Exciton–LO-phonon interaction in zinc-compound quantum wells

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The properties of optical phonons and excitons in four zinc-compound quantum wells, in which the well layers are made of ternary mixed crystals, are investigated theoretically. The optical phonon frequencies and the electron–LO-phonon coupling constants of the well materials are calculated by using a mixed crystal theory. The interface optical phonon energies and dispersion relations, the exciton binding energies and the exciton–LO-phonon interaction energies are calculated as functions of the well width and the concentration x. The effects of the mixed crystal on the exciton–optical-phonon interaction in the quantum wells are discussed.

Exciton–LO-phonon interaction is considered very important in the electronic and optical properties of quantum wells (QW's). The investigation of the exciton–LO-phonon interaction in QW's has been attracting much attention and has been studied by many authors.[1–6] All the studies have given us much useful information about the properties of excitons in QW's, but there still exist substantial problems. For example, although most of the experiments have been carried out in II-VI and III-V QW's that have been composed of mixed crystals, the ''two-mode'' effect of the mixed crystals on the excitonic properties of the QW's has not been investigated theoretically up to now, to the best of our knowledge. Almost all the published theoretical works used the effective phonon approximation[7] for simplicity. In fact, the electric and optical properties of the mixed crystals are somewhat different from those of binary crystals, especially in the phenomena relating with the LO phonons in the QW's. It is necessary to investigate the effect of mixed crystals on the excitonic properties of the QW's in detail in view of recent extensive experimental studies on the light emitting mechanism of the II-VI wide gap compounds QW's.[8–15]

Very recently we have proposed a theory of electron–LO-phonon interaction in mixed crystals[16] and a polaron theory in QW's composed of mixed crystals.[17] In the present paper we extend our work to excitons in the QW's composed of mixed crystals and study the property of the exciton–LO-phonon interaction systems in the mixed crystal QW's. Some interesting features of the exciton-phonon system in mixed crystal QW's are found and discussed.

We consider a sandwich structure that involves layers of mixed materials. The z axis is chosen perpendicular to the heterointerfaces. The well layer (denoted as 1) is in the region of $|z|\leqslant d$, and is made of an $A_xB_{1-x}C$ type ternary mixed crystal. The barrier layer (denoted as 2) is in the region of $|z|>d$ and is made of a binary semiconductor. This kind of QW structure is noted as binary/ternary/binary (BTB) QW's. The Hamiltonian of an exciton in the well interacting with optical phonons is expressed as[6,17]

$$H=\sum_{l=e,h}\left\{\frac{P_{l\|}^2}{2m_{l\|}}+\frac{P_{lz}^2}{2m_{lz}}+V_l(z_l)\right\}+V_{eh}(\rho,z)+\sum_{\beta}\sum_{\mathbf{k}}\hbar\omega_\beta(\mathbf{k})a_{\mathbf{k}\beta}^{\dagger}a_{\mathbf{k}\beta}+\sum_{\beta}\sum_{\mathbf{k}}\{V_{k\beta}[L_{k\beta}(z_e)e^{i\mathbf{k}\cdot\boldsymbol{\rho}_e}-L_{k\beta}(z_h)e^{i\mathbf{k}\cdot\boldsymbol{\rho}_h}]a_{\mathbf{k}\beta}+\text{H.c.}\}. \quad (1)$$

Where $l=e,h$ refer to the electron and hole, respectively. $\mathbf{P}_l=(\mathbf{P}_{l\|},P_{lz})$, $\mathbf{r}_l=(\boldsymbol{\rho}_l,z_l)$, and $m_l=(m_{l\|},m_{lz})$ are the momentum, position, and effective band mass of the particles. $\mathbf{P}_{l\|}$ and $\boldsymbol{\rho}_l$ are two-dimensional vectors in the x-y plane. $V_l(z_l)$ is the QW potential experienced by the electron or the hole. The finite square QW model is used in present paper. $V_{eh}(\rho,z)$ is the Coulomb potential between the electron and the hole. The width of the QW is W $(=2d)$. $\mathbf{r}=\mathbf{r}_e-\mathbf{r}_h=(\boldsymbol{\rho},z)$ is the relative position between the electron and the hole. In Eq. (1) $a_{\mathbf{k}\beta}^{\dagger}(a_{\mathbf{k}\beta})$ is the creation (annihilation) operator of an optical phonon with frequency $\omega_\beta(\mathbf{k})$ and wave vector $(\mathbf{k},k_\beta)$. From our previous study (Ref. 17) on polarons in mixed crystal QW's, it is known that in a BTB QW there are two slab-LO-phonon modes in the well layer, one half-space LO phonon mode in the barrier layer, and six interface optical (IO) phonon modes near the interfaces of the QW. The expressions of $\omega_\beta(\mathbf{k})$, $V_{k\beta}$, and $L_{k\beta}$ have been given in Ref. 17.

