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Physica B 215 (1995) 397–403

PHYSICA B

Effects of electron–phonon interaction on exciton binding energy in a quantum well

Der-San Chuu,[a,*] Win-Long Won,[b] Jui-Hsiang Pei[c]

[a] *Institute of Physics, National Chiao Tung University, Hsinchu 30050, Taiwan, ROC*
[b] *Department of Electrophysics, National Chiao Tung University, Hsinchu 30050, Taiwan, ROC*
[c] *Department of Science and Mathematics Education, National Hsin Chu Normal College, Hsinchu, Taiwan, ROC*

Received 19 January 1994; revised 13 March 1995

Abstract

Using Lee–Low–Pines transformation and perturbative variational method, the effect of electron–phonon interaction on the exciton binding energy in a $Ga_{1-x}Al_xAs$ quantum well can be solved analytically. Our results show that the phonon effect on binding energy of heavy-hole exciton is always larger than that of light-hole exciton and the correction of polaron effect on the exciton binding energy in a quantum well cannot be neglected.

1. Introduction

Recently the growth of alternate layers of controllable different semiconductors and relatively sharp interfaces makes the experimental studies of excitonic states possible. The exciton in the quantum well behaves like a quasi-two-dimensional hydrogen atom. The exciton binding energies in a quantum well have been studied extensively in recent years. The binding energy for an exciton is typically 100 to 1000 times smaller than that of a hydrogen atom. In the previous works, the hole is usually treated as a particle with either the heavy-hole mass or the light-hole mass. Most of the previous calculations employed the variational approach [1–4]. Some works [5, 6] included the nonparabolicity of the conduction band, and the degeneracy of the valence band. It is known that an electron staying in a low-lying level of conduction band of a polar crystal will interact strongly with the longitudinal optical mode of lattice vibrations [7–12]. On the basis of the strong coupling scheme, Ercelebi and Özdincer [8] calculated the ground-state binding energy of the exciton–phonon system in GaAs/GaAlAs quantum-well structures and found that the corrections due to electron–phonon coupling are rather significant. Degani and Hipólito [9] also found the polaronic contribution to the exciton binding energies is quite significant and increases with decreasing well thickness. Rücker et al. [12] calculated the electron–LO-phonon scattering rate in quasi-two-dimensional systems, based on a fully microscopic description of the phonon spectra and concluded that interface phonons are of great importance. In this work, we concentrate on the effect of electron–phonon coupling on the binding energy of Wannier excitons in quantum-well structures. A perturbative variation technique [13] is employed to construct a trial wave function. The interaction between the electron and surface phonon and the electron and bulk longitudinal phonon are taken into account. Lee–Low–Pines transformation is applied to separate the electron and phonon variables.

* Corresponding author.

2. Theory

The Hamiltonian of an exciton in a GaAs slab sandwiched between two semi-infinite slabs of GaAs–$Ga_{1-x}Al_xAs$ interacting with the longitudinal optical phonon can be expressed as

$$H = H_e + H_h + H_{e\text{-}h} + H_{sp} + H_{sp\text{-}e} + H_{sp\text{-}h} + H_{bp} + H_{bp\text{-}e} + H_{bp\text{-}e} + H_{bp\text{-}h}, \quad (1)$$

where

$$H_e = -\frac{\hbar^2}{2m_e}\nabla_e^2 + V_{ew},$$

$$H_h = -\frac{\hbar^2}{2m_h}\nabla_h^2 + V_{hw},$$

$$H_{e\text{-}h} = \frac{-e^2}{\varepsilon_0|\boldsymbol{r}_e - \boldsymbol{r}_h|},$$

$$H_{sp} = \sum_q \hbar\omega_s a_q^\dagger a_q,$$

$$H_{sp\text{-}e} = \sum_q (\Gamma_q[\exp(-q_z|z_e - L/2|) + \exp(-q_z|z_e - L/2|)]e^{i\boldsymbol{q}_r\cdot\boldsymbol{r}_{e//}}a_q + \text{h.c.}),$$

