Laboratory of Excited State Processes, Changchun Institute of Physics,
Academia Sinica, Changchun[1]*) (a) and*
Department of Physics, Inner Mongolia National Teacher's College, Tongliao[2]*) (b)*

The Influence of the Interaction between Phonons on the Properties of the Exciton in Polar Crystals

By

Wei Xiao (a, b) and Jing-Lin Xiao (a, b)

Some properties of excitons in polar crystals are studied. The effective Hamiltonian of the exciton in polar crystals is derived by a perturbation method. If we consider the interaction between phonons of different wave vectors in the recoil process, the influence on the effective potential between electron and hole, the self-trapping energy, and the self-trapping condition are discussed.

1. Introduction

Since Haken [1] studied excitons in polar crystals for the first time, the excitons in polar crystals have been of considerable interest. One reason is that it provides a significant example for two interacting fermions in a boson field. Another more serious one is that the obtained energies can be used to explain the optical spectra near the absorption edge of semiconductors. The main approaches applied to this problem can be divided into five types: variational calculations [2 to 4], perturbation theory [5, 6], combined variational and perturbational approaches [7, 8], Green's function methods [9, 10], and functional integration method [11, 12]. Gu et al. [13, 14], using a perturbation method, investigated the properties of intermediate-coupling excitons in a quantum well and of an exciton–phonon system in a polar semiconductor quantum well. Recently, Sumi and Sumi [15] discussed the self-trapping of excitons interacting with phonons via short-range potentials. The result shows that when excitons can move three-dimensionally, the self-trapping state appears suddenly as a strongly localized one when the coupling constant exceeds a certain critical value. However, this problem is still far away from an exact solution up to now. Chen et al. [16] calculated the ground state energy of the strongly bound exciton–phonon system by means of a concise variational approach. In fact, so far research of the exciton only was restricted to approximations and calculations where the interaction between phonons of different wave vectors in the recoil process is neglected. The properties of the surface polaron, which considers the corresponding interaction, have been discussed by the perturbation method by the present authors and coworkers [17].

The purpose of this paper is to explore the effect of the corresponding interaction on the properties of the exciton by using the perturbation method. The effective Hamilto-

---

[1]) Changchun 130021, People's Republic of China.
[2]) Tongliao 028043, People's Republic of China.

nian of the exciton in polar crystals is derived. If we consider the corresponding interaction, the influence on the effective interaction potential between the electron and the hole, the self-trapping energy, and the self-trapping condition of the exciton in polar crystals is discussed.

2. The Hamiltonian

The Hamiltonian H of the exciton–phonon system in polar crystals can be written as

$$
H = -\frac{\hbar^2}{2M} \nabla_{\mathbf{R}}^2 - \frac{\hbar^2}{2\mu} \nabla_{\mathbf{r}}^2 - \frac{e^2}{\varepsilon_\infty r} + \sum_{\mathbf{w}} \hbar\omega a_{\mathbf{w}}^+ a_{\mathbf{w}}
$$
$$
+ \sum_{\mathbf{w}} (V_{\mathbf{w}} a_{\mathbf{w}} \, \mathrm{e}^{i\mathbf{w}\cdot\mathbf{R}} \, \zeta_{\mathbf{w}} + \mathrm{h.c.}) , \tag{1a}
$$

$$
V_{\mathbf{w}} = \frac{4\pi i e}{\sqrt{V}} \left(\frac{c\hbar\omega}{8\pi\mathbf{w}^2} \right)^{1/2} , \tag{1b}
$$

$$
c = \frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} , \tag{1c}
$$

$$
\zeta_{\mathbf{w}} = \mathrm{e}^{-i\beta_1 \mathbf{w}\cdot\mathbf{r}} - \mathrm{e}^{i\beta_2 \mathbf{w}\cdot\mathbf{r}} , \tag{1d}
$$

$$
\beta_1 = \frac{m_e}{M} , \qquad \beta_2 = \frac{m_h}{M} , \qquad M = m_e + m_h , \qquad \mu = \frac{m_e m_h}{m_e + m_h} , \tag{1e}
$$

where M, μ, $\mathbf{R}$, $\mathbf{r}$ are the mass of the centre of mass, reduced mass, mass centre coordinate, and relative coordinate, respectively. m_e and m_h are the mass of the electron and the hole, β_1 and β_2 the fraction of the mass of the electron and hole, respectively, $a_{\mathbf{w}}^+$ and $a_{\mathbf{w}}$ are the creation and annihilation operators of the LO phonons with the wave vector $\mathbf{w}$, and $\hbar\omega$ is the LO-phonon energy. V is the normalization volume of the crystal, and the dynamical and static dielectric constants are labelled ε_∞ and ε_0, respectively.

