Self-localization of excitons in quasi-one-dimensional and quasi-two-dimensional semiconductors

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Exciton-phonon interaction can arise in quasi-one-dimensional and quasi-two-dimensional semiconductors if the electron and hole in the exciton are spatially separated. This interaction is due to the change of the electron-hole Coulomb interaction energy under flexural deformations of the filaments or planes. The energy of the exciton and its effective mass, which can differ by tens of hundreds of times from its bare mass for a number of real values of the coupling constant, are calculated for the regions of strong, weak, and intermediate coupling. These effects are not connected with the formation of electron or hole polarons in the crystal and can take place even under conditions when the interaction of the free electrons and holes with the lattice vibrations is negligibly small.

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INTRODUCTION

It is known that the interaction of an electron with lattice vibrations in dielectrics leads to the appearance of the so-called polaron effects.[1] Polaron effects can be significant also for large-radius excitons in those cases when the electron-phonon interaction essentially renormalizes the effective masses of the electrons and of the holes.[2] In quasi-one-dimensional and quasi-two-dimensional semiconductors, however, a strong interaction between the excitons and the lattice is possible even at a negligibly small constant of interaction between individual quasi particles (electrons and holes) and the lattice.

Indeed, for excitons in which the electron and the hole are localized on different filaments or planes, a unique interaction takes place between the exciton and the lattice, and is due to the change of the Coulomb energy of the electron and hole when these filaments (planes) are deformed. On the other hand, if the electron and hole are localized on the same filament (plane), then the indicated interaction does not take place. This difference between the interactions of the aforementioned excitons with the lattice should lead to a number of singularities. In particular exciton series with relatively narrow and broad lines should coexist in the exciton absorption and emission spectra. It must be borne in mind that the discussed effect of modulation of the energy of the Coulomb interaction of the electron and the hole by the lattice vibrations can be of importance also in three-dimensional semiconductors, provided that the allowed electron and hole energy bands are sufficiently narrow in these semiconductors. From this point of view, a distinguishing feature of quasi-one-dimensional and quasi-two-dimensional semiconductors is precisely the fact that the electron and hole bands are extremely narrow for certain directions, because of singularities in their structure. It is clear, of course, that a similar effect takes place also in quasi-one-dimensional and quasi-two-dimensional metals and semimetals, where the Coulomb interaction of quasiparticles of like sign should also be modulated.[1)]

We confine ourselves in this paper to a discussion of the properties of excitons with spatially-separated electrons and holes. This discussion is timely in connection with the rapidly developing experimental research on the physical properties of quasi-one-dimensional and quasi-two-dimensional semiconductors (see, e.g., [3,4] and the literature cited there). The energy and the effective mass of the exciton are calculated on the basis of Feynman's method of continual integration,[5] which was developed by him in polaron theory. The results take the form of an interpolation that goes over into the results obtained independently in the limits of strong and weak exciton-phonon coupling.

1. EXCITON IN A QUASI-ONE-DIMENSIONAL SYSTEM

When considering the problem of an exciton in which the electron and hole are localized on two different filaments, we choose a coordinate system with origin is halfway between these filaments, x and y axes in the plane of the filaments, and the x axis directed along the filaments.

In a quasi-one-dimensional semiconductor, the interaction at macroscopic distances between an electron and a hole situated on different filaments, takes the form[6]

$$V(x) = -\frac{e^2}{\varepsilon_\perp^{1/2}\varepsilon_\parallel^{1/2}(x^2/\varepsilon_\parallel + d^2/\varepsilon_\perp)^{1/2}}, \tag{1}$$

where $\varepsilon_\parallel = \varepsilon_{xx}$ and $\varepsilon_\perp = \varepsilon_{yy} = \varepsilon_{zz}$ are the principal values of the dielectric tensor, d is the distance between the filaments, and x is the projection of the vector joining the electron and the hole on the direction of the filaments. If the filaments are deformed, then in first order in the site displacements Eq. (1) acquires an increment

$$\frac{\partial V(x)}{\partial d}(u_1 - u_2),$$

where u_1 and u_2 are the displacemements perpendicular to the filaments at the points of the first and second charges. Expanding u_1 and u_2 in the normal coordinates $q_{k\lambda}$, we find that the interaction $\tilde{V}$ between the particles, with allowance for the possible deformation, is given by

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$$\tilde{V}(x)=V(x)+V'(x)\sum_{k,\lambda}\frac{ne_{k\lambda}}{(NM)^{1/2}}q_{k\lambda}\left[\exp\left\{ik_x x_0+ik_y\frac{d}{2}+ik_x\frac{x}{2}\right\}\right.$$
$$\left.-\exp\left\{ik_x x_0-ik_y\frac{d}{2}-ik_x\frac{x}{2}\right\}\right], \qquad (2)$$

where

$$V'(x)=\frac{de^2}{\varepsilon_\perp^{1/2}\varepsilon_\parallel}\left(\frac{d^2}{\varepsilon_\perp}+\frac{x^2}{\varepsilon_\parallel}\right)^{-1/2}.$$

In (2), x_0 is the coordinate of the center of gravity, n is the normal along the y axis, N is the total number of sites, M is the mass of the site, and λ is the number of the phonon branch. We assume that the unit cell contains one site.

