized in Ref. 15.

The intermediate coupling method is used to treat the exciton–LO-phonon interaction. The Hamiltonian H and the exciton state $|\Psi\rangle$ are transformed to $\tilde{H}=U^{-1}HU$ and $|\tilde{\Psi}\rangle=U^{-1}|\Psi\rangle$ by performing the unitary transformation with the unitary operator $U(F_{s\sigma})$:

$$U(F_{s\sigma})=\exp\left(\sum_{s\sigma}\left[F_{s\sigma}^*(\mathbf{r}_e,\mathbf{r}_h)a_{s\sigma}-F_{s\sigma}(\mathbf{r}_e,\mathbf{r}_h)a_{s\sigma}^{\dagger}\right]\right). \tag{2.11}$$

The form of $F_{s\sigma}$ is taken to be

$$F_{s\sigma}(\mathbf{r}_e,\mathbf{r}_h)=v_{s\sigma}[f_{s\sigma}^{(e)}S_{s\sigma}^*(\mathbf{r}_e)-f_{s\sigma}^{(h)}S_{s\sigma}^*(\mathbf{r}_h)], \tag{2.12}$$

where $f_{s\sigma}^{(e)}$ and $f_{s\sigma}^{(h)}$ are variational parameters to be determined later. The choice of the form $F_{s\sigma}$ in Eq. (2.12) corresponds to the intermediate coupling method, being similar to the choices in Refs. 16 and 17 for the bulk and Ref. 18 for the quantum well. The most important term, i.e., the zero phonon term of the transformed Hamiltonian $\tilde{H}=U^{-1}HU$, is given by

$$\tilde{H}_0=\sum_{j=e,h}\left[\frac{1}{2m_j}(\mathbf{p}_j+\mathbf{j}_j)^2+V_{\rm conf}^{(j)}+\sum_{s\sigma}\frac{\hbar^2}{2m_j}\left|\nabla_jF_{s\sigma}\right|^2\right]+\sum_{s,\sigma}\hbar\omega_{s\sigma}|F_{s\sigma}|^2-\frac{e^2}{\epsilon_\infty|\mathbf{r}_e-\mathbf{r}_h|}-\sum_{s,\sigma}\hbar\omega_{s\sigma}v_{s\sigma}\{[S_{s\sigma}(\mathbf{r}_e)-S_{s\sigma}(\mathbf{r}_h)]F_{s\sigma}+\text{c.c.}\}. \tag{2.13}$$

Here $\mathbf{j}_j$ is defined as $\mathbf{j}_j=-(i\hbar/2)\Sigma_{s,\sigma}[F_{s\sigma}^*\nabla_jF_{s\sigma}-F_{s\sigma}\nabla_jF_{s\sigma}^*]$. For the trial function to the transformed state $|\tilde{\Psi}\rangle$, the product of the exciton state $|\Phi_{\rm ex}\rangle$ and the zero phonon state $|0\rangle$, i.e., $|\tilde{\Psi}\rangle=|\Phi_{\rm ex}\rangle|0\rangle$, is chosen. For the lowest exciton state $|\Phi_{\rm ex}\rangle$, we choose the following form:

$$\Phi_{\rm ex}(\mathbf{r}_e,\mathbf{r}_h)=\frac{1}{\sqrt{\mathcal{N}}}j_0(\pi r_e/R)j_0(\pi r_h/R)\exp[-\beta|\mathbf{r}_e-\mathbf{r}_h|]. \tag{2.14}$$

Here $\mathcal{N}$ is a normalization constant and β is a variational parameter. This wave function is valid for a strong confinement case such as $R\leqslant(2\sim3)a_B$, where a_B is the exciton Bohr radius in the bulk.[14]

We can choose that both $f_{s\sigma}^{(e)}$ and $f_{s\sigma}^{(h)}$ are real and satisfy the relations $f_{(n,l,m)1}^{(j)}=f_{(n,l,-m)1}^{(j)}$ and $f_{(l,m)2}^{(j)}=f_{(l,-m)2}^{(j)}$, for which $\mathbf{j}_j=0$ holds. From the variational condition $\partial\langle\tilde{\Psi}|\tilde{H}|\tilde{\Psi}\rangle/\partial f_{s\sigma}^{(j)}=0$, the variational parameters $f_{s\sigma}^{(j)}$ is obtained as

$$f_{s\sigma}^{(j)}=\frac{(C_{s\sigma}^{(i)}+A_{s\sigma}+B_{s\sigma})(A_{s\sigma}-B_{s\sigma})}{(C_{s\sigma}^{(i)}+A_{s\sigma})(C_{s\sigma}^{(j)}+A_{s\sigma})-B_{s\sigma}^2}(i\neq j). \tag{2.15}$$