As usual, we eliminate the centre-of-mass coordinate $\mathbf{R}$ from (1a) by the unitary transformation

$$
U_1 = \exp \left(i \left(\mathbf{K} - \sum_{\mathbf{w}} a_{\mathbf{w}}^+ a_{\mathbf{w}} \mathbf{w} \right) \cdot \mathbf{R} \right) , \tag{2a}
$$

where $\hbar\mathbf{K}$ is the total momentum of the system which is a constant of motion. The structure of the unitary operator U_2 is well known from the literature on coherent states. We have

$$
U_2 = \exp \left(\sum_{\mathbf{w}} a_{\mathbf{w}}^+ f_{\mathbf{w}} - a_{\mathbf{w}} f_{\mathbf{w}}^* \right) , \tag{2b}
$$

$$
f_{\mathbf{w}} = -\frac{2M}{\hbar^2} \frac{V_{\mathbf{w}} \zeta_{\mathbf{w}}}{\mathbf{w}^2 + u^2} , \tag{2c}
$$

$$
\frac{\hbar^2 u^2}{2M} = \hbar\omega . \tag{2d}
$$

$f_{\mathbf{w}}(f_{\mathbf{w}}^*)$ are the variational parameters. The transformed Hamiltonian can be rewritten as

$$
\mathscr{H} = U_2^{-1} U_1^{-1} H U_1 U_2
$$
$$
= \mathscr{H}_{\mathrm{ex}}^0 + \mathscr{H}_{\mathrm{ex}}^1 + \mathscr{H}_{\mathrm{ex}}^2 + \mathscr{H}_0 + \mathscr{H}_1 + \mathscr{H}_2 , \tag{3a}
$$

where

$$\mathscr{H}_{\text{ex}}^0 = -\frac{\hbar^2}{2\mu}\,\nabla_{\mathbf{r}}^2 - \frac{e^2}{\varepsilon_\infty r} + \frac{\hbar^2}{2\mu}\sum_{\mathbf{w}}\frac{\mathbf{w}^2}{(\mathbf{w}^2+u^2)^2}\,|A_{\mathbf{w}}|^2\,, \tag{3b}$$

$$\mathscr{H}_{\text{ex}}^1 = \frac{\hbar^2}{\mu}\sum_{\mathbf{w}}\frac{i\mathbf{w}\cdot\nabla_{\mathbf{r}}}{\mathbf{w}^2+u^2}\,(a_{\mathbf{w}}^+ A_{\mathbf{w}}^* + a_{\mathbf{w}} A_{\mathbf{w}})\,, \tag{3c}$$

$$\mathscr{H}_{\text{ex}}^2 = \frac{\hbar^2}{2\mu}\sum_{\mathbf{w}}\frac{\mathbf{w}^2}{(\mathbf{w}^2+u^2)^2}\,[a_{\mathbf{w}}^+ a_{\mathbf{w}}^+ (A_{\mathbf{w}}^*)^2 + a_{\mathbf{w}} a_{\mathbf{w}}(A_{\mathbf{w}})^2$$

$$+ 2a_{\mathbf{w}}^+ a_{\mathbf{w}}\,|A_{\mathbf{w}}|^2] + \frac{\hbar^2}{2\mu}\sum_{\mathbf{w}\neq\mathbf{w}'}\frac{\mathbf{w}\cdot\mathbf{w}'}{(\mathbf{w}^2+u^2)\,(\mathbf{w}'^2+u^2)}$$

$$\times (a_{\mathbf{w}}^+ A_{\mathbf{w}}^* + a_{\mathbf{w}} A_{\mathbf{w}})\,(a_{\mathbf{w}'}^+ A_{\mathbf{w}'}^* + a_{\mathbf{w}'} A_{\mathbf{w}'})\,, \tag{3d}$$

$$\mathscr{H}_0 = \frac{\hbar^2}{2M}\sum_{\mathbf{w}} a_{\mathbf{w}}^+ a_{\mathbf{w}}^+ (\mathbf{w}^2+u^2) + \frac{\hbar^2 K^2}{2M} - \frac{2M}{\hbar^2}\sum_{\mathbf{w}}\frac{|V_w|^2}{\mathbf{w}^2+u^2}\,, \tag{3e}$$

$$\mathscr{H}_1 = -\frac{\hbar^2}{M}\sum_{\mathbf{w}}(a_{\mathbf{w}}^+ a_{\mathbf{w}} + a_{\mathbf{w}}^+ f_{\mathbf{w}} + a_{\mathbf{w}} f_{\mathbf{w}}^* + f_{\mathbf{w}} f_{\mathbf{w}}^*)\,\mathbf{K}\cdot\mathbf{w}\,, \tag{3f}$$

$$\mathscr{H}_2 = \frac{\hbar^2}{2M}\sum_{\mathbf{w}\neq\mathbf{w}'}\mathbf{w}\cdot\mathbf{w}'(a_{\mathbf{w}}^+ a_{\mathbf{w}'}^+ a_{\mathbf{w}} a_{\mathbf{w}'} + a_{\mathbf{w}'}^+ a_{\mathbf{w}} a_{\mathbf{w}'} f_{\mathbf{w}}^*$$