Strongly-coupling approximation. There exists a region of parameters in which the strong-coupling approximation of polaron theory is valid. In this case we can obtain the state of the exciton in the deformed crystal, by assuming the deformation to be static.[7] In a quasi-one-dimensional semiconductor, the dielectric tensor can be essentially anisotropic: $\varepsilon_\parallel \gg \varepsilon_\perp$.[3] Then, as shown earlier[8], there exist macroscopic bound states of an electron and a hole with an internal-motion dimension $(\bar{x})^2 \ll d^2\varepsilon_\perp/\varepsilon_\parallel$, and we can therefore put $V'(x)=V'(0)$ in (2). If x_0 is the dimension of the region of motion of the exciton as a unit, then (2) contains $k_x \lesssim 1/x_0$. Assume that $\bar{x} \ll \bar{x}_0$ (this assumption will be confirmed by the final results), then the term $k_x x/2$ in (2) can be neglected and the interaction (2) takes the form

$$\tilde{V}(x)=V(x)+\frac{V_0}{d}\sum_{k,\lambda}\frac{ne_{k,\lambda}}{(NM)^{1/2}}q_{k\lambda}e^{ik_x x_0}2i\sin\frac{k_y d}{2}, \qquad (3)$$

where $V_0=V(x=0)$. We retain in (3) the dependence of $V(x)$ on x, but neglect it in the second term ($V'(x)\to V'(0)=V_0/d$), since it already contains the small factor u_1-u_2. It is seen from (3) that in this approximation the variables of the internal motion and of the motion of the pair as a unit are separated, so that the problem is easily solved.

Let us find the stationary state by starting from a variational principle for the Schrödinger equation. Assuming the electron mass and the hole mass to be different, we write down the functional of the energy in terms of the normalized functions of the relative motion $\chi(x)$ and of the motion of the center of gravity $\varphi(x_0)$:

$$F_1=\int dx_0\,dx\,\varphi^*(x_0)\chi^*(x)\left[-\frac{\hbar^2}{4m}\frac{\partial^2}{\partial x_0^2}-\frac{\hbar^2}{m}\frac{\partial^2}{\partial x^2}+V(x)\right.$$
$$\left.+\frac{V_0}{d}\sum_{k,\lambda}\frac{ne_{k,\lambda}}{(NM)^{1/2}}q_{k\lambda}e^{ik_x x_0}2i\sin\frac{k_y d}{2}\right]\varphi(x_0)\chi(x)+\frac{1}{2}\sum_{k,\lambda}\omega_{k\lambda}^2 q_{k\lambda}^* q_{k\lambda}, \qquad (4)$$

where $\omega_{k\lambda}$ is the phonon frequency.

At $\bar{x}^2 \ll d^2\varepsilon_\parallel/\varepsilon_\perp$ the potential $V(x)$ can be expanded up to terms proportional to x^2, and then the vanishing of the variational derivative with respect to $\chi(x)$

$$\frac{\delta F_1}{\delta\chi(x)}=0$$

leads to an oscillator equation for $\chi(x)$.

The energy of the ground state of the internal motion of the exciton is equal to[8]

$$E_0=-V_0+{}^1/_2\hbar\omega_0,$$
$$V_0=\frac{e^2}{d(\varepsilon_\parallel\varepsilon_\perp)^{1/2}}, \qquad \omega_0^2=\frac{2e^2\varepsilon_\perp^{1/2}}{md^3\varepsilon_\parallel^{3/2}}.$$

Varying with respect to $q_{k\lambda}$, with allowance for the fact that $\varepsilon_{-k\lambda}q_{-k\lambda}=\varepsilon_{k\lambda}q_{k\lambda}^*$, we obtain

$$q_{k\lambda}=\frac{V_0 ne_{k\lambda}}{d\omega_{k\lambda}^2(NM)^{1/2}}2i\sin\frac{k_y d}{2}\left[\int_{-\infty}^{\infty}dx_0 e^{ik_x x_0}|\varphi(x_0)|^2\right]. \qquad (5)$$

We eliminate $q_{k\lambda}$ from (4), and then

$$F_1=E_0+\int dx_0\,\varphi^*(x_0)\left[-\frac{\hbar^2}{4m}\frac{\partial^2}{\partial x_0^2}\right]\varphi(x)$$
$$-\sum_{k,\lambda}\frac{V_0^2|ne_{k\lambda}|^2}{2d^2NM\omega_{k\lambda}^2}4\sin^2\frac{k_y d}{2}\left|\int dx_0 e^{ik_x x_0}|\varphi(x_0)|^2\right|^2. \qquad (6)$$

