Exciton-phonon interaction effects in quantum wells

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Properties of excitons interacting with interface optical phonons and confined longitudinal optical phonons in semiconducting compound quantum wells are investigated theoretically. By using the nonseparable trial wave function the exciton binding energies and the interaction energies of excitons with every phonon mode in two typical quantum-well structures, GaAs/AlAs and CdSe/ZnSe, are calculated as a function of the well width. Some interesting features of the exciton-phonon interaction system in quantum wells are found and discussed. The theoretical result gives a reasonable explanation of the agreement of some simple exciton theories with experiments. [S0163-1829(97)03228-1]

In recent years the investigation of the property of excitons in quantum wells (QW's) has become an important topic of the condensed-matter physics. Most of the works[1–5] that studied the exciton state in a QW considered mainly the confinement effects of the well potential, including or not the finite-barrier potential, valence-band mixing, nonparabolicity of the dispersion relation, mass and dielectric constants mismatch, etc. Qualitative and quantitative results of the exciton binding energy in QW's were reported in agreement with experimental results. There are many works[6–13] that consider exciton-phonon interaction effects in QW's by using the bulk phonon approximation. Some authors found that the polaronic effects on the binding of the Wannier exciton in a QW are rather noticeable and increase with decreasing well thickness.[6–8] It was soon pointed by one of the present authors[9] that the polaronic effects in a QW are not as large as those in Refs. 6–8, in which the large polaron self-energy shifts of the electron and hole subbands are neglected. Koinov[12] presented a theory of excitons in QW's based on the Bethe-Salpeter equation. He found that in the limit of the thin layer the interaction of excitons with LO phonons caused a decrease in the exciton binding energy of the order of 18%. This theoretical result is different from other authors' results.

Very recently some improved theories[14–18] have been introduced to study the exciton-phonon interaction in QW's. By including interface optical (IO) phonon effect, Gu and co-workers,[15,16] and also Chuu *et al.*[17] have studied the IO and LO phonon effects on excitons in polar semiconductor QW's, but the interaction expression of IO phonons with a charge carrier was extended directly from the surface phonon theory, which is not entirely the correct picture for the QW cases. Xie and Chen[18] have studied the ground state of both heavy- and light-hole excitons in a $GaAs/Ga_{1-x}Al_x$ As QW. Based on the work of Mori and Ando,[19] they took all the phonon modes into account, namely, the four branches of IO phonons, the confined LO phonon, and the half-space LO phonon. But, unfortunately, some miscalculations are found in their paper, which made the expressions and also the numerical results in their paper not entirely correct. Up to now, the roles of the phonons, especially IO phonons, in the QW exciton state have not been very clear. A detailed study of the exciton-phonon interaction in a QW remains an important work in order to understand the optical and electronic properties of QW structures, also in view of their applications to new devices.

In this paper we report an investigation of the interaction of the exciton with IO-phonon and confined LO-phonon modes in polar semiconductor QW's. The aim of the present paper is to give a clear picture of the polaronic effects on QW exciton states by a rigorous variational calculation. The results are shown to be valid throughout the entire well width range, corresponding in the thin and thick limits to two- and three-dimensional situations, respectively. Two typical QW structures, GaAs/AlAs and CdSe/ZnSe, are put into the calculation: one is a III-V polar compound QW with a weak polaronic effect and other is a II-VI polar compound QW with a strong polaronic effect. Some interesting features of the exciton-phonon system in QW's are found and discussed.