$$+ a_{\mathbf{w}}^+ a_{\mathbf{w}} a_{\mathbf{w}'} f_{\mathbf{w}'}^* + a_{\mathbf{w}}^+ a_{\mathbf{w}'}^+ a_{\mathbf{w}} f_{\mathbf{w}'} + a_{\mathbf{w}}^+ a_{\mathbf{w}'}^+ a_{\mathbf{w}'} f_{\mathbf{w}}$$

$$+ a_{\mathbf{w}} a_{\mathbf{w}'} f_{\mathbf{w}}^* f_{\mathbf{w}'}^* + a_{\mathbf{w}'}^+ a_{\mathbf{w}} f_{\mathbf{w}}^* f_{\mathbf{w}} + a_{\mathbf{w}}^+ a_{\mathbf{w}'} f_{\mathbf{w}}^* f_{\mathbf{w}'}$$

$$+ a_{\mathbf{w}}^+ a_{\mathbf{w}'} f_{\mathbf{w}'}^* f_{\mathbf{w}} + a_{\mathbf{w}}^+ a_{\mathbf{w}} f_{\mathbf{w}'}^* f_{\mathbf{w}'} + a_{\mathbf{w}}^+ a_{\mathbf{w}'}^+ f_{\mathbf{w}} f_{\mathbf{w}'}$$

$$+ a_{\mathbf{w}}^+ f_{\mathbf{w}} f_{\mathbf{w}'}^* f_{\mathbf{w}'} + a_{\mathbf{w}'}^+ f_{\mathbf{w}'} f_{\mathbf{w}}^* f_{\mathbf{w}} + a_{\mathbf{w}} f_{\mathbf{w}}^* f_{\mathbf{w}'}^* f_{\mathbf{w}'}$$

$$+ a_{\mathbf{w}'} f_{\mathbf{w}'}^* f_{\mathbf{w}}^* f_{\mathbf{w}} + f_{\mathbf{w}}^* f_{\mathbf{w}'}^* f_{\mathbf{w}} f_{\mathbf{w}'})\,, \tag{3g}$$

$$A_{\mathbf{w}} = \frac{2M}{\hbar^2}\,V_{\mathbf{w}}(\beta_1\,e^{-i\beta_1\mathbf{w}\cdot\mathbf{r}} + \beta_2\,e^{i\beta_2\mathbf{w}\cdot\mathbf{r}})\,. \tag{3h}$$

$\mathscr{H}_{\text{ex}}^2$ and $\mathscr{H}_2$ are terms describing the interaction between phonons of different wave vectors in the recoil process.

3. Perturbation Calculation

We regard $(\mathscr{H}_{\text{ex}}^0 + \mathscr{H}_0)$ as the unperturbed Hamiltonian of the exciton–phonon system, and $(\mathscr{H}_{\text{ex}}^1 + \mathscr{H}_{\text{ex}}^2 + \mathscr{H}_1 + \mathscr{H}_2)$ as the perturbation part in the perturbation calculation. The unperturbed ground state wave function of the system is $\phi = \varphi(r)\,|0\rangle$ where $\varphi(r)$ is the normalized exciton wave function. $|0\rangle$ is the zero phonon state, which satisfies

$$a_{\mathbf{w}}\,|0\rangle = 0\,,$$
$$a_{\mathbf{w}}^+\,|0\rangle = \varphi(|\mathbf{w})\,,$$
$$\langle 0\,|\,0\rangle = 1\,, \tag{4}$$

where $\varphi(|\mathbf{w})$ is the wave function of one phonon with wave vector $\mathbf{w}$.

The effective unperturbed Hamiltonian of the exciton may be expressed as

$$H_{\text{eff}}^0 = \langle 0 | \mathscr{H}_{\text{ex}}^0 + \mathscr{H}_0 | 0 \rangle$$

$$= \frac{\hbar^2 K^2}{2M} - \frac{\hbar^2}{2\mu} \nabla_r^2 - \frac{e^2}{\varepsilon_0 r} - \alpha \hbar \omega \left(2 - \frac{\beta_1^2 + \beta_2^2}{2\beta_1 \beta_2} \right)$$

$$- \alpha \hbar \omega \, e^{-ur} - \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \frac{e^2}{r} \, e^{-ur} , \tag{5a}$$

$$\alpha = \frac{Me^2}{\hbar^2 u} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) , \tag{5b}$$

where α is the coupling parameter of the exciton with phonon. The first-order perturbation energy induced by $(\mathscr{H}_{\text{ex}}^1 + \mathscr{H}_{\text{ex}}^2 + \mathscr{H}_1 + \mathscr{H}_2)$ is zero. Now we are ready to calculate the second-order perturbation energy,

$$\Delta E = -\sum_n{}' \frac{|(\mathscr{H}_1 + \mathscr{H}_{\text{ex}}^1 + \mathscr{H}_{\text{ex}}^2 + \mathscr{H}_2)_{0n}|^2}{E_n - E_0}$$

$$= -\frac{\alpha}{6} \left(\frac{\hbar^2 K^2}{2M} + \frac{\beta_1^2 + \beta_2^2}{\beta_1 \beta_2} \frac{\hbar^2}{2\mu} \nabla_r^2 \right)$$

$$- \alpha^2 \hbar \omega \left(\frac{A}{\beta_1^2 + \beta_2^2} + \frac{D}{2\beta_1 \beta_2} + a' \right) . \tag{6}$$