Here $i,j=e$ or h and

$$A_{s\sigma}=\langle\Phi_{\rm ex}||S_{s\sigma}(\mathbf{r}_e)|^2|\Phi_{\rm ex}\rangle=\langle\Phi_{\rm ex}||S_{s\sigma}(\mathbf{r}_h)|^2|\Phi_{\rm ex}\rangle, \tag{2.16}$$

$$B_{s\sigma}=\langle\Phi_{\rm ex}|S_{s\sigma}(\mathbf{r}_e)S_{s\sigma}^*(\mathbf{r}_h)|\Phi_{\rm ex}\rangle, \tag{2.17}$$

$$C_{s\sigma}^{(j)}=\frac{\hbar^2}{2m_j\omega_{s\sigma}}\langle\Phi_{\rm ex}||\nabla_jS_{s\sigma}(\mathbf{r}_j)|^2|\Phi_{\rm ex}\rangle. \tag{2.18}$$

TABLE I. Material parameters used in the calculation: the electron mass m_e, the hole mass m_h, the static dielectric constant ϵ_0, the high frequency dielectric constant ϵ_∞, and the bulk LO-phonon energy $\hbar\omega_{\rm LO}$ (Ref. 19), the Fröhlich electron–LO-phonon coupling constant α_e, the hole–LO-phonon coupling constant α_h, the polaron radius of electron $R_p^{(e)}$, the polaron radius of hole $R_p^{(h)}$, and the exciton Bohr radius a_B. Units of mass, energy, and length are free electron mass, meV, and angstrom, respectively.

	m_e (m)	m_h (m)	ϵ_0	ϵ_∞	$\hbar\omega_{\rm LO}$ (meV)	α_e	α_h	$R_p^{(e)}$ (Å)	$R_p^{(h)}$ (Å)	a_B (Å)
GaAs	0.067	0.625	13.8	10.9	36.3	0.08	0.24	39.6	13.0	95.3
CdSe	0.1	0.4	9.3	6.1	26.5	0.42	0.85	36.2	18.1	36.7

Then, the exciton energy $E_{\rm ex}$ is calculated from the numerical minimization of the expectation value for an energy with respect to the only remaining variational parameter β, i.e.,

$$E_{\rm ex}=\min_{\beta}\{\langle\Psi|H|\Psi\rangle\}$$
$$=\min_{\beta}\Bigg(\langle\Phi_{\rm ex}|H_{\rm ex}|\Phi_{\rm ex}\rangle - \sum_{s\sigma}\hbar\omega_{s\sigma}|v_{s\sigma}|^2(A_{s\sigma}-B_{s\sigma})(f_{s\sigma}^{(e)}+f_{s\sigma}^{(h)})\Bigg). \tag{2.19}$$

In order to see the strength of the exciton-phonon coupling, we pay attention to the following exciton-phonon interaction energy $E_{\text{ex-ph}}$:

$$E_{\text{ex-ph}}=\Bigg(-\sum_{s\sigma}\hbar\omega_{s\sigma}|v_{s\sigma}|^2(A_{s\sigma}-B_{s\sigma})(f_{s\sigma}^{(e)}+f_{s\sigma}^{(h)})\Bigg)_{\beta}, \tag{2.20}$$

which is the contribution due to the exciton–LO-phonon interaction in Eq. (2.19) for the value of β determined. We also calculate the virtual phonon number N involved in the exciton state Ψ. This is given by

$$N=\left\langle\Psi\left|\sum_{s,\sigma}a_{s\sigma}^{\dagger}a_{s\sigma}\right|\Psi\right\rangle, \tag{2.21}$$

which indicates the involvement of LO phonons in the exciton state $\Psi=U|\Phi_{\rm ex}\rangle|0\rangle$.

