
ENERGY SPECTRA OF EXCITONS OF VARIOUS RADII

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UDC 538
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The energy spectra, dispersion laws, widths of exciton bands, and effective masses for excitons of large and small radii have been investigated. The account of the crystal lattice discreteness allows us to reveal some new features of excitons: the anisotropy of their relative motion and the dependences of the Rydberg exciton constant and the exciton band width on the main quantum number n .

1. Introduction

Developed in the pioneer works of Frenkel [1], Peierls [2], Wannier [3], and Mott [4], the theory of excitons has been experimentally confirmed for most dielectric and semiconductor crystals. Its further elaboration has occurred in two directions, as stated by Knox [5]: the studies of “the exciton structure” and “the dynamics of excitons.” As the former direction, Knox means the determination of excited electron states depending on the wave vector of an exciton in the ideal lattice. According to [5], the latter direction includes the study of the interaction of excitons with one another and with other fields or particles. The present work can be referred to the studies of the exciton structure.

The majority of theoretical and experimental works in this field has considered the dynamics of excitons. In this case, one of the well-known structural models such as the model of Frenkel or Wannier–Mott excitons and the model of excitons with charge transfer is used. These works account for the interaction of excitons with phonons, photons, defects of a crystal lattice or admixtures, exciton-exciton interaction, etc. (see, e.g.,

[6–8]). If the interaction of excitons with other particles or fields is weak, then the exciton structure varies slightly. However, under strong interactions or the high intensities of exciting fields, the structure of excitons can be significantly changed. For example, under the strong phonon-exciton interaction, the self-localized excitons [8, Sections 12, 13] or the polaron states of electrons and holes can be created. The strong photon-exciton interaction leads to the creation of exciton polaritons [8, Section 3]. At the high intensities of exciting fields, the many-electron or many-exciton complexes, exciton droplets, and other dynamical structures can be formed.

At present, the excitons in low-dimensional systems (membranes, films, linear polymers, and biopolymers) [9, 10] and in small volumes (quantum dots and wires) [11, 12] are intensively studied. Such an attention to these objects is obviously related to the possibility of their practical use in electronics and other branches of industry. These works are based on two-three above-mentioned structural models of excitons. The discreteness of a crystal lattice is considered rarely [9, 13], as well as the excitons with intermediate radii [5, 8, 14].

Consider the most studied model of Wannier–Mott excitons with the energy spectrum [5]

$$E_n(k) = E_g + \frac{\hbar^2 k^2}{2m_{\text{ex}}} - \frac{R_{\text{ex}}}{n^2}, \quad (1)$$

where E_g – forbidden zone width, $m_{\text{ex}} = m_e + m_h$, m_e , m_h – effective masses of an electron and a hole, respectively, $R_{\text{ex}} = \mu e^4 / (2\varepsilon^2 \hbar^2)$ – Rydberg constant of an exciton, $\mu = (m_e m_h) / m_{\text{ex}}$, ε – dielectric

constant, k — wave vector of an exciton, and n is the principal quantum number of an exciton. Formula (1) holds only near the exciton band bottom. The numerous modifications and improvements of this model are restricted by the region of small k . Moreover, the band structure for the whole scope of the Brillouin zone was not practically studied.

The goal of the present paper is, firstly, the construction of a more general model of excitons which would describe all the known excitons as partial cases. Then we will take the discreteness of a crystal lattice into account for excitons with small and intermediate radii, which is the second purpose of our work.

2. Hamiltonian. Schrödinger Equation

The Hamiltonian with the electron-hole interaction for Wannier–Mott excitons with intermediate and small radii looks as