In (6), the first term being proportional to the coupling constant α is the energy, which neglects the interaction between phonons of different wave vectors in the recoil process, the second term being proportional to the squared coupling constant α^2 is an extra energy, which considers the corresponding interactions.

Finally, using (5a) and (6) (see Appendix), the effective Hamiltonian of the exciton can be expressed as

$$H_{\text{eff}} = \frac{\hbar^2 K^2}{2M^*} + \frac{p^2}{2\mu^*} - \frac{e^2}{\varepsilon_0 r} + V_{\text{eff}} - E_{\text{tr}} , \tag{7a}$$

$$V_{\text{eff}} = \alpha \hbar \omega \, e^{-ur} - \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \frac{e^2}{r} \, e^{-ur} - \alpha^2 \hbar \omega \left(\frac{1}{2\beta_1^3 \beta_2^3} D + a \right) , \tag{7b}$$

$$E_{\text{tr}} = \alpha \hbar \omega \left(2 - \frac{\beta_1^2 + \beta_2^2}{2\beta_1 \beta_2} \right) + \alpha^2 \hbar \omega \left[\frac{\beta_1^2 + \beta_2^2}{2\beta_1^2 \beta_2^2} \left(\frac{1}{12} - \frac{2}{9\pi} \right) + \frac{1}{6} \right] , \tag{7c}$$

$$M^* = \frac{M}{1 - \dfrac{\alpha}{6}} , \tag{7d}$$

$$\mu^* = \frac{\mu}{1 + \dfrac{\alpha}{3} \dfrac{\beta_1^2 + \beta_2^2}{2\beta_1 \beta_2}} , \tag{7e}$$

where V_{eff} is the effective interaction potential between the electron and the hole. M^* and μ^* are the effective mass of the mass centre and the effective reduced mass of an exciton. E_{tr} is the self-trapping energy of the exciton.

4. Results and Discussion

4.1 Effective interaction potential

The effective interaction potential can formally be divided into two parts,

$$V_{\text{eff}} = V_{\text{eff}}^1 + V_{\text{eff}}^2 , \qquad (8\text{a})$$

where

$$V_{\text{eff}}^1 = \alpha\hbar\omega \, e^{-ur} - \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right) \frac{e^2}{r} \, e^{-ur} , \qquad (8\text{b})$$

$$V_{\text{eff}}^2 = -\alpha^2\hbar\omega \left(\frac{1}{2\beta_1^3\beta_2^3} \, D + a\right) . \qquad (8\text{c})$$

V_{eff}^1 stands for the screening potential of the electron and the hole resulting from the screening action of the polarizing lattice. They are called Yukawa and Born-Mayer potentials, which omit the corresponding interaction and are independent of the relative magnitudes of the electron and hole masses. V_{eff}^2 represents the extra induced potential being proportional to the squared coupling constant α^2, which considers the corresponding interaction and depends on the relative magnitudes of the electron and hole masses. In Table 1, the data for the GaAs crystal are given. Fig. 1 shows the variation in the effective interaction potentials V_{eff}^1 and V_{eff}^2 of the exciton in a GaAs crystal with relative coordinate r. From the figure, one can see that the effective interaction potentials V_{eff}^1 and V_{eff}^2 decrease with increasing relative coordinate r. When $r < 5$ Å, the effective interaction potential V_{eff} only includes V_{eff}^1, whereas V_{eff}^2 is neglected. When $r > 5$ Å, the effective interaction potential V_{eff} includes not only V_{eff}^1, but there exists also an extra induced potential V_{eff}^2, which considers the corresponding interaction.

4.2 Self-trapping energy and self-trapping conduction

Equation (7c) shows that the self-trapping energy of the exciton in polar crystals, induced by the interactions of electron and hole with phonons, depends on the exciton–phonon coupling parameter α and the relative magnitudes of the electron and hole masses. In (7c), the first term being proportional to the coupling constant α is the self-trapping energy, which neglects the corresponding interaction. The second term being proportional to the squared coupling constant α^2 describes extra self-trapping energy, which considers the corresponding interaction.