The last term in (6) can be represented in the form

$$-\int dx\,dx'\,\alpha(x-x')|\varphi(x)|^2|\varphi(x')|^2,$$
$$\alpha(x-x')=\frac{2V_0^2}{d^2NM}\sum_{k,\lambda}\frac{|ne_{k\lambda}|^2\sin^2(k_y d/2)}{\omega_{k\lambda}^2}e^{ik_x(x-x')}.$$

The kernel $\alpha(x-x')$ decreases rapidly with increasing $|x-x'|$, and therefore, recognizing that $\varphi(x)$ varies slowly, we can reduce (6) to the form

$$F_1=\frac{\hbar^2}{4m}\int\left|\frac{\partial\varphi}{\partial x}\right|^2 dx-\alpha\int|\varphi(x)|^4 dx, \qquad (7)$$

where

$$\alpha=\frac{2V_0^2}{d^2NM}\sum_{k,\lambda,k_x=0}\frac{|ne_{k\lambda}|^2}{\omega_{k\lambda}^2}\sin^2\frac{k_y d}{2}=\frac{e^4}{2\pi d^4\rho\varepsilon_\perp\varepsilon_\parallel}B(k_D)\left(\frac{1}{c_l^2}+\frac{1}{2c_t^2}\right),$$
$$B(k_D)=\int_0^{k_D d}\sin^2\frac{x}{2}\frac{dx}{x}.$$

The constant α, in particular, is determined by the phonon spectrum of the crystal. To estimate α we have used the Debye model, where $\omega_{k1}=c_l k$, $\omega_{k2,3}=c_t k$, and $k_D \sim \pi/d$. It is seen that $B(k_D)\approx 1$ and is at any rate weakly dependent on the parameter k_D. This justifies the use of the Debye approximation and means in fact that the details of the phonon spectrum in the region of large k are immaterial in the calculation of α. We note that in the continual model of the lattice, which is not used here,[2)] we have $k_D \to \infty$ and consequently $\alpha \to \infty$, this being due to the divergence of the deformation energy of an infinitesimally thin filament in an elastic continuum. As will be shown below, for excitons in quasi-two-dimensional systems, no such divergence arises, so that in this case it is also possible to use the continual approximation.

The equation $\delta F_1(x)/\delta\varphi(x)=0$ has an exact solution[9]

$$\varphi(x)=\left(\frac{\varkappa}{2}\right)^{1/2}\frac{1}{\operatorname{ch}\varkappa x},$$

where $\varkappa=2\alpha m/\hbar^2$. In this case $F_1=-\alpha^2 m/3\hbar^2$.

To find the translational mass of the new quasiparticle

consisting of the exciton plus the deformation, we shall assume, following Landau and Pekar,[10] that the exciton and deformation move as a unit with velocity v. Taking into account additionally in (4) the kinetic energy of the lattice motion

$$\frac{1}{2}\sum_{\mathbf{k},\lambda}|\dot{q}_{\mathbf{k}\lambda}|^2,$$

we obtain a contribution proportional to v^2, whence

$$m^{**}=2m+\frac{e^4\varkappa^3}{\pi d^2\varepsilon_\perp\varepsilon_\parallel\rho c_t^4}\left(\frac{3}{4}+\frac{c_t^4}{8c_l^4}\right)\ln\frac{1}{\varkappa d}.$$

For the parameter $\varepsilon_\parallel\varepsilon_\perp=100$, $c_l=0.7\cdot10^5$ cm/sec, $c_t=\frac{1}{2}c_l$, $d=6$ Å, and $|F_1|=2.8\cdot10^{-3}$ eV we have $m^{**}\sim200$ m.

Let us make a few remarks concerning the conditions under which the foregoing results are valid. We note first that we have taken into account above only linear distortions of the lattice. This is justified if the displacements u of the filaments are small in comparison with the distances between them. Changing from the normal coordinates (5) to the displacements, we obtain $u/d\sim V_0/d^2\bar{x}_0c_l^2\rho$. For a wide range of parameters and, in particular, for those of them which were used by us, we indeed have $u/d\ll1$.

We note also that, as seen from the foregoing, in our case the wave function and the energy of the obtained state are characterized by only one coupling constant α, although in our problem there are three independent parameters with the dimensions of length: d, $\bar{x}$, and $\bar{x}_0$, and consequently there should be in the general case at least two independent coupling constants characterizing the states of the system. The reason why we are left with only one coupling constant α in our case is that the functional (7) used above, as already emphasized, has been written under the assumption that the radius $\bar{x}$ of the internal motion in the exciton is small in comparison with the dimension $\bar{x}_0$ of the self-localization region. In this limit ($\bar{x}/\bar{x}_0\ll1$) the internal dimension of the exciton along the x axis drops out and the problem reduces to an analysis of the interaction of a structureless particle with the lattice, i.e., to a situation that is described already by only one coupling constant. We note that the inequality $\bar{x}\ll\bar{x}_0$ can also be written in the form $\hbar\omega_0\gg|F_1|$, where ω_0 is the frequency of the internal motion in the exciton. Explicit expressions for F_1 and ω_0 were given above. We can verify that the indicated inequality is indeed satisified for the parameters employed above.