$$H = H_e + H_h + H_{eh}, \quad (2)$$

where $H_e = \sum_n E'_g a_n^+ a_n + \sum_{n,m} I_{nm} a_n^+ a_m$, $H_h = \sum_{n,m} J_{nm} b_n^+ b_m$, $H_{eh} = \sum_{n,m} U_{nm} a_n^+ b_m^+ a_n b_m$, E'_g — the distance between the centers of the conduction and valence bands, a_n^+, a_n, b_n^+, b_n — the creation and annihilation operators of electrons (a) and holes (b), $n = (n_1, n_2, n_3)$ — numbers of lattice nodes, I_{nm} — the electron exchange energy, $I_{nn} = 0$, J_{nm} — the same for a hole, U_{nm} — the Coulomb (or another) interaction energy. The exchange integrals I_{nm} , J_{nm} quickly decrease as functions of the distance $|\vec{r}_n - \vec{r}_m|$. Therefore, it is possible to use the approximation of nearest neighbors for I_{nm} , J_{nm} . We have six nonzero coefficients $I_{n\alpha} \neq 0$ for a cubic lattice ($\alpha = 1, 2, \dots, 6$). Let $I_{n\alpha} \equiv -I$; $J_{n\alpha} \equiv -J$. The signs of I, J correspond to the positive effective masses of an electron and a hole if $I > 0, J > 0$.

On the two-particle states $|\psi\rangle = \sum_{n,m} f_{nm} a_n^+ b_m^+ |0\rangle$, the Schrödinger equation $H|\Psi\rangle = E|\Psi\rangle$ yields the equation for f_{nm} in the form

$$-\sum_{\alpha=1}^6 (I f_{n\alpha} + J f_{\alpha m}) + (U_{nm} - E') f_{nm} = 0, \quad (3)$$

where $E' = E - E'_g$, $f_{n1} \equiv f_{(n_1+1, n_2, n_3)(m_1, m_2, m_3)}, \dots, f_{6m} \equiv f_{(n_1, n_2, n_3)(m_1, m_2, m_3+1)}$.

3. Separation of Variables. Relative Motion

We introduce the new variables

$$\nu_i = \frac{I m_i + J n_i}{I + J}, \quad l_i = n_i - m_i; \quad i = 1, 2, 3. \quad (4)$$

They are discrete analogs of the center-of-mass and relative coordinates. We set

$$f_{nm} = e^{ik\nu} e^{iq l} \cdot C(l), \quad (5)$$

where $k = (k_1, k_2, k_3)$, $k\nu = \sum_{i=1}^3 k_i \nu_i$, the same is implied for q , $C(l) \equiv C(l_1, l_2, l_3)$, $C(l)$ can be complex-valued. If we set $\tan q_i = \frac{J \sin \beta_i - I \sin \alpha_i}{I \cos \alpha_i + J \cos \beta_i}$, $\alpha_i = \frac{J}{I+J} k_i$, $\beta_i = \frac{I}{I+J} k_i$, then the variables can be separated, and we get the system of equations describing the relative motion

$$\sum_{i=1}^3 L_i (C(l_i + 1) + C(l_i - 1)) + (U_l - E') C(l) = 0. \quad (6)$$

Here, $L_i = \pm \sqrt{I^2 + J^2 + 2IJ \cos k_i}$, $U_l \equiv U_{nm}$, $C(l_2 \pm 1) \equiv C(l_1, l_2 \pm 1, l_3)$. The negative sign of L_i corresponds to the bound states of an electron and a hole. We restrict ourselves to this case in what follows.

4. Excitons with Large Radii

If $C(l)$ in (6) is a smooth function of l_i , it is possible to use the continual approximation: $l \rightarrow (x, y, z)$, $C(l) \rightarrow \Psi(x, y, z)$, $U_l \rightarrow U(x, y, z)$, where x, y, z are the dimensionless coordinates. Let $\Psi(x \pm 1, y, z) \approx \Psi(x, y, z) \pm \frac{d\Psi}{dx} + \frac{1}{2} \frac{d^2\Psi}{dx^2}$, and let the same hold for the variables y, z .

The conditions for the continual approximation to be true are

$$|\Psi| \gg \left| \frac{d\Psi}{dx} \right| \gg \left| \frac{d^2\Psi}{dx^2} \right| \gg \dots, \quad (7)$$

and the same holds for y and z .