III. RESULTS AND DISCUSSIONS

As typical examples for III-V and II-VI semiconductor quantum dots we consider GaAs and CdSe quantum dots and calculate exciton energy, exciton–LO-phonon interaction energy, and virtual phonon number, using the above formulation. The material parameters for GaAs and CdSe, used in the calculation, are given in Table I. The value of the dielectric constant for the nonpolar glass matrix in the barrier region ϵ_d is chosen to be 6, which is the value for the Pyrex1710.

The calculated results for the exciton energy $E_{\rm ex}$, the exciton-phonon interaction energy $E_{\text{ex-ph}}$, and the virtual phonon number N are shown in Figs. 1–3. The corresponding values for the bulk GaAs (CdSe) are -16.5 (-50.0) meV for $E_{\rm ex}$, -9.74 (-19.0) meV for $E_{\text{ex-ph}}$, and 0.109 (0.168) for N: these values are calculated by the intermediate coupling method of Refs. 14 and 16 and are indicated by the arrows in Figs. 1–3.

The exciton energy $E_{\rm ex}$ is shown in Fig. 1(a) for GaAs and Fig. 1(b) for CdSe. It is seen in Fig. 1 that exciton energy approaches the bulk value for the larger dot and increases for the decrease of the dot radius. This increase of the exciton energy for the smaller dot reflects the increase of the confinement effect: in the small QD the leading term of the lowest exciton energy without exciton-phonon interaction is given by $E_{\rm ex}=\hbar^2\pi^2/2\mu R^2$, where μ denotes the reduced mass for an electron and a hole.[14] This yields a rapid increase

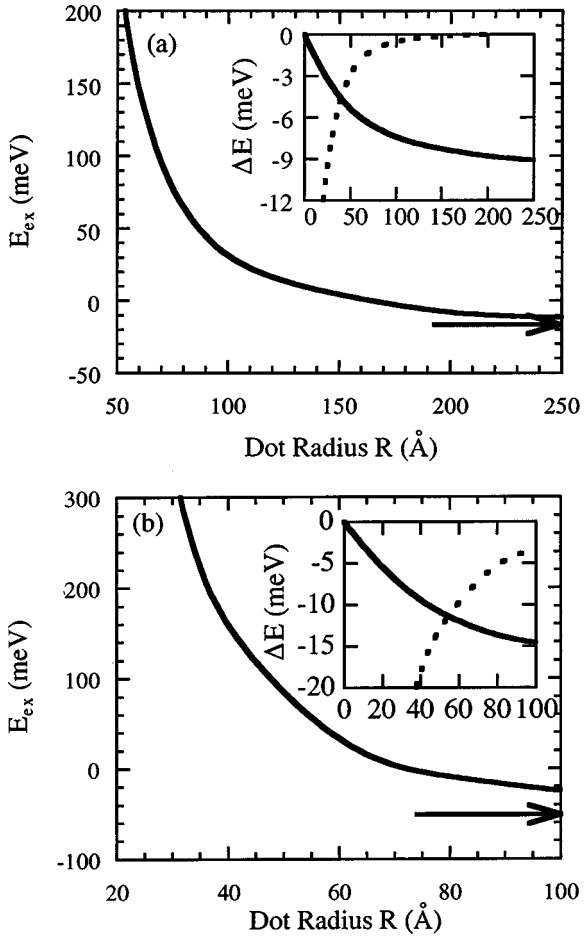

FIG. 1. The dot radius R dependence of the lowest exciton energy $E_{\rm ex}$ for GaAs (a) and CdSe (b) spherical quantum dots embedded in a glass matrix with $\epsilon_d=6$. The bulk values for $E_{\rm ex}$ are indicated by arrows. In the inset the differences from two simple models, i.e., the bare exciton model (the dashed line) and the shallow exciton model (the dotted line), are shown.