In addition to this inequality, the use of the strong-coupling approximation also calls for satisfaction of the inequality $|F_1|\gg\hbar\omega_D$. This relation, for sufficiently "rigid" lattices, may not take place (these lattices have a low energy $|F_1|$ and, for example for the parameters indicated above, we have $|F_1|\sim\hbar\omega_D$). It is precisely for this reason that we develop in the next section a more general theory of intermediate coupling. This theory will make it possible also to dispense with the already discussed limitation $\bar{x}\ll\bar{x}_0$.

Intermediate coupling. Feynman's path-integral method[5] yields in the entire range of parameters fairly accurate results that go over in limiting cases into the solutions obtained in the strong and weak coupling approximations. If the system energies expressed in terms of the coordinates and their derivatives, then, according to[5], the partition function $\mathrm{Sp}\exp(-\beta H)$ (and consequently the free energy

$$F=-\beta^{-1}\ln\mathrm{Sp}\,e^{-\beta\hat{H}},$$

where β^{-1} is the temperature) can be expressed in the form of a path integral

$$\mathrm{Sp}\,e^{-\beta\hat{H}}=\int e^{-S}Dx_0Dx\,Dq_1Dq_2\ldots, \tag{8}$$

where S is a function of the trajectories in configuration space:

$$S=\frac{1}{\hbar}\int_0^{\beta\hbar}dt\Big[m\dot{x}_0^2(t)+\frac{1}{4}m\dot{x}^2(t)+\frac{1}{4}m\omega_0^2x^2(t)+\frac{1}{2}\sum_{\mathbf{k},\lambda}(|\dot{q}_{\mathbf{k}\lambda}(t)|^2-\omega_{\mathbf{k}\lambda}^2+|\dot{q}_{\mathbf{k}\lambda}|^2)+\frac{2iV_0}{d(NM)^{1/2}}\sum_{\mathbf{k},\lambda}\mathbf{n}e_{\mathbf{k}\lambda}q_{\mathbf{k}\lambda}(t)\,e^{ik_xx_0(t)}\sin\frac{k_yd+k_xx(t)}{2}\Big] \tag{9}$$

Each path starts at the instant of time $t=0$ and terminates at $t=\beta\hbar$ at one and the same point.

The integration over all q_1, q_2, ... was carried out by Feynman in his solution of the polaron problem, and we shall use this integration to eliminate the phonon coordinates. It can be shown that in our case

$$S=\frac{1}{\hbar}\int_0^{\beta\hbar}m\dot{x}_0^2(t)\,dt+\frac{1}{\hbar}\int_0^{\beta\hbar}\left(\frac{m\dot{x}^2(t)}{4}+\frac{m\omega_0^2x^2(t)}{4}\right)dt-\frac{2V_0^2}{d^2}\sum_{\mathbf{k},\lambda}\frac{|\mathbf{n}e_{\mathbf{k}\lambda}|^2}{\omega_{\mathbf{k}\lambda}}\int_0^{\beta\hbar}\int\exp\left[ik_x(x_0(t)-x_0(s))-\omega_{\mathbf{k}\lambda}|t-s|\right]\cdot\sin\frac{k_yd+k_xx(t)}{2}\sin\frac{k_yd+k_xx(s)}{2}\,dt\,ds. \tag{10}$$

It is impossible to integrate further with respect to Dx_0 and Dx, because of the complicated form of the last term of (10). The continual integral can be calculated only if S is an integral of functions in the form $x^2(t)$, $x(t)$, $\dot{x}^2(t)$. Just as in the polaron problem,[5] we use a variational method. In place of an exciton that interacts with a lattice, we consider an exciton in which, besides a direct Coulomb interaction between the electron and the hole, each of the particles interacts with a fictitious particle of mass M_e via the potentials $f(x_e-x_e)^2/2$ and $f(x_h-x_e)^2/2$, where x_e, x_h, x_e are respectively the coordinates of the electron, the hole, and the fictitious particle, while f and M_e are variational parameters.

After eliminating the coordinate x_e, the introduced action takes the form

$$S_0=\frac{1}{\hbar}\int_0^{\beta\hbar}dt\,m\dot{x}_0^2(t)+\frac{1}{\hbar}\int_0^{\beta\hbar}\left(\frac{m\dot{x}^2(t)}{4}+\frac{m\omega_0^2x^2(t)}{4}\right)dt+\frac{C}{2\hbar}\int_0^{\beta\hbar}\int_0^{\beta\hbar}dt\,ds|x_0(t)-x_0(s)|^2e^{-W|t-s|}, \tag{11}$$

where

$$W=(2f/M_e)^{1/4},\quad C=\tfrac{1}{4}M_eW^3,\quad \omega^2=\omega_0^2+f/m.$$

The continual integral with the action S_0 can be calculated exactly, and the difference between S and S_0 is taken into account in the form of a correction

$$F=F_0+\beta^{-1}\langle S-S_0\rangle, \tag{12}$$

$$F_0=\frac{1}{\beta}\ln\int e^{-S_0}Dx_0\,Dx, \tag{13}$$

$$\langle S-S_0\rangle=\int (S-S_0)e^{-S_0}Dx_0\,Dx\Big/\int e^{-S_0}Dx_0\,Dx. \tag{14}$$