$$H_{sp\text{-}h} = \sum_q (\Gamma_q[\exp(-q_z|z_h - L/2|) + \exp(-q_z|z_h + L/2|)]e^{i\boldsymbol{q}_r\cdot\boldsymbol{r}_{h//}}a_q + \text{h.c.}),$$

$$H_{bp} = \sum_k \hbar\omega_b b_k^\dagger b_k,$$

$$H_{bp\text{-}e} = \sum_k W_{ek}\theta(z_e)\cos k_z z_e e^{i\boldsymbol{k}_r\cdot\boldsymbol{r}_{e//}}b_k + \text{h.c.},$$

$$H_{bp\text{-}h} = \sum_k W_{hk}\theta(z_h)\cos k_z z_h e^{i\boldsymbol{k}_r\cdot\boldsymbol{r}_{h//}}b_k + \text{h.c.},$$

where $V_{ew}(z_e)$ and $V_{hw}(z_h)$ are the well potentials seen by the electron and hole; a_q is the annihilation operator for optical surface (SO) phonons of wave vector $\boldsymbol{q} = (\boldsymbol{q}_r, \boldsymbol{q}_z)$ and frequency ω_s; b_k is the annihilation operator for the optical bulk (BO) phonons of wave vector $\boldsymbol{k} = (\boldsymbol{k}_r, \boldsymbol{q}_z)$ and frequency ω_j; H_{sp} is the surface phonon energy; $H_{sp\text{-}e}(H_{sp\text{-}h})$ is the interacting Hamiltonian between the surface phonon and the electron (hole) in the well. H_{bp} is the bulk phonon energy, and $H_{bp\text{-}e}$ ($H_{bp\text{-}h}$) is the interacting Hamiltonian between the bulk phonon and the electron (hole) in the well. The interaction strengths are

$$\Gamma_q = i\left(\frac{\pi\hbar\omega_s e^2}{\varepsilon^* Aq}\right)^{1/2},$$

$$\varepsilon^* = \left(\frac{\varepsilon_0 - 1}{\varepsilon_0 + 1} - \frac{\varepsilon_\infty - 1}{\varepsilon_\infty + 1}\right)^{-1},$$

$$\theta(z) = \begin{cases} 0, & z > L/2 \text{ or } z < -L/2, \\ 1, & -L/2 < z < L/2, \end{cases}$$

$$W_{jk} = \frac{1}{k}\frac{U^j}{\sqrt{V}}, \quad V\text{: crystal volume},$$

$$U^j = \hbar\omega_j\sqrt{4\pi\eta_j\left(\frac{\hbar}{2m_j\omega_j}\right)^{1/2}},$$

$$\eta_j = e^2\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)\sqrt{\frac{m_j}{2\hbar^3\omega_j}},$$

$$j = e, h,$$

where ε_0 and ε_∞ are the static and high-frequency dielectric constants, ω_s and ω_j are the dispersionless SO and BO phonon energies.

Now applying the first Lee–Low–Pines transformation [14], one obtains

$$\begin{aligned} H^0 = {} & H_e + H_h + H_{e\text{-}h} \\ & + \sum_q\left(\hbar\omega_s + \frac{\hbar^2q^2}{2m_e} + \frac{\hbar^2q^2}{2m_h}\right)(a_q^\dagger + f_q^*)(a_q + f_q) \\ & + \sum_q \Gamma_q e^{-q|z_e|}(a_q + f_q) + \text{h.c.} \\ & + \sum_q \Gamma_q e^{-q|z_h|}(a_q + f_q) + \text{h.c.} \\ & + \sum_k\left(\hbar\omega_b + \frac{\hbar^2k_r^2}{2m_e} + \frac{\hbar^2k_r^2}{2m_h}\right)(b_k^\dagger + g_k^*)(b_k + g_k) \\ & + \sum_k W_{ek}\theta(z_e)\cos k_z z_e(b_k + g_k) + \text{h.c.} \\ & + \sum_k W_{hk}\theta(z_h)\cos k_z z_h(b_k + g_k) + \text{h.c.} \end{aligned} \quad (2)$$