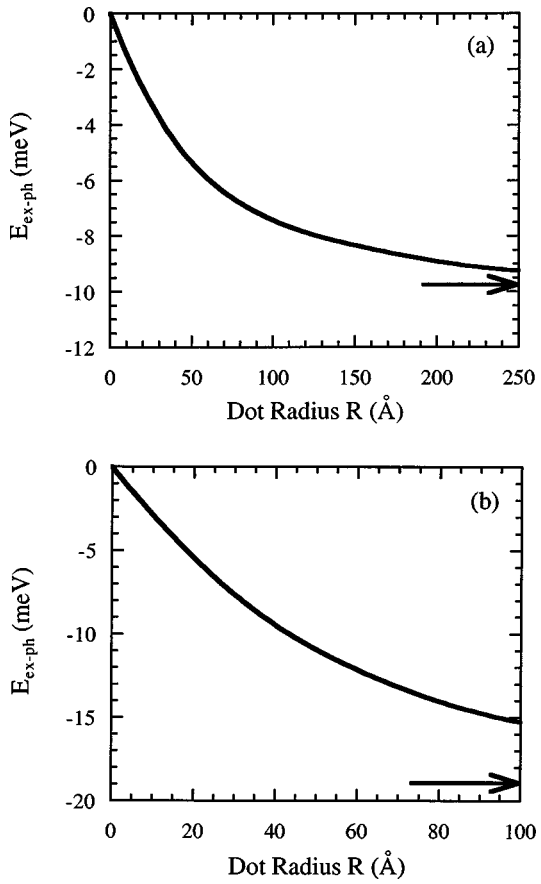

FIG. 2. The dot radius R dependence of the exciton-phonon interaction energy $E_{\text{ex-ph}}$ for GaAs (a) and CdSe (b) spherical quantum dots embedded in a glass matrix with $\epsilon_d=6$. The bulk values for $E_{\text{ex-ph}}$ are indicated by arrows.

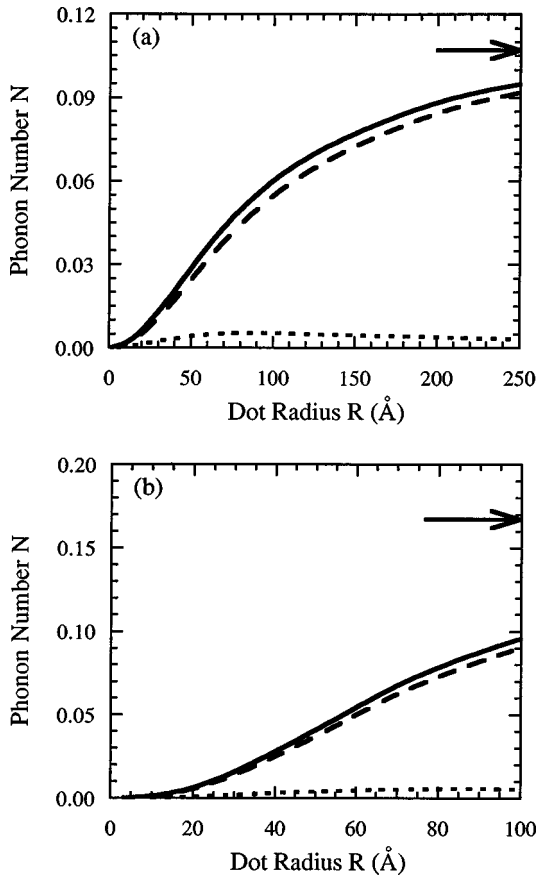

FIG. 3. The dot radius R dependence of the virtual phonon number N for GaAs (a) and CdSe (b) spherical quantum dots embedded in a glass matrix with $\epsilon_d=6$. The solid line stands for the virtual phonon number N, the dashed line for the contribution of the bulk-type phonon, and the dotted line for the contribution of the interface-type phonon. The bulk values for N are indicated by arrows.

of the exciton energy for the very small QD as seen in Fig. 1. In order to see the effects of the LO phonon on the exciton energy in detail, we also calculate the lowest exciton energies for the two simple models. One is the bare exciton model, where exciton-phonon interaction is neglected completely, and then exciton energy $E_{\text{ex}}^{(b)}$ is obtained from

$$E_{\text{ex}}^{(b)}=\min_{\beta}\{\langle\Phi_{\text{ex}}|H_{\text{ex}}|\Phi_{\text{ex}}\rangle\}, \tag{3.1}$$

where the exciton Hamiltonian H_{ex} is given by Eq. (2.2). Another is the shallow exciton model for which the following effective Hamiltonian is used:

$$H^{(s)}=\sum_{j=e,h}\left(\frac{p_j^2}{2m_j}+V_{\text{conf}}^{(j)}\right)-\frac{e^2}{\epsilon_0|\mathbf{r}_e-\mathbf{r}_h|}-(\alpha_e+\alpha_h)\hbar\omega_{\text{LO}}, \tag{3.2}$$

and then the exciton energy can be obtained from

$$E_{\text{ex}}^{(s)}=\min_{\beta}\{\langle\Phi_{\text{ex}}|H^{(s)}|\Phi_{\text{ex}}\rangle\}. \tag{3.3}$$