The self-trapping condition of the exciton is $E_{\text{tr}} > 0$, we have

$$\left(3 + \frac{\alpha}{6}\right)(\beta_1\beta_2)^2 - \left(\frac{1}{2} + \frac{\alpha}{12} - \frac{2\alpha}{9\pi}\right)(\beta_1\beta_2) + \left(\frac{1}{24} - \frac{1}{9\pi}\right)\alpha > 0 . \qquad (9)$$

Table 1

The parameters for a GaAs crystal taken from [19, 20]

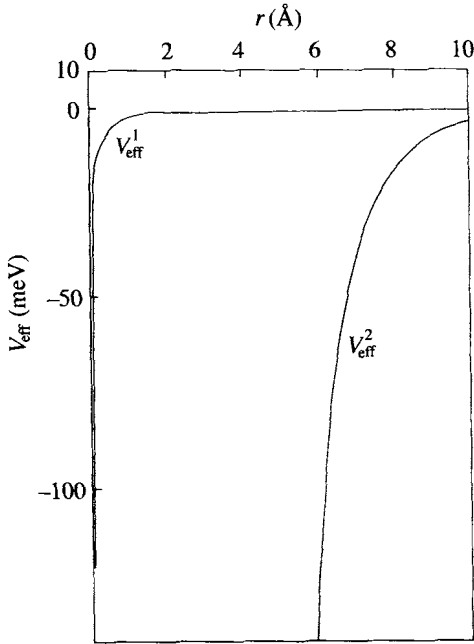| material	ε_0	ε_∞	$\hbar\omega_s$ (meV)	β_1	β_2	α
GaAs	12.5	10.9	35.2	0.123	0.877	0.1997

r (Å)

Fig. 1. The relation between the effective interaction potentials V_{eff}^1 and V_{eff}^2 of the exciton in a GaAs crystal with relative coordinate r

According to the solving process of secondary inequality, we can know that the exciton is self-trapped when $\alpha > 3.2789$ and $\beta_1 \beta_2$ take arbitrary values. When $0 < \alpha < 3.2789$, we have

$$\beta_1 \beta_2 > C_+, \qquad \beta_1 \beta_2 < C_-, \tag{10a}$$

$$C_\pm = \frac{\left(\dfrac{1}{2} + \dfrac{\alpha}{12} - \dfrac{2\alpha}{9\pi}\right) \pm \sqrt{\left(\dfrac{1}{2} + \dfrac{\alpha}{12} - \dfrac{2\alpha}{9\pi}\right) - 2\left(6 + \dfrac{\alpha}{3}\right)\left(\dfrac{1}{24} - \dfrac{1}{9\pi}\right)\alpha}}{6 + \dfrac{\alpha}{3}}. \tag{10b}$$

Because $\beta_1 + \beta_2 = 1$, the maximum value of $\beta_1 \beta_2$ is equal to 0.25, i.e.,

$$0 < \beta_1 \beta_2 < 0.25. \tag{11a}$$

Thus, we have

$$C_+ < \beta_1 \beta_2 < 0.25, \tag{11b}$$

$$0 < \beta_1 \beta_2 < C_-. \tag{11c}$$

Solving equations (11a) to (11c), we can obtain that the exciton is self-trapped when the exciton–phonon coupling parameter is in the range $0 < \alpha < 3.2789$ and

$$\beta_1 < \beta_1', \qquad \beta_1^{(0)} < \beta_1 < \beta_1^{(1)}, \qquad \beta_1 > \beta_1''; \tag{12a}$$

$$\beta_1' = \frac{1 - \sqrt{1 - 4C_-}}{2}, \qquad \beta_1^{(0)} = \frac{1 - \sqrt{1 - 4C_+}}{2}, \tag{12b}$$

$$\beta_1^{(1)} = \frac{1 + \sqrt{1 - 4C_+}}{2}, \qquad \beta_1'' = \frac{1 + \sqrt{1 - 4C_-}}{2}. \tag{12c}$$
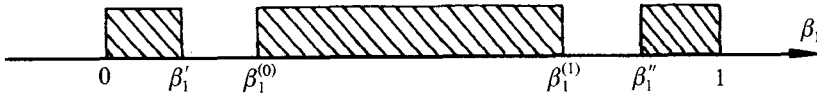
Fig. 2. The self-trapping range of the exciton $(0 < \alpha < 3.2789)$

In summary, the self-trapping condition of the exciton in polar crystals is that, when the exciton–phonon coupling parameter α is in the range $0 < \alpha < 3.2789$, $0 < \beta_1 < \beta_1'$, $\beta_1^{(0)} < \beta_1 < \beta_1^{(1)}$, $\beta_1'' < \beta_1 < 1$ are the self-trapping ranges. Fig. 2 shows the self-trapping ranges of the exciton in polar crystals. From Fig. 2 one can see that the space between β_1', $\beta_1^{(1)}$ and $\beta_1^{(0)}$, β_1'' increases with decreasing coupling parameter α. As $\alpha \to 0$, the self-trapping range of the exciton is identical with that obtained if the interaction between phonons of different wave vectors in the recoil process is neglected, i.e., $0.2113 < \beta_1 < 0.7889$. When $\alpha = 3.2789$, $\beta_1^{(0)} = \beta_1'$, $\beta_1^{(1)} = \beta_1''$, all ranges join together, for an arbitrary value of β_1 the exciton in polar crystals is self-trapped, when the exciton–phonon coupling parameter α is in the range $\alpha \geq 3.2789$.