In the above derivation, we have neglected the terms involving the virtual phonon–electron and virtual phonon–hole interaction. Now assuming the trial wave function to be a product of exciton part and phonon part, we have

$$|\Psi\rangle = \phi(r)|\psi\rangle \quad (3)$$

where $\phi(r)$ depends on the exciton coordinates and $|\psi\rangle$ depends on the phonon coordinates. For lower-lying polaron states, $|\psi\rangle$ is taken as the phonon vacuum state $|0\rangle$. Now applying second Lee–Low–Pines transformation [14], one obtains finally

$$\langle H^0\rangle = \langle\phi(r)|H_e + H_h + H_{e\text{-}h}|\phi(r)\rangle - \sum_q \frac{|\Gamma_q|^2|\beta_{eq}+\beta_{hq}|^2}{\hbar\omega_s + \hbar^2q^2/2m_e + \hbar^2q^2/2m_h} - \sum_k \frac{|W_{ek}\alpha_{ek} + W_{hk}\alpha_{hk}|^2}{\hbar\omega_b + \hbar^2k_r^2/2m_e + \hbar^2k_r^2/2m_h}, \quad (4)$$

where

$$\beta_{eq} = \int_{-L/2\le z_e,\, z_h\le L/2} [\exp(-q|z_e - L/2|) + \exp(-q|z_e + L/2|)] \times |\phi(r)|^2\, dx_e\, dy_e\, dz_e\, dx_h\, dy_h\, dz_h,$$

$$\beta_{hq} = \int_{-L/2\le z_e,\, z_h\le L/2} [\exp(-q|z_h - L/2|) + \exp(-q|z_h + L/2|)] \times |\phi(r)|^2\, dx_e\, dy_e\, dz_e\, dx_h\, dy_h\, dz_h,$$

$$\alpha_{ek} = \int_{-L/2\le z_e,\, z_h\le L/2} \theta(z_e)\cos k_z z_e\, |\phi(r)|^2\, dx_e\, dy_e\, dz_e\, dx_h\, dy_h\, dz_h,$$

$$\alpha_{hk} = \int_{-L/2\le z_e,\, z_h\le L/2} \theta(z_h)\cos k_z z_h\, |\phi(r)|^2\, dx_e\, dy_e\, dz_e\, dx_h\, dy_h\, dz_h.$$

For excitonic part, we employ the perturbative and variational method by first adding a term $\lambda e^2/(\varepsilon_0\sqrt{x^2+y^2})$ to and then substracting the same term from the hamiltonian and rearranged as

$$H = H_{z_e} + H_{z_h} + H_{xy} + H'(\lambda) = H_0(\lambda) + H'(\lambda), \quad (5)$$

where

$$H_{z_e} = \frac{-\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2} + V_{ew}(z_e),$$

$$H_{z_h} = \frac{-\hbar^2}{2m_h}\frac{\partial^2}{\partial z_h^2} + V_{hw}(z_h),$$

$$H_{xy} = \frac{-\hbar^2}{2\mu_\pm}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) - \frac{\lambda e^2}{\varepsilon_0\sqrt{x^2+y^2}},$$

$$H'(\lambda) = \frac{\lambda e^2}{\varepsilon_0\sqrt{x^2+y^2}} - \frac{e^2}{\varepsilon_0|\boldsymbol{r}_e - \boldsymbol{r}_h|},$$

and the height of the potential well seen by the electron (hole) is assumed to be V_e (V_h). The Hamiltonian H_0 can be solved exactly while the Hamiltonian $H'(\lambda)$ is treated as a perturbation with λ as a variational parameter. Up to the first-order energy correction, the total energy can be expressed as

$$E_g = E_g^{(0)}(\lambda) + \Delta E_g^{(1)}(\lambda) = 4\lambda_0^2 R + \frac{16\lambda^2e^2}{a^2\varepsilon_0^2}\int_{-\infty}^{\infty} dz_e\, |f_e(z_e)|^2 \int_{-\infty}^{\infty} dz_h\, |f_h(z_h)|^2 \times\left\{\frac{a}{4} + |z_e - z_h| - \frac{\pi}{2}|z_e - z_h| \times\left[H_1\left(\frac{4\lambda|z_e - z_h|}{a}\right) - N_1\left(\frac{4\lambda|z_e - z_h|}{a}\right)\right]\right\}, \quad (6)$$

where $H_1(x)$ and $N_1(x)$ are the Struve and Neumann functions of order 1 [15]. The parameter λ can be obtained by requiring $\partial E_g/\partial\lambda = 0$ and the ground state energies E_g is then obtained. The binding energy of exciton is defined as $-E_g$.