We use the same variational method as Ref. 6 to study the excitonic properties of some zinc-compound QW's that have been extensively studied for the green-blue light emitting diodes and lasers.[8–15,18] In the present paper $Zn_{1-x}Cd_xSe/ZnSe$, $Zn_{1-x}Cd_xS/ZnS$, $Zn_{1-x}Cd_xTe/ZnTe$ and $ZnS_{1-x}Se_x/ZnS$ QW's are put into numerical calculation.

TABLE I. Physical parameters used in the calculations. All data are taken from Ref. 20. ω_L and ω_T are in units of meV. m_e and m_h are in units of the free electron mass. a is the lattice constant in units of Å. E_g is the energy band gap in units of eV.

	ϵ_0	ϵ_∞	ω_L	ω_T	m_e	m_h	a	E_g
ZnSe	7.6	5.4	31.4	26.4	0.13	0.57	5.668	2.82
ZnTe	9.67	7.28	25.6	21.9	0.13	0.6	6.10	2.39
ZnS	8.0	5.1	43.2	34.3	0.34	1.76	5.410	3.78
CdSe	10.16	6.2	25.9	20.5	0.12	0.45	6.052	1.83
CdTe	10.2	7.1	21.0	17.4	0.09	0.72	6.482	1.61
CdS	8.45	5.32	37.3	30.0	0.14	0.51	5.825	2.57

The physical parameters used in this paper are listed in Table I. The lattice constants, the band gaps, and the electron and hole band masses of the mixed crystals are estimated by the linear interpolation scheme due to the lack of definite knowledge. The ratios of the conduction-band offset to the valence-band offset are assumed as 0.8:0.2, 0.7:0.3, 0.7:0.3, and 0.2:0.8 for the $Zn_{1-x}Cd_xSe/ZnSe$, $Zn_{1-x}Cd_xS/ZnS$, $Zn_{1-x}Cd_xTe/ZnTe$, and $ZnS_{1-x}Se_x/ZnS$ QW's, respectively. We have calculated the same problem with different barrier parameters and found that excitonic properties of the QW's are not sensitive to the barrier parameters if the well width is not too narrow and the parameters are in a reasonable range.

In order to give a complete understanding of the mixed crystal effects, we show at first the properties of the mixed crystals in the wells of the QW's based on our theoretical calculations.[16] The optical-phonon frequencies of the mixed crystals are plotted in Fig. 1. The results show that there are two LO and two TO phonon frequencies in the mixed crystals theoretically. The strength of each LO-phonon mode interacting with carriers is reflected in the Fröhlich electron-phonon coupling constant, which is defined as[17]

$$\alpha_{Lj}=\left(\frac{m_e e^4}{2\hbar^3\omega_{Lj}\bar{\epsilon}_{1j}^2(\omega_{Lj})}\right)^{1/2}, \quad j=1a,1b. \tag{2}$$

The Fröhlich coupling constants of the mixed crystals are calculated and the results are plotted in Fig. 2. This figure shows clearly the dependence of the coupling constants with the concentration x. In fact, if one wants to make a judgment on whether a mixed crystal is a ''two mode'' or ''one mode'' mixed crystal, the best way is to see the variation of the Fröhlich coupling constants with the concentration x. Our results show that the $ZnS_{1-x}Se_x$ crystal is a two-mode mixed crystal and the others are one-mode mixed crystals. In the one-mode mixed crystal the electron–LO-phonon interaction is contributed mainly from the La phonon mode. For the $Zn_{1-x}Cd_xS$ crystal the contribution from the Lb mode is very small and can be neglected. But for the $Zn_{1-x}Cd_xSe$ and $Zn_{1-x}Cd_xTe$ crystals the Lb mode phonons give obvious contributions to the polaronic effects in a part of the composition range, despite that these two crystals are one-mode mixed crystals.