Direct calculation of (13) and (14) as $\beta\to 0$ yields

$$F_1=\frac{\hbar(\omega-\omega_0)^2}{4\omega}+\frac{\hbar(V-W)^2}{4V}+E_0-\sum_{\mathbf{k},\lambda}\frac{V_0^2|\mathbf{ne}_{\mathbf{k}\lambda}|^2}{\omega_{\mathbf{k}\lambda}NMd^2}$$
$$\times\int_0^\infty dt\exp\left\{-\omega_{\mathbf{k}\lambda}t-\frac{\hbar k_x^2}{4m}\left[\frac{W^2t}{V^2}+\frac{4C}{WV^3}(1-e^{-tV})\right]\right\}\cdot$$
$$\cdot\left\{\exp\left[-\frac{\hbar k_x^2}{4m\omega}(1-e^{-\omega t})\right]-\cos k_yd\exp\left[-\frac{\hbar k_x^2}{4m\omega}(1+e^{-\omega t})\right]\right\}, \tag{15}$$

where

$$V^2=W^2+4C/W^32m,\quad \omega^2=\omega_0^2+V^2-W^2.$$

It is seen from (15) that a substantial contribution to the integral is made by the times $t\sim 1/\omega_D$, and therefore $k_x^2\lesssim 4m\omega_{\mathbf{k}}V^2/\hbar W^2$. Consequently, the expansion of $V(x)$ in a series in small x is valid for $x^2\ll d^2\varepsilon_\parallel/\varepsilon_\perp$ if

$$\hbar W^2/4m\omega_DV^2\ll d^2\varepsilon_\parallel/\varepsilon_\perp.$$

The inequality is least satisfied in the weak-coupling limit $V^2/W^2\to 1$. For example, at $d=10$ Å and $\omega_D\sim 10^{-2}$ eV, this inequality reduces to the requirement that the anisotropy be sufficient: $\varepsilon_\parallel/\varepsilon_\perp\gg 2$. With increasing coupling force ($V^2/W^2>1$) the series expansion of $V(x)$ becomes more and more justified.

Variation of the functional F_1 was carried out by a numerical method. We have also calculated the exciton translational mass, using Feynman's method.[5] In order of magnitude, the effective mass coincides with M_e. The results are given in Table I.

TABLE I. Energy and effective mass of exciton.

$\varepsilon_\parallel$	3	5	7	10	12	15	20	30
				$d=5$ A				
α_0	2.7	1.6	1.1	0.8	0.7	0.5	0.4	0.3
ω_0	111	75.5	58.7	44.9	39.2	33.1	26.7	19.7
V	69	33	20	11	8	5	3	1
W	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
E	34.8	14.5	8.1	4.3	3.3	2.3	1.5	0.9
m **	58 000	12 000	4000	1000	500	170	50	
				$d=6$ A				
α_0	1.3	0.8	0.6	0.4	0.33	0.26	0.2	0.1
ω_0	84.3	57.5	44.7	34.2	29.8	25.2	20.3	15
V	27	11	6	3	3	2	1	1
W	0.5	0.5	0.5	0.75	1	1.25	1.25	1.25
E	10.5	4.3	2.5	1.5	1.2	0.9	0.7	0.5
m **	7700	1100	260	30	16	5		
				$d=7$ A				
α_0	0.7	0.4	0.3					
ω_0	66.9	45.6	35.4					
V	10	3	2					
W	0.5	0.75	1					
E	3.7	1.7	1.1					
m **	850	30						

Note. The energy is measured in units of $\hbar\omega_D$ ($\omega_D=10^{13}$ sec^{-1}). The mass is measured in units of $2m$ (m is the electron mass): c_l $c_l=0.7\times 10^6$ cm/sec; $\varepsilon_\perp=3$; $c_t/c_l=0.5$; $\rho=1$ g/cm^3; a_0 is the dimensionless coupling constant: $a_0=\frac{1}{4}\pi^{-3/2}(m/\hbar\omega_D)^{1/2}e^4/\rho d^4c_l^2\hbar\varepsilon_\perp\varepsilon_\parallel$.