3. Results and discussion

We have calculated the binding energies of the heavy-hole (E_{BH}) and the light-hole (E_{BL}) excitations of GaAs quantum wells for different Al concentrations $x = 0.15$ and 0.3 as a function of the well width L. The physical parameters are adopted from the previous works [16, 17]. The reduced masses in the x–y plane for the heavy hole ($J = 3/2$) and the light hole ($J = 1/2$) excitons are taken as $0.04m_0$ and $0.051m_0$ respectively.

Fig. 1 shows the comparison of the binding energies of the light-hole and heavy-hole excitons with and without the electron–phonon interaction for different well widths. One can see from Fig. 1 that

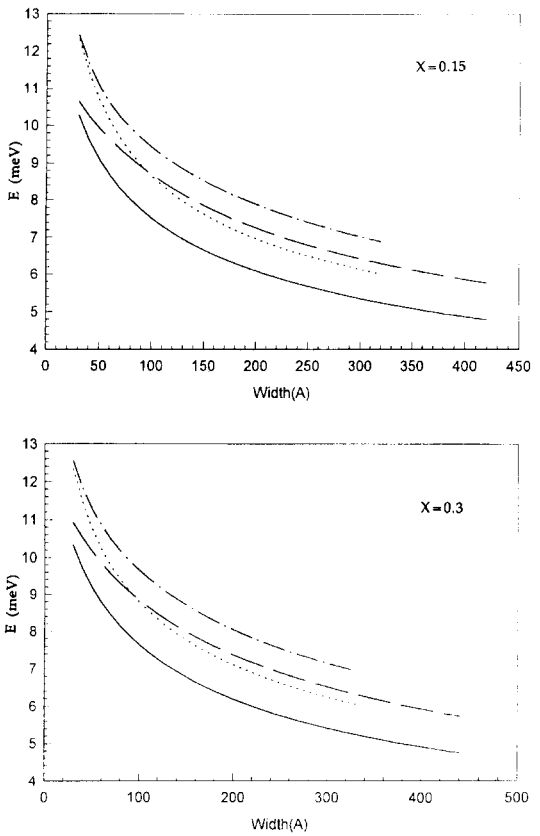

Fig. 1. The comparison of binding energies of heavy-hole exciton and light-hole exciton with and without the polaron effect for Al concentration $x = 0.15$ and 0.3. Solid (dotted) line is the binding energy of heavy- (light-) hole exciton. Broken (dotted and broken) line is the binding energy of heavy- (light-) hole exciton with surface and bulk phonon effects.

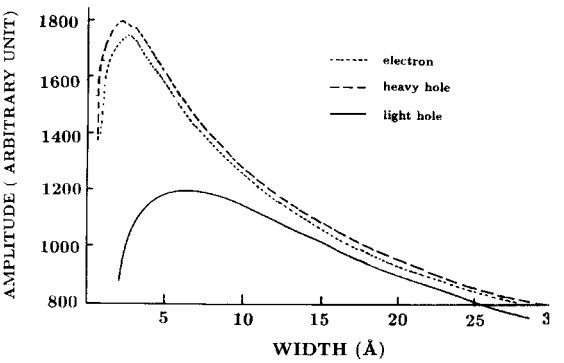

Fig. 2. The variation of amplitudes of wave functions of electron, heavy-hole exciton and light-hole exciton as a function of well width.