Since we are interested only in the exciton-phonon interaction problem, the simple models of the QW and the exciton will be chosen in this paper. The Hamiltonian of an exciton interacting with optical phonons in a polar semiconductor QW is expressed as

$$H=\sum_j \left\{\frac{P_{j\|}^2}{2m_{j\|}}+\frac{P_{jz}^2}{2m_{jz}}+V(z_j)\right\}-\frac{e^2}{\epsilon_\infty r}+H_{ph}+H_i, \quad (1)$$

where $j=1$ and 2 refer to the electron and hole, respectively. $\mathbf{P}=(\mathbf{P}_{j\|},P_{jz})$, $\mathbf{r}=(\rho_j,z_j)$, and $m=(m_{j\|},m_{jz})$ are the momentum, position, and effective band mass of the particles. $\mathbf{P}_{j\|}$ and $\boldsymbol{\rho}$ are two-dimensional vectors in the x-y plane. $V(z_j)$ is the infinite-height square well barrier potential for the electron ($j=1$) and hole ($j=2$). The width of the QW is W ($=2d$). $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2=(\boldsymbol{\rho},z)$ is the relative position of the electron and hole. ϵ_∞ is the optical dielectric constant of the well material.

The free phonon Hamiltonian H_{ph} and the exciton–optical-phonon interaction Hamiltonian H_i are given by

$$H_{ph}=\sum_\beta \sum_{\mathbf{k}} \hbar\omega_\beta(\mathbf{k})a_{\mathbf{k}\beta}^\dagger a_{\mathbf{k}\beta}, \quad (2)$$

$$H_i=\sum_\beta \sum_{\mathbf{k}} \sum_j [\theta_j V_{\mathbf{k}\beta}L_{\mathbf{k}\beta}(z_j)\exp(i\mathbf{k}\cdot\rho_j)a_{\mathbf{k}\beta}+\text{H.c.}], \quad (3)$$

where $a^{\dagger}_{\mathbf{k}\beta}(a_{\mathbf{k}\beta})$ is the creation (annihilation) operator of an optical phonon with frequency $\omega_{\beta}(\mathbf{k})$ and wave vector $(\mathbf{k},k_{\beta})$. Here β denotes the mode index: $\beta=m$ refers to the confined LO phonon in the well material and $\beta=(\sigma,p)$ to the IO phonon. The index $p(=+,-)$ refers to the symmetric and antisymmetric IO-phonon modes and $\sigma(=+,-)$ to the high- and low-frequency IO-phonon modes, respectively. The dispersion relations of the frequency $\omega_{\beta}(\mathbf{k})$ and the interaction coefficient $V_{\mathbf{k}\beta}L_{\mathbf{k}\beta}(z_j)$ are well understood and the readers who are interested in it can refer to the Refs. 20–23. $\theta_1=1$ and $\theta_2=-1$ show that the electron– and hole–optical-phonon interactions have opposite signs.

The above Hamiltonian is so complicated that the accurate eigenfunction and eigenvalue are almost impossible to find. Therefore, some approximation method must be used. In this paper we adopt an improved variational method,[9,22,23] which has been used extensively to study the exciton-phonon interaction problem in QW's. The trial wave function is assumed to take the form

$$|\Psi\rangle=U|0\rangle|\Psi_{ex}\rangle, \tag{4}$$

where $|0\rangle$ is the zero-phonon state, U is a unitary transformation operator, which is defined as

$$U=\exp\left(\sum_{\beta}\sum_{\mathbf{k}}\sum_{j} f_{\mathbf{k}\beta j}L_{\mathbf{k}\beta}(z_j)\exp(-i\mathbf{k}\cdot\boldsymbol{\rho}_j)a^{\dagger}_{\mathbf{k}\beta}-\text{H.c.}\right), \tag{5}$$

$f_{\mathbf{k}\beta j}$ is the variational parameter, which will be determined by the variational condition

$$\delta\langle\Psi|H|\Psi\rangle/\delta f_{\mathbf{k}\beta j}=0, \tag{6}$$

and $|\Psi_{ex}\rangle$ is the exciton wave function in the QW and is chosen as

$$\Psi_{ex}=N\exp\left(-\frac{r}{\lambda}\right)\cos\left(-\frac{\pi}{2d}z_1\right)\cos\left(-\frac{\pi}{2d}z_2\right), \tag{7}$$

where N is determined by normalization conditions and λ is a variational parameter that will be determined by the procedure of minimizing the ground-state energy of the system. The trial wave function Ψ_{ex} not separable in spatial coordinates can give out reasonable energies throughout the entire well width range[1] and has been used extensively by many authors.