We take $U(x, y, z) = -\frac{e^2}{\varepsilon ar}$ for the Coulomb interaction, where $r = \sqrt{x^2 + y^2 + z^2}$, ε and a — dielectric and lattice constants. Then relation (6) yields

$$L_1 \frac{d^2\Psi}{dx^2} + L_2 \frac{d^2\Psi}{dy^2} + L_3 \frac{d^2\Psi}{dz^2} - \left(\frac{e^2}{\varepsilon ar} + E' - 2(L_1 + L_2 + L_3) \right) \Psi = 0. \quad (8)$$

Equation (8) has the anisotropy term due to the wave vector k of an exciton. If the exciton moves

along a diagonal of the cube ($|k_1| = |k_2| = |k_3|$), Eq. (8) corresponds to the isotropy:

$$L(k)\Delta\Psi - \left(\frac{e^2}{\varepsilon ar} - 6L(k) + E'\right)\Psi = 0, \quad (9)$$

where $L(k) = L_i$, $k = |k_i|$.

Equation (9) which is similar to that for the hydrogen wave function gives the energy spectrum

$$E_n(k) = E'_g - 6\sqrt{I^2 + J^2 + 2IJ \cos k} - \frac{e^4}{4\varepsilon^2 a^2 \sqrt{I^2 + J^2 + 2IJ \cos k}} \cdot \frac{1}{n^2}, \quad (10)$$

where $n = 1, 2, \dots$ – the principle quantum number.

It follows from (10) that

- the Rydberg constant of an exciton depends on k ;
- the dispersion law, $E_n(k)$, depends on n and can be nonmonotone;
- the widths of exciton bands are finite and depend on n .

The monotonicity of $E_n(k)$ depends on the dimensionless parameter $\gamma = \frac{1}{2IJ}(U_a^2/(24n^2) - (I^2 + J^2))$, where $U_a = e^2/(\varepsilon a)$. If $|\gamma| \geq 1$, then the dispersion law $E_n(k)$ is monotone, and $k_1 = 0$, $k_2 = \pi$ correspond to the minimum and maximum of $E_n(k)$, respectively. The exciton band width is $\Delta E_n = E_n(\pi) - E_n(0)$, i.e.,

$$\Delta E_n = \begin{cases} 12J \left(1 - \frac{U_a^2}{24n^2(I^2 - J^2)}\right), & \text{if } I > J, \\ 12I \left(1 - \frac{U_a^2}{24n^2(I^2 - J^2)}\right), & \text{if } I < J. \end{cases}$$

If $|\gamma| < 1$, the maximum of $E_n(k)$ arises at $k_3 = \arccos \gamma$. At $k_1 = 0$, $k_2 = \pi$, it has two minima. But conditions (7) can be broken in this case. It is possible to present inequality (7) as

$$R_n \gg a, \quad (11)$$

where $R_n = 3an^2L(k)/U_a$ – the intermediate radius of an exciton in the state characterized by n, k . The function $L(k)$ has minimum at $k = \pi$. Then inequality (11) gives $|I - J| \gg U_a/3n^2$ which together with the inequality $|\gamma| < 1$ imply that $\frac{1}{2\sqrt{6}n} > \frac{|I \pm J|}{U_a} \gg \frac{1}{3n^2}$. It is satisfied only at large n and small $|I - J|/U_a$. Since condition (11) is more strict than $|\gamma| < 1$, the nonmonotone behavior of $E_n(k)$ can be evidently revealed for the excitons with intermediate radii.

It is possible to use the perturbative methods for solving Eq. (8), if $L(k_i)$ satisfies the condition

$|L(0) - L(\pi)|/|L(0)| \ll 1$ or if $|k_1| \approx |k_2| \approx |k_3|$. Then we can get a small correction to $E_n(0)$ in (10). Equation (8) is anisotropic, but the effective mass is isotropic. At small k , the anisotropy is negligibly small. In this case, we have two small parameters k and $|\Delta\Psi|/|\Psi|$, which follows from (7). In order to keep the quantities to be of the same order, we must decompose relation (6) in a different way: in Ψ to within k_i^2 and in $\frac{d^2\Psi}{dx^2}$, $\frac{d^2\Psi}{dy^2}$, $\frac{d^2\Psi}{dz^2}$ getting $L(k_i) \approx L(0)$. Thus, we obtain the equation

$$-(I + J)\Delta\Psi - \left(\frac{U_a}{r} + E' + 6(I + J) - \frac{2IJ}{I + J}k^2\right)\Psi = 0.$$

It is exactly the equation for the wave function of a Wannier-Mott exciton. The energy coincides with (1) if we set $\mu = \hbar^2/2a^2(I + J)$, $m_{\text{ex}} = \hbar^2(I + J)/4IJa^2$, $E_g = E' - 6(I + J)$.