the polaron effect cannot yield pronounced difference for different Al concentrations x. The reason that the exciton binding energies in quantum wells do not depend significantly on the barrier height may be explained as follows. In our calculation, the effective masses of the electron and hole in $Ga_{1-x}Al_xAs$ quantum wells were assumed as $m_e = (0.067 + 0.083x)m_0$, $m_+ = (0.45 + 0.2x)m_0$, and $m_- = (0.082 + 0.068x)m_0$, where m_0 is the free-electron mass. These effective masses do not yield pronounced difference for different Al concentrations, which is why our calculated binding energies cannot yield significant difference as x varies from 0.15 to 0.3. From Fig. 1 one can also note that for a given value of x, the effect of electron–phonon interaction on either light-hole or heavy-hole excitons are significant, especially for larger well width. However, the total binding energy of light-hole exciton including the polaron effect is always larger than that of heavy-hole exciton. Fig. 2 shows the variation of the amplitude of wave function as a function of well width for electron, heavy-hole exciton and light-hole exciton. One can see that the amplitudes decrease with increasing well width. However, for small well width the wave functions for electron or exciton decrease rapidly as well width approaches zero. This is because the leakage of the electron happens as the width becomes very small. Figs. 3 and 4 show the influence of the surface and bulk phonon on the heavy-hole and the light-hole exciton binding energies for Al concentration $x = 0.15$ and 0.3, respectively. Here, the influence is defined as $(E^* - E_0)/E^*$, where E^* and E_0 are the exciton binding energies with and without the polaron effect. One can see from Figs. 3 and 4 that the Al concentrations cannot yield significantly different phonon influence on exciton binding energies. Fig. 3 also shows that the surface phonon plays more important role as the well width ranges from 80 Å to 100 Å. One may also note that the influence of the surface phonon on the binding energy of exciton is small for small well width. The influence becomes important as the well width ranges from 80 Å to 100 Å and then decreases again as the well width becomes larger. For

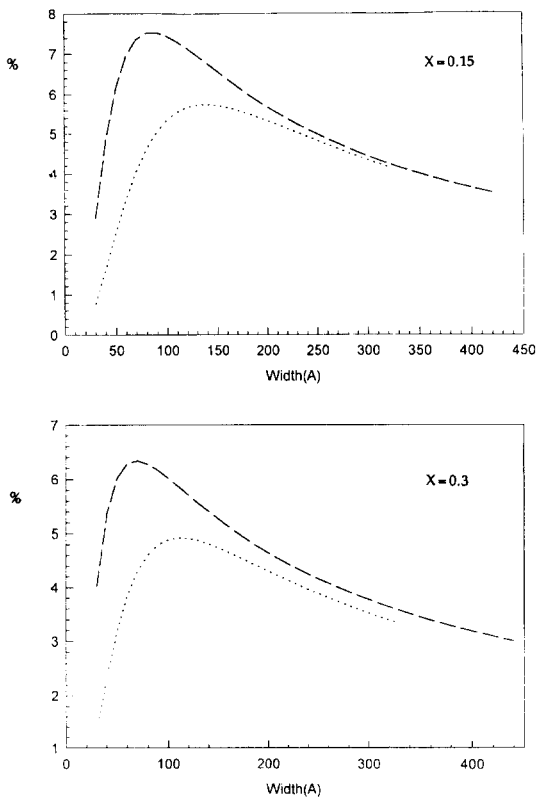

Fig. 3. The variation of percentage of surface phonon effect on heavy-hole-exciton binding energy (broken line) and light-hole exciton binding energy (dotted line) as a function of the well width L for concentration $x = 0.15$ and 0.3.