The ground-state energy E_g and its binding energy E_b of the exciton-phonon system in a QW are calculated, respectively, by the process of

$$E_g=\min_{\lambda}E(\lambda)=\min_{\lambda}\langle\Psi|H|\Psi\rangle, \tag{8}$$

$$E_b=E(\lambda\rightarrow\infty)-E_g, \tag{9}$$

where $E(\lambda\rightarrow\infty)$ (noted as E_{po}) is just the sum of the free polaron energies of the electron and hole in the same QW.

The heavy-hole excitons in two typical QW structures, GaAs/AlAs and CdSe/ZnSe, have been studied numerically. GaAs and AlAs are III-V compounds in which the polaronic effects are very weak and the effective exciton Bohr radii are very large. CdSe and ZnSe are of II-VI compounds in which the Fröhlich interactions are relatively strong and the exciton radii are small. The two QW structures have been extensively studied both experimentally and theoretically. The material parameters are listed in Table I. Quantitative investigations on the typical QW structures will give us a general knowledge of the QW exciton-phonon interaction problems.

Although our theory can give correct theoretical results in the two-dimensional limit, it should not be compared directly with experiments since the infinite barrier approximation is not reasonable in this case. As we have discussed in Ref. 22, if the probability of the particle penetrating into the finite-height barrier region is much smaller, the perfect carrier confinement approximation is reasonable. The conduction-(valence-) band discontinuities are 1.3 eV (0.23 eV) (Ref. 27) for the GaAs/AlAs QW and 0.826 eV (0.23 eV) (Ref. 25) for the CdSe/ZnSe QW, respectively. After quantum-mechanical calculations we found that the probability of the electron and hole moving out of the finite square QW's is less than 0.01 in the range of the well width larger than one polaron radius. Thus we can say that our calculations in this paper give reasonable results in the range of the well width larger than one polaron radius. The polaron radius is defined by $R_P=(\hbar/2m_e\omega_L)^{1/2}$, which is 39.75 Å in the GaAs/AlAs QW (Ref. 24) and 33.23 Å in the CdSe/ZnSe QW,[26] respectively.

In Fig. 1 we plotted the binding energies of the exciton-phonon systems in the GaAs/AlAs and CdSe/ZnSe QW's as a function of the well width. The energies of the exciton without an interaction with phonon and with dielectric constants ϵ_0 and ϵ_{∞} are calculated by the same trial wave function Ψ_{ex}; the corresponding binding energies are denoted as

TABLE I. Material parameters ω_L and ω_T are in units of meV and m_e and m_h are in units of the mass of a free electron. α is the Fröhlich coupling constant of the electron.

Substance	ϵ_0	ϵ_{∞}	ω_L	ω_T	m_e	m_h	α
GaAs[a]	13.18	10.89	36.25	33.29	0.067	0.62	0.068
AlAs[a]	10.06	8.16	50.09	44.88	0.150	0.76	0.126
CdSe[b]	9.3	6.1	26.54	21.50	0.13	0.45	0.46
ZnSe[b]	8.33	5.9	30.48	25.65	0.171	0.60	0.432

[a]Reference 24.

[b]m_h from Ref. 25, others from Ref. 26.