The transition to the case of strong coupling and of excitons with internal-motion dimension $\bar{x}\ll\bar{x}_0$ is effected in (15) as $\omega_0\to\infty$ and $V\gg W$, ω_D. If $\bar{x}_0\gg d$, then a substantial contribution to the integral (15) is made by small k_x, and it is therefore possible to sum over $\mathbf{k}$, setting $k_x=0$ in $\omega_{\mathbf{k}\lambda}$ and $\mathbf{e}_{\mathbf{k}\lambda}$. As a result, F_1 takes the form

$$F_1=\hbar V/4-(m/\pi\hbar)^{1/4}\alpha V^{1/4}. \tag{16}$$

The minimum of the energy $F_1=-\alpha^2m/\pi\hbar^2$ is reached at $V=4\alpha^2m/\pi\hbar^2$ (α is defined by formula (8)). In this case $W\sim\omega_D$. An exact solution for the functional (7) yields $F_1=-\alpha^2m/3\hbar^2$.[9] If the exciton dimension in the undeformed system is large in comparison with $\bar{x}_0$, corresponding to $\omega_0\ll V$, then we have in the strong-coupling limit $\omega\approx V$ and

$$F_1=\hbar V/2-(mV/2\pi\hbar)^{1/2}\alpha. \tag{17}$$

Comparison of (16) and (17) shows that in the strong coupling approximation the case $\bar{x}\gg\bar{x}_0$ differs from the case $\bar{x}\ll\bar{x}_0$ only in an inessential renormalization of the coupling constant. Thus, allowance for the finite dimension of the internal motion yields $F_1=-m\alpha^2/4\pi\hbar^2$.

We note that when $\omega_0\ll V$ the energy does not depend at all on the frequency ω_0 determined by the direct Coulomb interaction. This might mean that a bound state of two electrons or two holes with $\omega_0^2<0$ is possible. Actually, however, in the presence of a deformation the interaction between identical particles differs only in sign from (1), and it is necessary to add to d the deformation-induced change of the distance $2u(x)\ll d$ between the particles along the y axis. Differentiating $V(x)$ we obtain the force acting between the particles:

$$-\frac{\partial V}{\partial x}=\frac{e^2}{2\varepsilon_\perp\varepsilon_\parallel^{1/2}}\left(\frac{d^2}{\varepsilon_\perp}+\frac{x^2}{\varepsilon_\parallel}\right)^{-3/2}\left(\frac{2d}{\varepsilon_\perp}\frac{\partial u}{\partial x}+\frac{2x}{\varepsilon_\parallel}\right). \tag{18}$$

It is seen therefore that at $\partial u/\partial x<0$ and $|\partial u/\partial x|>|x|\varepsilon_\perp/d\varepsilon_\parallel$ the interaction is of the attraction type. Thus, at a sufficient anisotropy $\varepsilon_\parallel\gg\varepsilon_\perp$, in a certain region x, a more important role in the dependence of the energy on the distance is assumed by the change $(d+2u(x))^2/\varepsilon_\perp$, which leads to an effective attraction between the particles, as compared with $x^2/\varepsilon_\parallel$. Consequently, at a fixed deformation, the energy of interaction between identical particles, as a function of the relative distance, can have a local minimum at $x=0$.

It is important to investigate the stability of the obtained state. Let the deflection of one filament as a result of the deformation be $u(x)$, and that of the second filament $(-u(x))$. Then the elastic energy is defined as the minimum of the functional

$$\frac{1}{2}\sum_{\mathbf{k},\lambda}\omega_{\mathbf{k}\lambda}^2|q_{\mathbf{k}\lambda}|^2$$

under the conditions

$$\sum_{\mathbf{k},\lambda}\frac{\mathbf{ne}_{\mathbf{k}\lambda}}{(NM)^{1/2}}q_{\mathbf{k}\lambda}\exp\left(\pm ik_y\frac{d}{2}+ik_xx\right)=\pm u(x).$$

It can be shown that the energy of a state in which the deformations are shifted relative to one another by $2a$, $(u(x) \to u(x+a),\ -u(x) \to -u(x-a))$ differ from the energy of a state with deformations $u(x)$ and $-u(x)$ by an amount

$$\delta F = -\sum_{k_x} \frac{2u^2(k_x) k_x^2 a^2}{\alpha^2(k_x) - \beta^2(k_x)} \beta(k_x), \tag{19}$$

where

$$\beta(k_x) = \sum_{k_y, k_z, \lambda} \frac{|ne_{k\lambda}|^2 \cos k_y d}{NM\omega_{k\lambda}^2}, \quad \alpha(k_x) = \sum_{k_y, k_z, \lambda} \frac{|ne_{k\lambda}|^2}{NM\omega_{k\lambda}^2}$$

$u(k_x)$ is the Fourier transform of $u(x)$.

Inasmuch as $\alpha^2(k_x) > \beta^2(k_x)$, the sign of δF is determined by the sign of $\beta(k_x)$. If the spectrum of $\omega_{k\lambda}$ is such that the frequency $\omega_{k\lambda}$ increases with increasing k_y when k_x and k_z are constant, then $\beta(k_x) > 0$ and $\delta F < 0$. Thus, for the acoustic spectrum, the obtained state has the maximum elastic energy. It can therefore be stable only if there is a sufficiently strong Coulomb attraction between the particles. In the presence of repulsion, the obtained state is unstable. It is also unstable in the case of attraction if the parameters are such that $\omega_0 \ll V$ (relatively weak attraction, corresponding to $\bar{x}_0 \ll \bar{x}$).