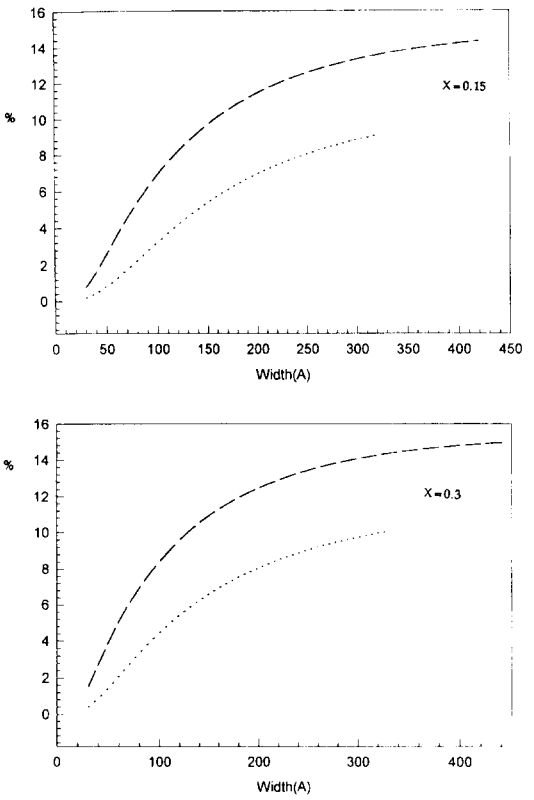

Fig. 4. The variation of bulk phonon effect on heavy-hole-exciton binding energy (broken line) and light-hole-exciton binding energy (dotted line) as a function of the well width L for concentration $x = 0.15$ and 0.3.

larger well width, the surface phonon effect on either the heavy-hole or light-hole exciton becomes almost the same. The reason for the small influence of the surface phonon in narrow quantum well can be explained as follows. For quantum well with finite barrier height, the leakage of wave function out of the well region is much larger for very narrow well width. This means that there is a larger probability for finding the exciton outside the quantum well and thus makes the interaction between the electron and the surface phonon less pronounced. One can also note that our calculated exciton binding energies do not decrease with the well thickness for very narrow quantum wells as was obtained already by previous works. We can explain this fact as follows. Since in our work we employed a perturbative variational technique to calculate the binding energy of the exciton, the perturbation term $H'(\lambda)$ contained in Eq. (3) becomes so important that the approximation is no longer valid for very narrow quantum wells. Therefore, in our calculation the exciton binding energies are not able to be reproduced correctly for quantum well with thickness less than 30 Å in Fig. 1. Fig. 4 shows that the bulk phonon effect on the binding energy of either light-hole or heavy-hole exciton is small for very narrow quantum well. However, the bulk phonon influence on the exciton binding energy becomes significant as the well width becomes larger and reaches a saturated value of about 14% for very wide quantum well. One can also see from Figs. 3 and 4 that the influence of either the surface phonon or bulk phonon on the heavy-hole exciton are always larger than that on

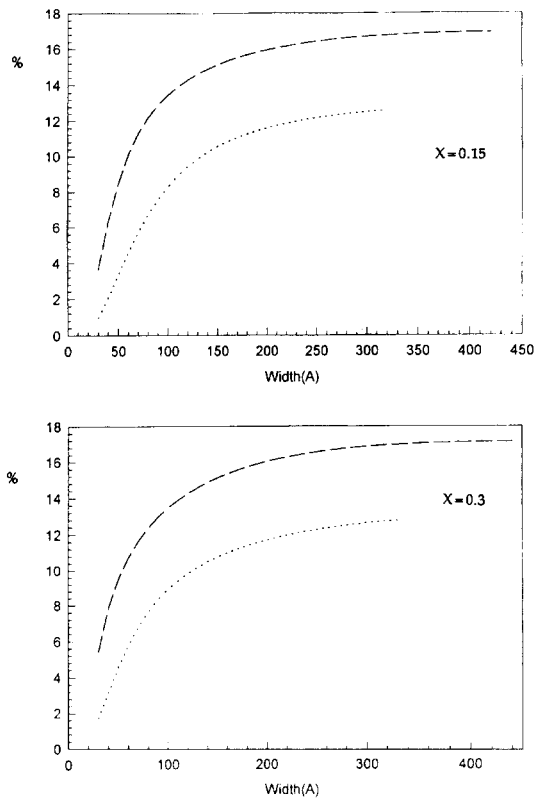

Fig. 5. The variation of total phonon effect on heavy-hole-exciton binding energy (broken line) and light-hole-exciton binding energy (dotted line) as a function of the well width L for concentration $x = 0.15$ and 0.3.