In this model, effects of the exciton-phonon interaction are described by the following: the Coulomb interaction between an electron and a hole is screened by the static dielectric constant ϵ_0 and both an electron and a hole receive the individual polaron self-energy shifts in the bulk, $-\alpha_e\hbar\omega_{\text{LO}}$ and $-\alpha_h\hbar\omega_{\text{LO}}$, respectively. This model is valid for the shallow exciton limit (for the very large radius exciton) with the small electron-phonon coupling case in the bulk, as in GaAs.[16,20] The inset in Fig. 1 shows the calculated values of the following energy differences between E_{ex} and $E_{\text{ex}}^{(b)}$ or $E_{\text{ex}}^{(s)}$:

$$\Delta E^{(b)}=E_{\text{ex}}-E_{\text{ex}}^{(b)}, \tag{3.4}$$

$$\Delta E^{(s)}=E_{\text{ex}}-E_{\text{ex}}^{(s)}. \tag{3.5}$$

From this figure, the following two points are seen. The first is that for the smaller dot $\Delta E^{(b)}$ becomes smaller, i.e., the exciton energy E_{ex} approaches the bare exciton energy $E_{\text{ex}}^{(b)}$: there the exciton-phonon coupling becomes smaller and eventually vanishes in the small dot limit, as pointed out in Ref. 4. The second is that for the larger dot the exciton energy deviates greatly from the bare exciton energy and is closer to the shallow exciton energy: this result indicates that the LO-phonon effects are important. In the inset in Fig. 1 we notice that the crossing of $\Delta E^{(b)}$ and $\Delta E^{(s)}$ occurs at the QD radius around 45 Å for the GaAs QD and 55 Å for the CdSe QD. These values are close to the sum of the electron polaron radius $R_p^{(e)}$ and the hole polaron radius $R_p^{(h)}$: from Table I the value of $R_p^{(e)}+R_p^{(h)}$ is obtained to be about 53–54

Å for both materials. Therefore we may say that the polaronic contribution of an exciton becomes much smaller when the dot radius becomes smaller than the sum of the electron and the hole polaron radii. In this region the cancellation of the polaron effects for an electron and a hole becomes very effective.

It is also seen clearly in Figs. 2 and 3 that in the limit of the small dot radius the exciton-phonon interaction energy $E_{\text{ex-ph}}$ as well as the virtual phonon number N become zero and, with the increase of the dot radius, $|E_{\text{ex-ph}}|$ and N increase and then gradually approach the bulk value. The vanishing of the polaronic effects in the small radius limit is natural because, as stated above, the cancellation of the polaronic effects due to the opposite charge of an electron and a hole becomes stronger for the smaller dot and then the polaronic effects of an exciton become smaller. The present calculation describes this situation very well.

Figure 3 shows generally that the polaronic contribution of the bulk-type phonon is much larger than that of the interface-type phonon. These figures also show the different behavior of the contributions due to both phonons. The bulk-type phonon, being quantized, is confined in the dot and its vibrational amplitude is zero at the interface of the dot. The interface-type phonon has a vibrational amplitude which is the largest at the interface of the dot and decreases to zero along the direction to the center of the dot. The difference of these vibrational amplitudes, appearing in the exciton-phonon coupling in Eqs. (2.7) and (2.8), and the cancellation of electron and hole polaron effects in the small dot lead to the quite different behavior of the polaronic contributions in Fig. 3; when the dot size increases, the contribution due to the bulk-type phonon increases from zero to the bulk values, while that due to the interface-type phonon increases from zero, reaches the maximum, and decreases.

In passing it is noted that we have also performed the calculation with the consideration of the image charge effects, which arises from the different dielectric constants inside and outside the QD.[21] The results obtained are essentially the same as above. For example, values of the exciton energy E_{ex} are 144.2 (146.4) meV for $R=60$ Å in GaAs and 160.6 (160.3) meV for $R=40$ Å in CdSe with (without) the image charge effects. Thus the image charge effects do not affect the discussion on the exciton-phonon interaction effect much in the present case.