It follows from the considerations advanced above that the only possible states of the exciton-plus-lattice system are those in which the dimension $\bar{x}_0$ of the deformation region is not small in comparison with the bare exciton radius $\bar{x}$. This conclusion is clear, of course, also from pure qualitative reasoning: in a rigid lattice, the deformation radius is large, and as the lattice becomes softer, this radius decreases, but cannot become much smaller than x.

Since the variational procedure used above[3)] does not lead to stable solutions at all values of the initial parameters of the problem (it turns out that $\bar{x}_0 \ll \bar{x}$ in the region of the considered unstable solutions), and stable solutions should exist even for these values of the initial parameters, we are faced with the problem of improving the variational procedure. It appears that such an improvement can be reached, for example, within the framework of a model in which the electron and the hole interact with different fictitious masses. In connection with the foregoing, we shall henceforth confine ourselves precisely to the parameter region in which $\bar{x}_0 > \bar{x}$.

For the case of strong coupling $F_1 \ll \omega_D$ is realized at $V^2 - W^2 \ll \omega_D^2$. In this limit the term $4C(1 - e^{-Vt})/WV^3$ in the exponential of (15) can be neglected. We then have for the energy of the state

$$F_1 = E_0 - \sum_{k\lambda} \frac{(ne_{k\lambda})^2 V_0^2}{\omega_{k\lambda} d^2 NM} \int_0^\infty dt \exp\left(-\omega_{k\lambda} t - \frac{\hbar k_x^2}{4m} t\right) \times \left[\exp\left\{-\frac{\hbar k_x^2}{4m\omega_0}(1 - e^{-\omega_0 t})\right\} - \cos k_y d \exp\left\{-\frac{\hbar k_x^2}{4m\omega_0}(1 + e^{-\omega_0 t})\right\}\right]. \tag{20}$$

The same expression for F_1 can be obtained by perturbation theory.

Weak-coupling approximation. The unperturbed functions of an exciton moving with momentum $\hbar p$ are $\psi_{pn} = L^{-1/2} \exp(ipx_0)\, \varphi_n(x)$ ($\varphi_n(x)$ are the oscillator wave functions $n = 1, 2, \ldots, L$ is the length of the filament). The perturbation is the second term of (2), in which it is necessary to put, for the reason indicated above, $V'(x) = V_0/d$. In second-order perturbation theory, the correction to the energy of an exciton with momentum $\hbar p$ is given by

$$F_1 = \sum_{n, q, \lambda} \frac{V_0^2 \hbar |ne_{q\lambda}|^2}{2NM\omega_{q\lambda} d^2} \frac{|J_n(-q_z)|^2}{\hbar^2 p^2/4m - \hbar^2 (p - q_z)^2/4m - \hbar\omega_{q\lambda} - \hbar n\omega_0}, \tag{21}$$

where

$$J_n(-q_z) = \int dx\, \varphi_n(x) \varphi_0(x) \left[\exp\left\{-\frac{i}{2}(q_z x + k_y d)\right\} - \exp\left\{\frac{i}{2}(q_z x + k_y d)\right\}\right].$$

Direct calculation yields

$$|J_n(-q_z)|^2 = \frac{4(q_z \bar{x})^{2n} f_n(k_y d/2)}{\pi n! 2^{3n}} \exp\left(-\frac{q_z^2 \bar{x}^2}{8}\right),$$

$f_n(z) = \cos^2 z$ if n is odd,
$f_n(z) = \sin^2 z$ if n is even.

The second factor in (21) can be represented in the form

$$|J_n(-q_z)|^2 \int_0^\infty dt \exp\left[-t\left(\frac{\hbar^2 p^2}{2m} - \frac{\hbar^2 (p - q_z)^2}{4m} - \hbar\omega_{q\lambda} - \hbar\omega_0 n\right)\right].$$

Summing over n and putting $p = 0$, we obtain expression (20). Thus, (15) goes over into the strong and weak coupling approximations.

2. QUASI-TWO-DIMENSIONAL SYSTEMS

The foregoing results can be easily generalized to include the case of a system of semiconducting planes. In the strong-coupling approximation, when $\bar{x} \ll \bar{x}_0$, the functional of the energy takes the form (4) with the substitution $k_x \to \mathbf{k}_\parallel$, where $\mathbf{k}_\parallel$, $\mathbf{x}_0$, $\mathbf{x}$ are already two-dimensional vectors lying in the plane of the layers. A functional of the type (6), which was obtained after eliminating the coordinates of the internal motion, cannot be minimized exactly with respect to $\varphi(\mathbf{x}_0)$ in the two-dimensional case. To estimate the minimum we therefore choose a simple form of a normalized trial function $\varphi(\mathbf{x}_0) = (2/\pi)^{1/2} \varkappa e^{-\varkappa|x|}$. The expression for F_2 depends essentially on $\omega_{k\lambda}$. For the Debye spectrum we have

$$F_2 = \hbar^2 \varkappa^2/4m - \beta\varkappa^2, \tag{22}$$

where

$$\beta = e^4 (2\pi d^3 \varepsilon_\parallel^2 \rho c_l^2)^{-1}, \quad \varepsilon_\parallel = \varepsilon_{xx} = \varepsilon_{zz}.$$

The y axis is directed perpendicular to the planes.