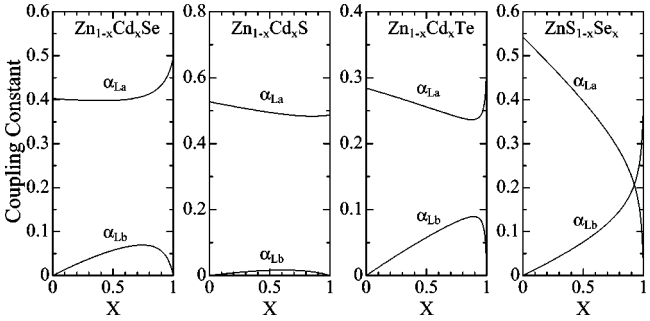

FIG. 2. Concentration dependence of the electron–LO-phonon coupling constants.

The mixed crystal effect changes completely the feature of the IO phonon in the mixed crystal QW's. Figure 3 displays the dependence of the IO-phonon energy with the concentration x in the case of the wave vector $k\to 0$. All of the six IO-phonon modes are plotted in the figure and show very different features. In order to show the importance of each IO-phonon mode, we have calculated the electron–IO-phonon coupling strength $\alpha_{Ipj}(k)$, which is defined similar to α_{Lj} by replacing ω_{Lj} and ϵ_{1j} with IO-phonon frequency and corresponding effective dielectric constant, respectively. Since the IO-phonon energies disperse with the wave vector k, the coupling strength $\alpha_{Ipj}(k)$ $(p=+,-;\ j=1,2,3.)$ also has dispersion relation with k. The dependence of $\alpha_{Ipj}(k)$ with the concentration x and k reflects the feature of the electron–IO-phonon interaction in the QW's. Figure 4 displays the dependence of the electron–IO-phonon coupling strength with the wave vector k in the case of $x=0.8$. Because only the long-wavelength optical phonons have a large effect on the changed particles we can presume only the ω_{+3} mode gives a large contribution to the polaronic effect of the QW's. From Fig. 3 we see that the phonon energy of the ω_{+3} mode in the case of $k\to 0$ is equal to the LO-phonon energy of barrier material and is independent of the concentration x. This interesting feature which resulted from the

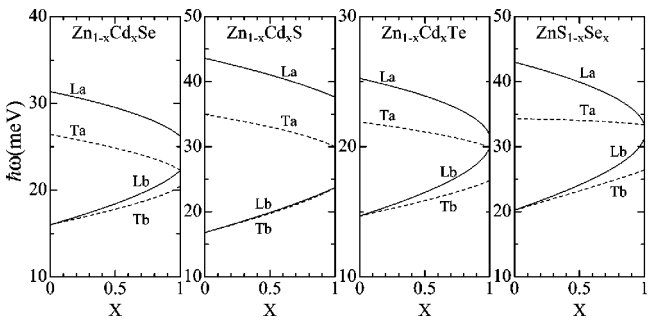

FIG. 1. Concentration dependence of transverse (dashed line) and longitudinal (full line) optical phonon energies.

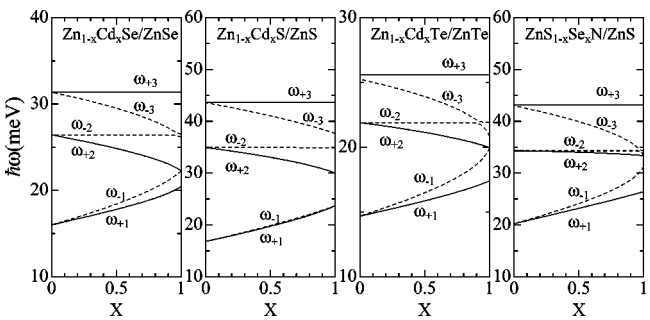

FIG. 3. Concentration dependence of IO-phonon energy ($k=0$).