The extra self-trapping energy, which considers the corresponding interaction, is given by

$$E_2 = \alpha^2 \hbar \omega \left[\frac{\beta_1^2 + \beta_2^2}{2\beta_1^2 \beta_2^2} \left(\frac{1}{12} - \frac{2}{9\pi} \right) + \frac{1}{6} \right]. \tag{13a}$$

The self-trapping energy, which omits the corresponding interaction, is

$$E_1 = \alpha \hbar \omega \left(2 - \frac{\beta_1^2 + \beta_2^2}{2\beta_1 \beta_2} \right). \tag{13b}$$
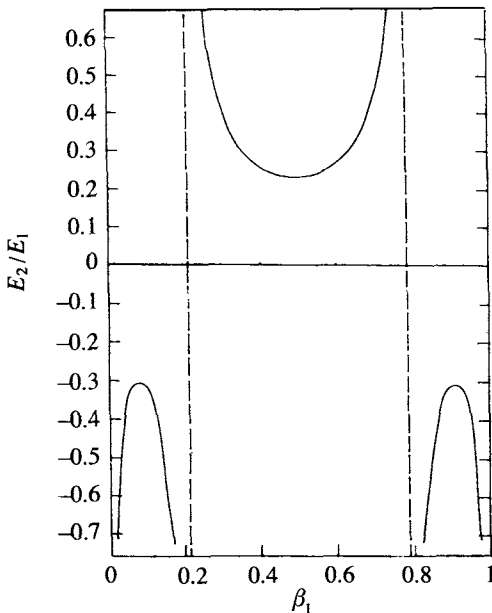
The ratio of E_2 and E_1, is

$$P = \frac{E_2}{E_1} = \frac{\dfrac{\beta_1^2 + \beta_2^2}{\beta_1 \beta_2} \left(\dfrac{1}{12} - \dfrac{2}{9\pi} \right) + \dfrac{\beta_1 \beta_2}{3}}{2\beta_1 \beta_2 - (\beta_1 - \beta_2)^2} \, \alpha. \tag{13c}$$

Fig. 3 gives a description of the variation of P with the fraction of the mass of the electron β_1 using α as unit of P. From the figure one can see that when β_1 is around 0, 0.2113, 0.7886, and 1, we have

Fig. 3. The relation between E_2/E_1 and β_1

Table 2
The material parameters for several crystals taken from [19, 20]

material	m_e/m_0 [1]	m_h/m_0	α_e	α_h	$\alpha = (\alpha_e^2 + \alpha_h^2)^{1/2}$ [2]	one-phonon [3] process	multi-phonon [4] process	E_2/E_1
CdSe	0.116	0.69	0.427	1.04	1.124	non-self-trapping	non-self-trapping	0.51
CuCl	0.44	3.6	2.00	5.73	6.07	non-self-trapping	self-trapping	2
GaAs	0.07	0.5	0.07	0.187	0.1977	non-self-trapping	non-self-trapping	0.07
ZnO	0.24	0.49	0.899	1.29	1.57	self-trapping	self-trapping	0.51

[1]) m_0 is the free-electron rest mass.
[2]) $\alpha = (\alpha_e^2 + \alpha_h^2)^{1/2}$ is taken from [18].
[3]) One-phonon process represents a process which omits the interaction between phonons of different wave vectors in the recoil process.
[4]) Multi-phonon process represents a process, which considers the corresponding interaction.

$E_2 \gg E_1$, i.e., the extra self-trapping energy E_2 is dominant. When β_1 takes other values, the magnitude of P depends on the exciton–phonon coupling strength α. For example, when $\alpha = 0.5$, for $\beta_1 = 0.5$, $E_2/E_1 = 10.8\%$, for $\beta_1 = 0.4$, $E_2/E_1 = 12.2\%$, for $\beta_1 = 0.3$, $E_2/E_1 = 20\%$.

From (13a) and (13b) one can see that when β_1 and β_2 are taking arbitrary values, $E_2 > 0$, whereas when β_1 (or β_2) is taking a value in the range $0 < \beta_1 < 0.2113$ and $0.7886 < \beta_1 < 1$, $E_1 < 0$, and when $0.2113 < \beta_1 < 0.7866$, $E_1 > 0$. Of course, when β_1 (or β_2) is in the range $0 < \beta_1 < 0.2113$ and $0.7886 < \beta_1 < 1$, $E_2/E_1 < 0$, whereas when $0.2113 < \beta_1 < 0.7886$, $E_2/E_1 > 0$.

In order to explain more clearly the influence on the self-trapping energy and the self-trapping condition of the exciton in polar crystals, which considers the corresponding interaction, numerical calculations, for the several crystals, as example, are performed and the results are given in Table 2.