5. Excitons of Small Radii

It is possible to solve system (6) in the discrete version by means of the method of successive approximations in the case where

$$|L_i| \ll U_a. \quad (12)$$

It is convenient to introduce the dimensionless quantities: $\lambda_i = -L_i/U_a > 0$, $\lambda_i \ll 1$, $\varepsilon = \frac{E'}{U_a}$,

$$u_l = -U_l/U_a, \quad u_l = \begin{cases} u_0, & \text{if } l = 0, \\ 1/\sqrt{l_1^2 + l_2^2 + l_3^2}, & \text{if } l \neq 0. \end{cases}$$

Then Eq. (6) can be transformed into

$$\sum_{i=1}^3 \lambda_i (C(l_i + 1) + C(l_i - 1)) + (\varepsilon + u_l)C(l) = 0. \quad (13)$$

We get the solutions in the zero-order approximation, if we put $\lambda_i = 0$ in (13). Then we have $(\varepsilon + u_l)C(l) = 0$. Provided $C_l \neq 0$, $\varepsilon = -u_l$. It is the energy in the zero-order approximation.

There are crystals (including molecular ones), for which $u_0 > 1$. In this case, the nearest energy level corresponds to Frenkel-like excitons. The next levels correspond to $l \neq 0$. In another crystals, $u_0 < 1$ or even $u_0 < 0$. Then the nearest energy level corresponds to $l \neq 0$, and the Frenkel-like excitons do not exist apparently. We consider the first case, $u_0 > 1$, and solve system (13) in the first-order approximation for the level $\varepsilon_0 = -u_0$. We take $\varepsilon_0 = -u_0 + \delta$, $C(0, 0, 0) = 1 - x$, $C(\pm 1, 0, 0) = y_1$, $C(0, \pm 1, 0) = y_2$, $C(0, 0, \pm 1) = y_3$, and $C(l) = 0$. We assume also that the quantities δ, x, y_i have the first order of smallness ($\sim \lambda_i$). In this case, system (13) together with the normalization condition

$\sum_l |C(l)|^2 = 1$ are satisfied to within $\sim \lambda_i$. Then $\delta = 0$, $x = 0$, $y_i = \frac{\lambda_i}{u_0 - 1}$.

In the second-order approximation, we introduce the corrections of the second order in $\sim \lambda_i^2$ and obtain

$$C(0, 0, 0) = 1 - 2 \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2}{(u_0 - 1)^2}, \quad C(\pm 1, 0, 0) = \frac{\lambda_1}{u_0 - 1},$$

$$C(0, \pm 1, 0) = \frac{\lambda_2}{u_0 - 1}, \quad C(0, 0, \pm 1) = \frac{\lambda_3}{u_0 - 1},$$

$$C(\pm 1, \pm 1, 0) = \frac{2\lambda_1\lambda_2}{(u_0 - 1)(u_0 - \frac{\sqrt{2}}{2})},$$

$$C(\pm 1, 0, \pm 1) = \frac{2\lambda_1\lambda_3}{(u_0 - 1)(u_0 - \frac{\sqrt{2}}{2})},$$

$$C(0, \pm 1, \pm 1) = \frac{2\lambda_2\lambda_3}{(u_0 - 1)(u_0 - \frac{\sqrt{2}}{2})},$$

$$C(\pm 2, 0, 0) = \frac{\lambda_1^2}{(u_0 - 1)(u_0 - \frac{1}{2})},$$

$$C(0, \pm 2, 0) = \frac{\lambda_2^2}{(u_0 - 1)(u_0 - \frac{1}{2})},$$

$$C(0, 0, \pm 2) = \frac{\lambda_3^2}{(u_0 - 1)(u_0 - \frac{1}{2})},$$

$$\varepsilon_0 = -u_0 - \frac{2(\lambda_1^2 + \lambda_2^2 + \lambda_3^2)}{u_0 - 1}.$$

The dispersion law, exciton band width, and effective mass are as follows:

$$E_0 = E'_g + U_0 + 2 \frac{3(I^2 + J^2) + 2IJ(\cos k_1 + \cos k_2 + \cos k_3)}{U_0 - U_a}, \quad (14)$$

$$\Delta E = \frac{2AIJ}{U_a - U_0}, \quad m_{\text{ex}} = \frac{\hbar^2(U_a - U_0)}{4IJa^2}.$$

In the third-order approximation, we get the corrections proportional to λ_i^3 .