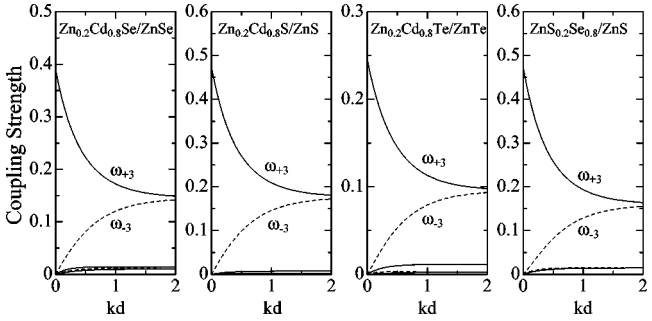

FIG. 4. Dispersion relation of the electron–IO-phonon coupling strength.

present theory is common for any BTB type QW. It should be noticed that, although $\omega_{+3}(k\rightarrow 0)=\omega_{L2}$ is held, the property of the ω_{+3} IO phonon is different essentially from the ω_{L2} bulk phonon mode. The origin and the location as well as the dispersion relation of the ω_{+3} mode are different from the LO phonon in the barrier. One should be careful in the analysis of the phonon spectrum of the QW's.

Since the ground state and the lowest excited state are important in experiments, we give theoretical calculations of the 1*S* and 2*S* excitons in the present section. In these calculations the subband states of the electron and the hole are assumed in the lowest subband states. Figure 5 shows that in all cases 1*S*-exciton binding energies are in the range of 17–46 meV for wide wells of 200 Å, and approach gradually the values of bulk excitons with increasing the width further. Even if the width is much larger than the exciton Bohr radii (30 Å–54 Å), a small width dependence of the binding energies is found. This fact is due to the quantum confinement that has effect on the excitons even when the well width is ten times larger than the exciton radii.[19] It is noticed that the 1*S*-exciton binding energies have maximum values when the well width is about 20 Å–30 Å, the larger the band offset the smaller the well width where the binding energy is maximum. However, the 2*S*-exciton binding energies do not show obvious variation with the well width and almost remain constants in the QW's. Since the published data of carrier masses and exciton binding energies for zinc-compound quantum wells are very different, it seems too difficult to present a theory that gives results in agreement with all experiments at present.

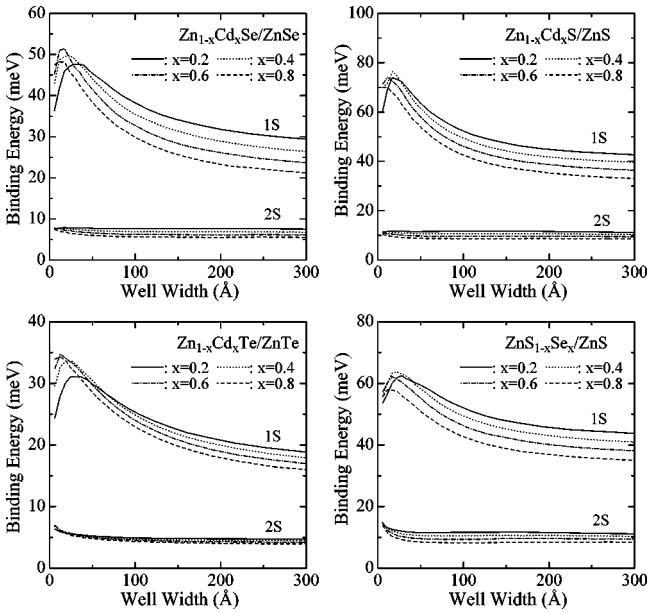

FIG. 5. Exciton binding energy as a function of the quantum well width.