the light-hole exciton, especially for the influence of surface phonon for very small well width. This is because the effective mass of the heavy hole along z-direction is heavier than that of the light hole, therefore, the heavy hole is bounded more tightly than light hole and yields a smaller interaction range. This in turn yields a larger effect on a small position for the surface phonon. The same reason can be applied to the bulk phonon case. Furthermore, although the corrections due to the electron–phonon interaction for the heavy- and light-hole excitons differ only slightly in our calculation, the binding energies of light-hole excitons were found to be always larger than those of heavy-hole excitons. This makes the influence of the electron–phonon interaction on the heavy-hole exciton to be always larger. Fig. 5 shows the total influence of the surface phonon and bulk on the binding energies of the light-hole and heavy-hole excitons. The comparison of Figs. 1 and 5 shows that although the total binding energy of light-hole exciton including the surface and bulk phonon effect is larger than that of the heavy-hole exciton, the polaron effect on the heavy-hole exciton plays more important role than that of light-hole exciton. Fig. 5 also shows that the total phonon effect (including the surface and bulk phonon effects) increases with increasing well width. Our calculated percentage of phonon effects reaches a saturation value of about 17.5% for heavy-hole exciton and 13.5% for light-hole exciton. Ercelebi [8] studied the electron (hole)-lattice interaction in quantum well and obtained more than 30% phonon effect as the well width becomes larger than 150 Å. Their results seem to be overestimated. Degani and Hipólito [9] studied the phonon effect on the binding energy of exciton and obtained a result of 26% to 20% phonon effect as the well width ranges form 10 Å to 150 Å. Rogers et al. [18] reported the exciton binding energies from low magnetic field measurements. Their observations show that the binding energies are 10 and 8 meV for heavy-hole excitons and are 11 and 9 meV for light-hole excitons for quantum wells with thickness of 75 Å and 100 Å, respectively. Table 1 lists the comparison of some available experimental data and the theoretical results of Ercelebi et al. [8] and ours. The values in the parenthesis do not include the electron–phonon effect. One can see from the table that our calculated results for both heavy and light excitons seem to be more reliable while comparing with the results of Ercelebi et al.

4. Summary

In summary, we have studied the optical surface and bulk phonon effects of the exciton binding energy. The results show that the polaronic effects are important and cannot be neglected. Our results manifest that the correction due to polaron effect on the binding energy of heavy-hole exciton is larger than that on the light-hole exciton.

Table 1
Comparison of binding energies (meV) of heavy-hole and light-hole excitons

Width (Å)	x	Hole level	Experimental data (meV)	Our results (meV)	Ref. [8]
75	0.4	H_1	10.5–11.5 (Ref. [19])	9.56 (8.34)	
75	0.4	L_1	11.3–12.3 (Ref. [19])	10.45 (9.75)	
75	0.35	H_1	10–12 (Ref. [18]), 9.3 (Ref. [6])	9.43 (8.23)	
75	0.35	L_1	11 (Ref. [18]), 11 (Ref. [6])	10.31 (9.62)	
80	0.15	H_1		9.12 (8.01)	10.60 (9.11)
80	0.15	L_1		9.92 (9.26)	14.19 (10.73)
80	0.30	H_1		9.34 (8.14)	10.60 (9.11)
80	0.30	L_1		10.17 (9.39)	14.19 (10.73)
92	0.35	H_1	9.5–10.5 (Ref. [19])	9.01 (7.82)	
92	0.35	L_1	11.2–12.2 (Ref. [19])	9.80 (9.14)	
100	0.35	H_1	13 (Ref. [20])	8.87 (7.60)	
100	0.35	L_1	10 (Ref. [19])	9.63 (8.91)	
110	0.35	H_1	8–9.5 (Ref. [18]), 8.4 (Ref. [6])	8.69 (7.43)	
110	0.35	L_1	9 (Ref. [19]), 11 (Ref. [6])	9.42 (8.68)	
112	0.3	H_1	12 (Ref. [21])	8.56 (7.29)	
112	0.3	L_1		9.31 (8.56)	
120	0.15	H_1		8.31 (7.13)	9.65 (8.06)
120	0.15	L_1		9.01 (8.17)	13.01 (9.37)
120	0.30	H_1		8.46 (7.23)	9.65 (8.06)
120	0.30	L_1		9.23 (8.32)	13.01 (9.37)

Acknowledgements

This work is partially supported by the National Science Council of ROC.

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