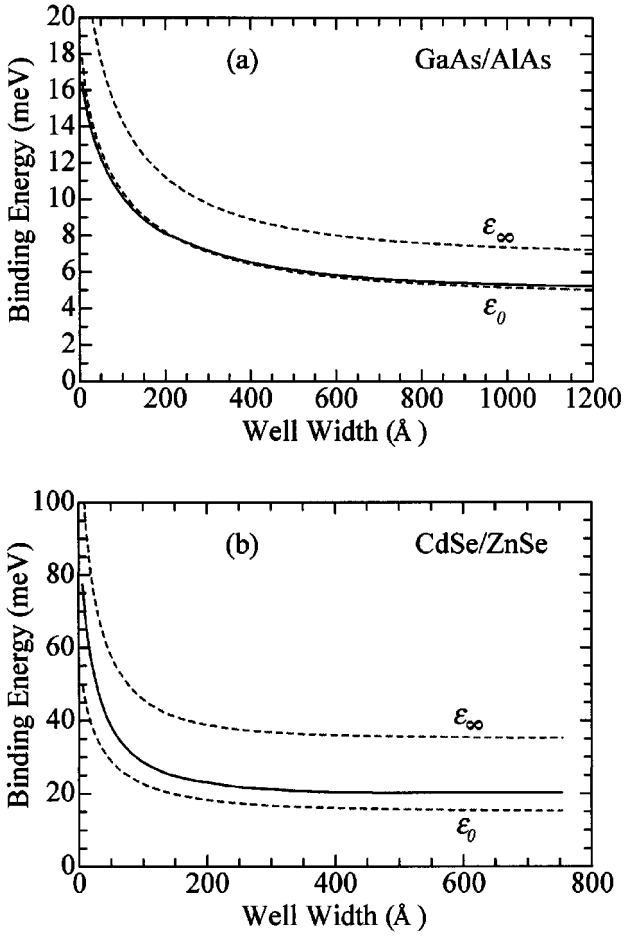

FIG. 1. Binding energies of the excitons as a function of the well width in the (a) GaAs/AlAs QW and (b) CdSe/ZnSe QW. The two dashed lines labeled with ϵ_0 and ϵ_∞ correspond to $E(\epsilon_0)$ and $E(\epsilon_\infty)$, respectively.

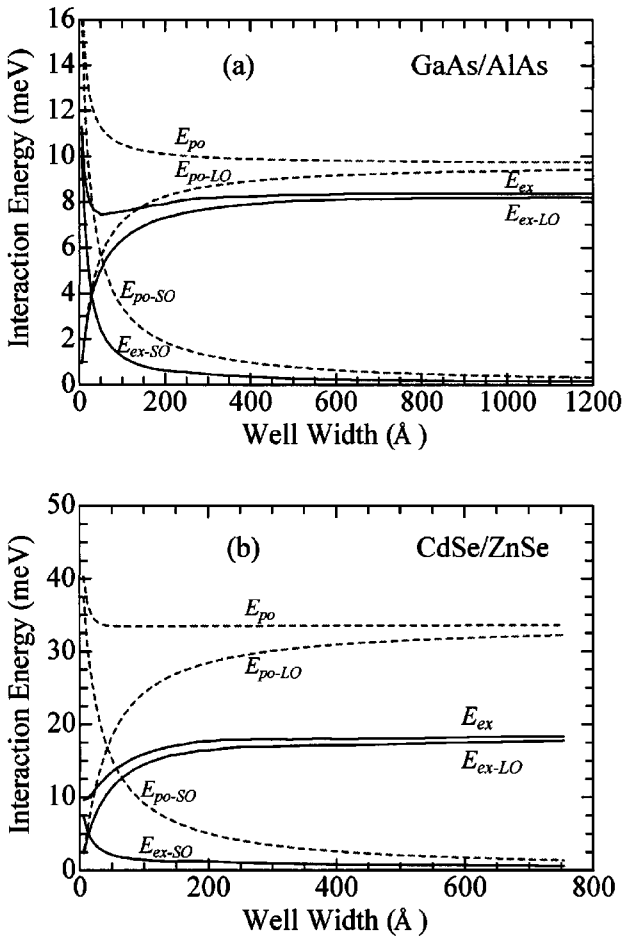

FIG. 2. Interaction energies of the exciton with phonons as a function of the well width in the (a) GaAs/AlAs QW and (b) CdSe/ZnSe QW. $E_{ex\text{-}LO}$ and $E_{ex\text{-}IO}$ are the interaction energies of the exciton with LO and IO phonons, respectively. $E_{ex\text{-}ph}$ is the interaction energy of the exciton with LO and IO phonons. The dashed lines correspond to the polaron energies of the electron and hole with LO and/or IO phonons.