The second energy level in the zero-order approximation corresponds to $|l| = 1$, and $\varepsilon_1 = -u_1 = -1$. This level is degenerate: $C(\pm 1, 0, 0) = C(0, \pm 1, 0) = C(0, 0, \pm 1) = \frac{\sqrt{6}}{6}$. It splits in the second-order approximation, and the wave function $C(l)$ is transformed by the representations of some point group. In the case where $k = 0$ or $|k_1| = |k_2| = |k_3|$, this group

involves the inversions and 2-, 3-, and 4-fold symmetry axes, which intersect at the point $l = 0$. If k_i is arbitrary, then the point group consists of the inversions and a 2-fold symmetry axis. The calculations and results are highly unwieldy in the last case. Therefore, we set forth the results for the isotropic case in the second-order approximation. The normalized totally symmetric wave function $C(l)$ (the identity representation) reads

$$C(\pm 1, 0, 0) = C(0, \pm 1, 0) = C(0, 0, \pm 1) = \frac{\sqrt{6}}{6} + O(\lambda^2),$$

$$C(0, 0, 0) = \frac{\sqrt{6}}{1 - u_0} \lambda,$$

$$C(\pm 1, \pm 1, 0) = C(\pm 1, 0, \pm 1) =$$

$$= C(0, \pm 1, \pm 1) = \frac{\sqrt{6}}{3} (2 + \sqrt{2}) \lambda,$$

$C(\pm 2, 0, 0) = C(0, \pm 2, 0) = C(0, 0, \pm 2) = \frac{\sqrt{6}}{3} \lambda$, where $O(\lambda^2)$ – small corrections of the order of $\sim \lambda^2$, all other $C(l)$ are proportional to $\lambda^2, \lambda^3, \dots$, $\varepsilon = -1 - 2\lambda^2 \left(\frac{3}{1-u_0} + 9 + 4\sqrt{2} \right)$. In this case, the energy

$$E_{11} = E'_g - U_a - \frac{2L^2(k)}{U_a} \left(\frac{3}{1 - u_0} + 9 + 4\sqrt{2} \right). \quad (15)$$

The energy in the second-order approximation,

$$E_{12} = E'_g - U_a - \frac{2L^2(k)}{U_a} (5 + 2\sqrt{2}), \quad (16)$$

corresponds to several states with different symmetries:

a) $C(\pm 1, 0, 0) = C(0, \pm 1, 0) = C(0, 0, \pm 1) = \pm \frac{\sqrt{6}}{6} + O(\lambda^2)$, all other $C(l)$ are proportional to $\lambda, \lambda^2, \dots$, or are zero.

b) $C(\pm 1, 0, 0) = \pm \frac{\sqrt{2}}{2} + O(\lambda^2)$, other $C(l) \sim \lambda, \lambda^2, \lambda^3, \dots$,

c) $C(1, 0, 0) = -iC(0, 1, 0) = -C(-1, 0, 0) = iC(0, -1, 0) = \frac{1}{2} + O(\lambda^2)$,

d) $C(\pm 1, 0, 0) = e^{i\frac{2\pi}{3}} C(0, \pm 1, 0) = e^{i\frac{4\pi}{3}} C(0, 0, \pm 1) = \pm \frac{\sqrt{6}}{6} + O(\lambda^2)$.