From the above discussion one can deduce that the influence of the corresponding interaction on the self-trapping energy and the self-trapping condition of the exciton in polar crystals cannot be neglected.

Appendix

Let us first calculate (6),

$$\Delta E = -\sum_n' \frac{|(\mathcal{H}_1 + \mathcal{H}_{ex}^1 + \mathcal{H}_{ex}^2 + \mathcal{H}_2)_{0n}|^2}{E_n - E_0}$$
$$= (\Delta E)_1 + (\Delta E)_{ex}^1 + (\Delta E)_{ex}^2 + (\Delta E)_2 + (\Delta E')_1$$
$$+ (\Delta E')_2 + (\Delta E')_3 + (\Delta E')_4 + (\Delta E')_5 + (\Delta E')_6 . \tag{A1}$$

Replacing the summation by integration, after straightforward calculation we obtain

$$(\Delta E)_1 = -\sum_n' \frac{|(\mathcal{H}_1)_{0n}|^2}{E_n - E_0} = -\frac{\alpha}{6} \frac{\hbar^2 K^2}{2M} , \tag{A2}$$

$$(\Delta E)^1_{\text{ex}} = -\sideset{}{'}\sum_n \frac{|(\mathscr{H}'_{\text{ex}})_{0n}|^2}{E_n - E_0} = -\frac{\alpha}{3}\left(\frac{\beta_1^2 + \beta_2^2}{2\beta_1\beta_2}\right)\frac{\hbar^2}{2\mu}\,\nabla_{\mathbf{r}}^2\,, \tag{A3}$$

$$(\Delta E)^2_{\text{ex}} = -\sideset{}{'}\sum_n \frac{|(\mathscr{H}^2_{\text{ex}})_{0n}|^2}{E_n - E_0} = -\alpha^2\hbar\omega(A + 2B + D)\,, \tag{A4}$$

where

$$A = \left(\frac{1}{12} - \frac{2}{9\pi}\right)\frac{\beta_1^2 + \beta_2^2}{2\beta_1^2\beta_2^2}\,,$$

$$B = \frac{\beta_1^2 + \beta_2^2}{2\beta_1\beta_2}\left[\frac{ur}{144} + \frac{1}{18} - \frac{49}{48}\frac{1}{ur} - \frac{1}{8}\frac{1}{(ur)^2} + \frac{23}{8}\frac{1}{(ur)^3}\right]\text{e}^{-ur}\,,$$

$$D = \frac{4}{\pi}\,\xi(r) + \frac{2}{(ur)^6}\left(\frac{1}{2} + \sqrt{2}\,ur\right)\text{e}^{-\sqrt{2}\,ur} + \frac{4}{(ur)^6} - \frac{2}{(ur)^5}(ur + 2)\,\text{e}^{-ur}$$

$$+ \frac{1}{4(ur)^6}\left[\frac{1}{3}\,(ur)^5 + \frac{5}{3}\,(ur)^4 + \frac{13}{6}\,(ur)^3 - \frac{3}{4}\,(ur)^2 - \frac{3}{2}\,ur - \frac{3}{2}\right]\text{e}^{-2ur}\,, \tag{A5}$$

$$\xi(r) = \frac{u^4}{r^6}\int\limits_0^\infty \frac{\text{e}^{-(w^2 + 2u^2)^{1/2}r}}{w(w^2 + u^2)^4\,(w^2 + 2u^2)}\,[wr^2(w^2 + 2u^2)^{1/2}\cos(wr)$$

$$- \sin(wr) + wr\,\cos(wr) - r(w^2 + 2u^2)^{1/2}\sin(wr)]\,\text{d}w\,, \tag{A6}$$

$$(\Delta E)_2 = -\sideset{}{'}\sum_n \frac{|(\mathscr{H}_2)_{0n}|^2}{E_n - E_0} = -\alpha^2\hbar\omega(a' + b)\,, \tag{A7}$$

where

$$a' = \tfrac{1}{6} + a\,, \tag{A8}$$

$$a = \frac{1}{6}\left[-(2 - ur)\,\text{e}^{-ur} + \left(\frac{6}{ur} + \frac{24}{(ur)^2} + \frac{96}{(ur)^3} + \frac{87}{(ur)^4} - \frac{18}{(ur)^5}\right)\text{e}^{-2ur}\right]\,, \tag{A9}$$

$$b = \frac{4\beta_1^2\beta_2^2}{(\beta_1^2 + \beta_2^2)^2}\,A - \frac{4\beta_1\beta_2}{\beta_1^2 + \beta_2^2}\,B + D\,, \tag{A10}$$

$$(\Delta E')_1 = -\sideset{}{'}\sum_n \frac{(\mathscr{H}^1_{\text{ex}})_{0n}\,(\mathscr{H}_1)^*_{0n} + (\mathscr{H}_1)_{0n}\,(\mathscr{H}^1_{\text{ex}})^*_{0n}}{E_n - E_0} = 0\,, \tag{A11}$$