Finally we mention the validity of the intermediate coupling method. The intermediate coupling method is valid when energy differences of the relevant exciton state from other exciton states are not much larger than the LO-phonon energy. This situation is realized for many bulk semiconductors and many QD systems with a QD radius that is not as small as $R>a_B$. The opposite limit occurs for the small QD such that $R\ll a_B$, where the exciton states are mainly governed by the individual confinement states of an electron and a hole. In this case the adiabatic approximation, which corresponds to the choice of the $\mathbf{r}_j$-independent $F_{s\sigma}=v_{s\sigma}g_{s\sigma}$ in Eq. (2.12), is valid. For this adiabatic choice, after some calculation in a way similar to that above, we obtain the result that the polaron effects of an electron and a hole completely cancel and thus there is no polaronic effect on the exciton. From the fact that there is no adiabatic contribution to the polaronic effects for an exciton, the intermediate coupling method is considered to be valid for the present problem. To confirm the validity of the intermediate coupling method further, we have also performed the calculation with the choice of $F_{s\sigma}=v_{s\sigma}[f_{s\sigma}^{(e)}S_{s\sigma}(\mathbf{r}_e)-f_{s\sigma}^{(h)}S_{s\sigma}(\mathbf{r}_h)+g_{s\sigma}]$, which can interpolate both the intermediate coupling and adiabatic cases and is the extension of the choice for the single polaron in QD (Ref. 15) to that for the exciton. The results obtained for the present GaAs and CdSe quantum dots are almost the same as those obtained by the intermediate coupling method in Figs. 1–3. Thus the intermediate coupling method is appropriate for the present exciton-phonon interaction problem in the QD system.

In the above we have seen that there are no adiabatic contributions to polaronic effects for an exciton in the present usual exciton model where both an electron and a hole can move in a dot. The situation is very different if we use the donorlike exciton model and eliminate the hole coordinate by the transformation as done in Ref. 7. There the problem essentially reduces to the bound polaron problem in QD systems and then the result of the large increase of the polaron effect on an electron in the small QD has been obtained in Ref. 7 as in the single polaron problem in QD.[15] Thus we think that it is not suitable to use the donorlike exciton model to discuss the LO-phonon effect (polaronic effects) on an exciton in a QD system, where the proper consideration of the cancellation of the polaronic effects for an electron and a hole is important.

IV. CONCLUSION

In the present work we have treated LO-phonon effects on an exciton in a quantum dot surrounded by a nonpolar barrier matrix in the perfect confinement case.

The variational calculation of the most basic quantities, i.e., exciton energy, exciton-phonon interaction energy, and virtual phonon number, clearly shows the nature of the involvement of the LO phonon in the exciton state: the vanishing of the polaronic effects in the small dot limit and the gradual increase to the bulk values for the larger dot. It is stressed that the detailed consideration of the LO-phonon effects on an exciton is necessary for the QD systems because of the large cancellation of the polaron effect of an electron and a hole for the small dot: the use of the expression derived for the bulk crystal, for example, the use of the Haken potential[20] for the effective interaction between an electron and a hole as done in Ref. 22, is not generally justified. We believe the present result reflects the nature of the LO-phonon effect common to many three-dimensionally confined systems. The appropriate theoretical analysis of physical phenomena such as Raman scattering and relaxation of the exciton state will also clarify the nature of the LO-phonon effect. We think that our result plays an important role in these studies.

Finally we mention remaining problems related to the present work. One is the effects of the finite potential barrier. In the present work we confine ourselves to the perfect confinement case of an exciton in the QD. If the potential barrier is finite, the penetration of the wave function to the barrier region becomes large for a smaller dot. The exciton-phonon interaction for this situation is interesting and needs study: it is expected that there the larger amplitude of the exciton

wave function in the nonpolar barrier matrix as well as the larger cancellation of the electron and hole polaron effect in QD yield the smaller polaronic effects for the small QD. Another problem is the effects of the polar barrier matrix. There are many experiments in the quantum dot system whose barrier region has LO phonons, such as a system of a polar quantum dot embedded in another polar crystal. So far there has been no theoretical work on the electron–LO-phonon interaction and polaronic effects for this problem, to which the present variational method can be applied also.

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