It is seen from (22) that for the quasi-two-dimensional systems the localized states are produced if $\beta > \hbar^2/4m$. In this case the energy gain increases monotonically with decreasing dimension of the deformed region, therefore the equilibrium value $\varkappa \gtrsim 1/\bar{x}$ and cannot be obtained within the framework of the employed approach, where it was assumed that $\varkappa\bar{x} \ll 1$. The condition $\beta > \hbar^2 (4m)^{-1}$ is satisfied, for example, at $\varepsilon_\parallel = 3$, $d = 8$ Å, $c_l = 0.7 \cdot 10^5$ cm/sec and $\rho = 1$ g/cm^3. The Feynman continual-integration

method yields in the case of a quasi-two-dimensional system an expression analogous to (15) for the energy F_2, except that the first two terms have an additional factor 2, and the integrand contains $\mathbf{k}_\parallel$ in place of k_x. As seen from the analysis of the strong-coupling approximation, there exists a critical interaction constant such that when the interaction is weaker the change of energy and mass of the exciton, due to the deformation, is small and can be calculated by perturbation theory. On the other hand, if the interaction exceeds the critical value, a strongly localized state is produced, the dimensional of which, as follows from an analysis of the expression for F_2, is determined by the details of the phonon spectrum $\omega_{\mathbf{k}\lambda}$. In particular, for a Debye spectrum, the dimension of the state is of the order of the interatomic dimension. To investigate such states it is therefore necessary to know the actual form of the interaction between the charges at short distances, and the form of the phonon spectrum at short wavelengths.

In quasi-one-dimensional and layered systems, the phonon spectrum is anisotropic and can have singularities due to flexural oscillations of the filaments or the planes.[11,12] In particular, if the elastic constants corresponding to the interaction of the atoms inside one plane are much larger than for the atoms situated in different planes, the spectrum takes the form[11]

$$\omega_{\mathbf{k}}^2 = s^2 k_\parallel^2 + A^2 a^2 k_\parallel^4 + \omega_1^2 \sin^2(dk_y/2), \tag{23}$$

where $s^2 \sim \omega_1^2 d^2$, $A^2 \sim \omega_0^2 a^2$ (a is the lattice constant in the plane), ω_1 is the frequency corresponding to the relative motion of the planes, and ω_0 is the frequency of the flexural oscillations of the plane, with $\omega_1 \ll \omega_0$. At $k_\parallel^2 \gg \omega_1/\omega_0 a^2 \equiv p^2$, the term $\propto k_\parallel^4$ predominates in the spectrum. It follows therefore that the principal role in the deformation of states with dimensions $\bar{x}_0 < 1/p$ is played by flexural deformations, the energy of which increases rapidly with decreasing $\bar{x}_0$ ($\omega_k^2 \sim 1/\bar{x}_0^4$). It is therefore clear that the dimension of the state is determined by the macroscopic parameter p^{-1}. For the spectrum (23), the energy as a function of the variational parameter $\varkappa$ takes the form (22), but $\beta = \beta(\varkappa)$, where

$$\beta(\varkappa) = \frac{2V_0^2}{d^2 NM} \sum_{\mathbf{k}} \frac{64\varkappa^4 \sin^2(k_y d/2)}{[(2\varkappa)^2 + k_\parallel^2]^3 \omega_{\mathbf{k}}^2}. \tag{24}$$

Assuming, for example, $\omega_0 = 10^{13}$ sec^{-1}, $\omega_1 = 3 \cdot 10^{12}$ sec^{-1}, $a = 3$ Å, $d = 5$ Å, and $\varepsilon_\parallel = 3$, we obtain by numerical calculation $\bar{x}_0 \sim 1/p$, $E = -0.01$ eV $\sim \hbar\omega_D$ and $m^{**} = 16\,m$. We note that the use of a spectrum similar to (23) for a quasi-one-dimensional crystal at sufficiently small ω_1 can increase the constant α [Eq. (8)] by several times.

In all the estimates, the exciton bare mass m^* was assumed to be equal to $2m$, where m is the mass of the electron in vacuum. It is seen from the foregoing that the coupling constant increases if the effective bare mass of the exciton is larger than $2m$. It follows from the results that m^{**} depends very strongly on α_0. Therefore a small increase of the exciton mass m^* leads to a substantial increase of m^{**} and E.

[1)] In other words, we have in mind a system Hamiltonian that includes terms representing mutual scattering of two quasiparticles (electrons or holes) with emission or absorption of a phonon, and corresponding to flexural oscillations.

[2)] See the expression for the energy of the elastic deformation of the lattice (last term of (4)).

[3)] As already shown, this procedure is suitable for the parameter region where $\bar{x}_0 \gg \bar{x}$.

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