$$(\Delta E')_2 = -\sideset{}{'}\sum_n \frac{(\mathscr{H}_1)_{0n}\,(\mathscr{H}_2)^*_{0n} + (\mathscr{H}_2)_{0n}\,(\mathscr{H}_1)^*_{0n}}{E_n - E_0} = 0\,, \tag{A12}$$

$$(\Delta E')_3 = -\sideset{}{'}\sum_n \frac{(\mathscr{H}_1)_{0n}\,(\mathscr{H}^2_{\text{ex}})^*_{0n} + (\mathscr{H}^2_{\text{ex}})_{0n}\,(\mathscr{H}_1)^*_{0n}}{E_n - E_0} = 0\,, \tag{A13}$$

$$(\Delta E')_4 = -\sideset{}{'}\sum_n \frac{(\mathscr{H}^1_{\text{ex}})_{0n}\,(\mathscr{H}_2)^*_{0n} + (\mathscr{H}_2)_{0n}\,(\mathscr{H}^1_{\text{ex}})^*_{0n}}{E_n - E_0} = 0\,, \tag{A14}$$

$$(\Delta E')_5 = -\sideset{}{'}\sum_n \frac{(\mathscr{H}^1_{\text{ex}})_{0n}\,(\mathscr{H}^2_{\text{ex}})^*_{0n} + (\mathscr{H}^2_{\text{ex}})_{0n}\,(\mathscr{H}^1_{\text{ex}})^*_{0n}}{E_n - E_0} = 0\,, \tag{A15}$$

$$(\Delta E')_6 = -\sum_n{}' \frac{(\mathscr{H}_{ex}^2)_{0n}\,(\mathscr{H}_2)_{0n}^* + (\mathscr{H}_2)_{0n}\,(\mathscr{H}_{ex}^2)_{0n}^*}{E_n - E_0} = -a^2(d+f)\,, \qquad (A16)$$

where

$$d = \frac{(\beta_1 - \beta_2)^2}{2\beta_1\beta_2}\, b\,, \qquad (A17)$$

$$f = \frac{1}{2\beta_1\beta_2}\, D\,. \qquad (A18)$$

Finally we can obtain

$$\Delta E = -\frac{a}{6}\left(\frac{\hbar^2 K^2}{2M} + \frac{\beta_1^2 + \beta_2^2}{\beta_1\beta_2}\frac{\hbar^2}{2\mu}\,\nabla_r^2\right) - a^2\hbar\omega\left(\frac{A}{\beta_1^2 + \beta_2^2} + \frac{D}{2\beta_1\beta_2} + a'\right)\,. \qquad (A19)$$

References

[1] H. HAKEN, Z. Phys. **146**, 527 (1956).
[2] G. MAHLER and U. SCHRÖDER, phys. stat. sol. (b) **61**, 629 (1974).
[3] M. MATSUURA and H. BUTTNER, Phys. Rev. B **21**, 679 (1980).
[4] G. IADONISI, F. BASSANI, and G. STRINATI, phys. stat. sol. (b) **153**, 611 (1989).
[5] S. WANG, C. K. MAHUTTE, and M. MATSUURA, phys. stat. sol. (b) **51**, 11 (1972).
[6] S. WANG and M. MATSUURA, Phys. Rev. B **10**, 3330 (1974).
[7] K. HATTORI, phys. stat. sol. (b) **76**, 281 (1976).
[8] M. MATSUURA and C. MAVROYANIS, Phys. Rev. B **13**, 2131 (1976).
[9] J. SAK, Phys. Rev. B **6**, 2226 (1972).
[10] S. D. MAHANTI and C. M. VARMA, Phys. Rev. B **6**, 2209 (1972).
[11] H. HAKEN, Z. Phys. **147**, 323 (1957); **155**, 223 (1959).
[12] J. ADAMOWSKI and B. GERLACH, Phys. Rev. B **23**, 2943 (1981).
[13] QIANLI YIANG, JINGQI MIAO, and SHIWEI GU, J. Phys.: Condensed Matter **1**, 10343 (1989).
[14] JINGQI MIAO, QIANLI YANG, and SHIWEI GU, Phys. Rev. B **40**, 9846 (1989).
[15] H. SUMI and A. SUMI, J. Phys. Soc. Japan **63**, 637 (1994).
[16] QINGHU CHEN, KELIN WANG, and SHAOLONG WAN, Phys. Rev. B **50**, 14583 (1994).
[17] JING LIN XIAO, TONG HAIXING, BAO QUAN SUN, and WEI XIAO, phys. stat. sol. (b) **183**, 425 (1994).
[18] S. W. GU, J. Science **27**, 339 (1982) (in Chinese).
[19] E. O. KANE, Phys. Rev. B **18**, 6849 (1978).
[20] E. KARTHEUSER, Polarons in Ionic Crystals and Polar Semiconductors, North-Holland Publ. Co., Amsterdam 1972.