The solution of system (13), $C(\pm 1, 0, 0) = -C(0, \pm 1, 0) = \frac{1}{2} + O(\lambda^2)$; $C(0, 0, 0) = C(0, 0, \pm 1) = 0$, $\varepsilon = -1 - 2\lambda^2(3 + \sqrt{2})$, corresponds to the energy

$$E_{13} = E'_g - U_a - \frac{2L^2(k)}{U_a} (3 + \sqrt{2}). \quad (17)$$

The succession of the energy levels $E_{1\alpha}$ (15)–(17) can be different, by depending on u_0 . If $u_0 > 1 + \frac{3}{4}(2 - \sqrt{2})$, then $E_{11} < E_{12} < E_{13}$; if $1 + \frac{1}{2}(2 - \sqrt{2}) < u_0 <$

$1 + \frac{3}{4}(2 - \sqrt{2})$, then $E_{12} < E_{11} < E_{13}$; and if $1 < u_0 < 1 + \frac{1}{2}(2 - \sqrt{2})$, then $E_{12} < E_{13} < E_{11}$.

The method of successive approximations can be used for the zero-approximation energy levels at $|l| = \sqrt{2}, \sqrt{3}, 2, \sqrt{5}, \sqrt{6}, \sqrt{8}, \dots$. Then we get the energy spectra

$$E_{l_1, l_2, l_3} = E'_g - \frac{U_a}{\sqrt{l_1^2 + l_2^2 + l_3^2}} + O(\lambda^2), \quad (18)$$

where $l_i = 1; 2; 3, \dots$, $O(\lambda^2)$ — the small corrections to the zero-approximation energy.

In the case $u_0 < 1$ or $u_0 < 0$, the solutions of system (13) are the same, however the lowest level corresponds to (15), and the energy of a Frenkel-like exciton (14) will be higher, or it does not exist.

We can obtain formula (18) without the usage of the approximation of nearest neighbors for I_{nm}, J_{nm} . The single condition for it is

$$|I_{nm}|, |J_{nm}| \ll U_a. \quad (19)$$

That is, the widths of the conduction and valence bands must be less than the Coulomb energy at the distance $r = a$. For the noncubic crystals, formula (18) will take another form. For the hexagonal lattice, formula (18) becomes

$$E_{l_1, l_2, l_3} = E'_g - \frac{U_a}{\sqrt{l_1^2 + l_2^2 + l_3^2 + l_1 l_2 + l_1 l_3 + l_2 l_3}} + O(\lambda^2), \quad (20)$$

and, for an arbitrary lattice with primitive translation vectors $\vec{a}_1, \vec{a}_2, \vec{a}_3$, it looks as

$$E_{l_1 l_2 l_3} = E'_g - \frac{e^2}{\varepsilon |l_1 \vec{a}_1 + l_2 \vec{a}_2 + l_3 \vec{a}_3|} + O(\lambda^2). \quad (21)$$

Similar multiplet structures of the exciton energy spectra can be revealed in all crystals, in which condition (19) takes place. The value of the splitting in a multiplet is proportional to $(I + J)^2/U_a$. The number of levels in the multiplet and their degeneracy depend on the exciton wave vector k . If $k = 0$ or $|k_1| = |k_2| = |k_3|$, the exciton states possess the higher symmetry, and the number of levels in the multiplet is minimum.

6. Conclusions

1. The account of the crystal lattice discreteness allows us to derive the following new features of large-radius excitons:

- a) Relative motion anisotropy due to the wave vector of an exciton k .

- b) Dispersion law results in a finite value of the exciton band width and its dependence on the principal quantum number n .
 - c) Dependence of the exciton Rydberg constant on k .
2. For small-radius excitons:
- a) The multiplet structure of the energy spectra is obtained in the second order of the successive approximations.
 - b) The parameters of the structure (the number of levels in a multiplet, their degeneracy and symmetry) depend on the wave vector k .
 - c) The value of the splitting in a multiplet is proportional to $(I + J)^2/U_a$.
 - d) The approximate energy spectrum depending on the unit cell geometry is obtained.

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Received 06.01.06

ЕНЕРГЕТИЧНІ СПЕКТРИ ЕКСИТОНІВ З РІЗНИМИ РАДІУСАМИ

М.І. Кислуха, Д.М. Тульчинська

Резюме

Досліджено енергетичні спектри, закони дисперсії, ширини екситонних зон та ефективні маси для екситонів великого та малого радіусів. Врахування дискретності кристалічної ґратки дає можливість виявити нові властивості екситонів: анізотропію відносного руху електрона і дірки, залежність постійної Рідберга екситона та ширини екситонної зони від головного квантового числа n .