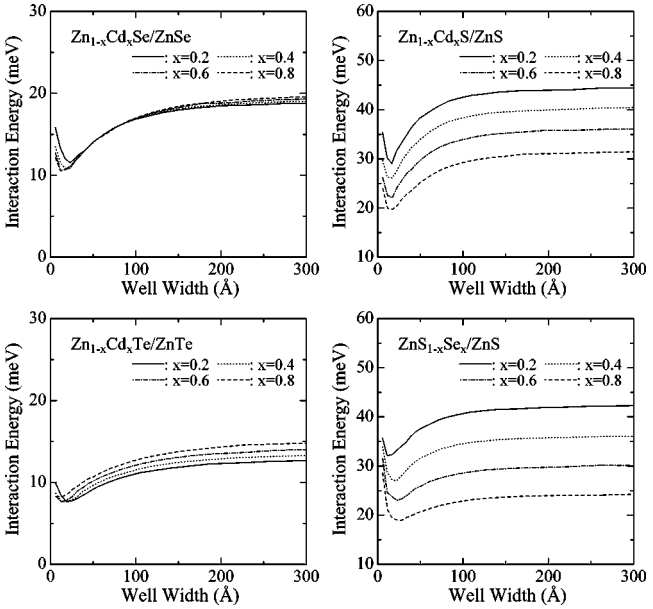

FIG. 6. The total interaction energy of the 1*s* exciton with LO phonons as a function of the well width.

The 1*S*-exciton–optical-phonon interaction energies in the zinc-compound QW's (with $x=0.2$, 0.4, 0.6, and 0.8) as a function of the well width are shown in Fig. 6. From this figure we understand that the variations of the interaction energies with the concentration x are very different for the QW's. The common feature is that the interaction energies are decreased with decreasing the well width, and have minimum values when the well width in the range of 20 Å–30 Å if x is not too small. In the exciton-related lasing mechanism in zinc-compound QW's,[10] it is essentially important for the exciton stability that the exciton binding energy is larger than the LO-phonon energy. From Fig. 5 we understand that the exciton state will be the most stable state if the well widths

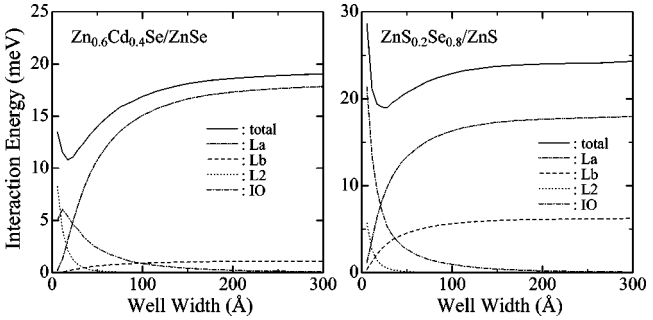

FIG. 7. Electron–LO-phonon interaction energy as a function of the well width. The solid line corresponds to the total polaron energy. The dash-double-dotted line represents the contribution from the $j=1a$ slab-LO-phonon mode, the dashed line from the $j=1b$ slab-LO-phonon mode, the dotted line from the half-space LO-phonon mode, and the dash-dotted line from the IO-phonon mode.

of the zinc-compound QW's are fabricated in the range of 20 Å–30 Å. We think this result is helpful for the design of the laser structures of the zinc-compound QW's.

In order to investigate the importance of every phonon mode in the exciton-phonon interaction, we have calculated the contribution to the total interaction energy from every phonon mode, two typical results are plotted in Fig. 7. It is seen that the half-space LO phonons and the IO phonons have effects on the exciton only in the very narrow well case. With increasing the well width the contribution of the slab-LO-phonon to the interaction energy increases and goes to the bulk value in the limit of $r \rightarrow \infty$. The importance of the slab-LO-phonon mode is determined by the concentration x. This property can be seen clearly from Figs. 2 and 7. Since the well material of the $ZnS_{1-x}Se_x/ZnS$ QW is a ''two-mode'' mixed crystal, the contribution of the $L1a$ mode is decreased from its maximum value to zero with increasing the concentration x from 0 to 1. At the same time, the $L1b$ is increased from zero to its maximum value. The two-mode behavior of the QW's composed of mixed crystal will appear in the optical experiments, such as the luminescence spectrum and the Raman spectrum.

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