$E(\epsilon_0)$ and $E(\epsilon_\infty)$, respectively, and are plotted in Fig. 1 for comparison. From the numerical results we see that the feature of the binding energy of the exciton–optical-phonon coupling system depends on the Fröhlich coupling strengths of the exciton in the well and in the barrier. In the case of the GaAs/AlAs QW, because the strength in the barrier is larger than in the well, the exciton binding energy is close to $E(\epsilon_0)$ as the well width is reduced, and when $W \leqslant 200$ Å it is even little lower than $E(\epsilon_0)$. The case of the CdSe/ZnSe QW is the opposite: the binding energy goes to $E(\epsilon_\infty)$ as the well width is reduced.

It is interesting that the theoretical result of the binding energy of the exciton-phonon system in the GaAs/AlAs QW is almost the same of $E(\epsilon_0)$ in the thin QW case ($W \leqslant 600$ Å). Now we understand with clear physical pictures why the simplest model, in which the polaronic effect was not considered, used by Bastard *et al.*[1] and many other authors can give reasonable results in agreement with the experiments on the GaAs-based QW's. Because the exciton Bohr radius [=115 Å (Ref. 24)] is much larger than the polaron radius and the Fröhlich coupling constant in the AlAs barrier is much larger than in the GaAs well, the polaronic effects made the Coulomb interaction in the exciton in the GaAs/AlAs QW screened by the static dielectric constant completely.

The interaction energies of the exciton with various phonon modes are plotted in Fig. 2. In order to give a clear picture of the role of the polaronic effects on the exciton states, the corresponding free polaron energies of the electron and hole interacting with the phonons in the same QW are plotted with dashed lines. From Fig. 2 we see some general features of the exciton–optical-phonon coupling system in polar semiconductor QW's. (i) The exciton–optical-phonon interaction is reduced with decreasing well width, except for the very thin well case where the infinite barrier model is not a reasonable model. (ii) As the well width is reduced to the thin limit, the importance of IO phonons increases drastically. The contribution is mainly from the ω_{++} phonon branch, which is also called the ω_{L2}-type branch because $\omega_{++} \rightarrow \omega_{L2}$ in the $d \rightarrow 0$ limit. Although the perfect carrier confinement approximation is used in the paper, in the narrow QW case the ω_{L2}-type IO phonon mode will be in the dominant position and the characteristics of the exciton-phonon interaction will appear to be similar to the features in the barrier material. (iii) If we compare the binding energy E_b in Fig. 1 with the exciton-phonon interaction

energy $E_{ex\text{-}ph}$ in Fig. 2, it should be realized that the exciton-phonon interaction plays an important role in the exciton state and affects the position of exciton peaks in the optical spectra of the QW structures and also the lifetime, linewidth, and so on.

Feature (i) is in contradiction with some published theoretical results,[6–8] but is consistent with the experiment results. Pelekanos *et al.*[28] found that there is a drastic reduction in the exciton-phonon coupling with decreasing well thickness in the (Cd,Zn)Se/ZnSe QW's. Similar studies in the GaAs-based systems also have reported the same but relatively smaller effects as one goes from the three- to the two-dimensional limit.[29] As shown in Fig. 2, our results are in qualitative agreement with the experiments.

In summary, we have present a theoretical investigation of the properties of the exciton–optical-phonon coupling system in two typical compound quantum wells, CdSe/ZnSe and GaAs/AlAs. The binding energies and the exciton–IO-phonon and –LO-phonon interaction energies in the QW's were calculated with an improved variational method. Some interesting features of the exciton–optical-phonon interaction in the two typical QW structures have been found and discussed.

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