Bound Electron and Phonon States in a Strong Magnetic Field Due to Nonlinear Electron-Phonon Interaction

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It is shown that in a quantizing magnetic field there exist bound electron and optical phonon states produced by electron-phonon interaction which is quadratic with respect to lattice displacements. The role of this interaction in the bound state problem is evident from the fact that the binding energy is proportional to the square of the quadratic interaction constant and to the fourth power of the linear interaction constant. A complete solution of the problem is obtained for a model Hamiltonian in which the interaction depends only on the momentum transfer. This solution corresponds to a local potential. In addition, an integral equation, which corresponds to a more realistic description of polarization interaction, is also derived and analyzed.

INTRODUCTION

IT has recently been shown[1,2] that the energy spectrum of an electron in a strong magnetic field, interacting with optical phonons, includes a set of levels which can be interpreted as the bound states of an electron and a phonon. These levels are located somewhat below the phonon energy $\hbar\omega_0$, calculated from the bottom of the lowest Landau zone, and are concentrated at this energy, which is the decay threshold. The ordinary Hamiltonian of electron-phonon interaction $H_{int}^{(1)}$ was used in[1,2]: this Hamiltonian is linear in the phonon amplitude u or, what amounts to the same thing, in the creation and annihilation operators of the phonons, b_q^+ and b_q. Here the binding energy W was shown to be proportional to β^4, where β is a dimensionless coefficient which is proportional to the interaction $H_{int}^{(1)}$ (in[1,2] it was assumed that $\alpha \sim \beta^2$). Naturally, along with $H_{int}^{(1)}$, there always exists the interaction $H_{int}^{(2)}$, which is quadratic in u; in most cases, the contribution from $H_{int}^{(2)}$ at low temperatures $T \ll \hbar\omega_0$ is small in comparison with the contribution from $H_{int}^{(1)}$ in terms of the parameter u_0/d, where u_0 is the amplitude of the zero oscillations and d the lattice constant. However, in the problem of the bound states of an electron and a phonon, the comparative roles of the linear and quadratic interactions must be determined differently. The reason for this is the following. The Hamiltonian $H_{int}^{(1)}$ contains b and b^+ linearly and does not conserve the number of phonons. Therefore, in order to obtain the Hamiltonian, by means of which we can consider the dynamical system "electron + single phonon," it is necessary to subject the Hamiltonian of the system "electron + phonon field" to a canonical transformation and, in some approximation, exclude the terms which do not conserve the number of phonons. This can be done, for example, if the coupling is weak, i.e., $H_{int}^{(1)}/\hbar\omega_0 \approx \beta \ll 1$, which will be assumed. After such a transformation, the role of interaction will be played by the term $\tilde{H}_{int}^{(1)}$, which contains b^+b and bb^+ and is proportional to β^2; precisely this term must be equal to $H_{int}^{(2)}$. It is then seen that the role of $H_{int}^{(2)}$ can be very important in the problem of the bound states.

It should be noted that, so far as excitons in molecular crystals are concerned, and also electrons in narrow-band semiconductors, it has already been shown[3] that the change in the vibration frequencies, described by $H_{int}^{(2)}$, makes as important contribution to the coupling energy as a shift in the equilibrium position of the oscillators, which is described by $H_{int}^{(1)}$.

In the following, we shall cinsider only the interaction of $H_{int}^{(2)}$ and shall assume that the terms bb and b^+b^+, which do not conserve the number of phonons, are also eliminated by the canonical transformation, so that the interaction has the form

$$H_{int} = \frac{1}{V} \sum_{\sigma q, \sigma'q'} \gamma_{\sigma'\sigma}(q', q)\, b_{\sigma'q'}^+ b_{\sigma q} e^{i(q-q')r}. \tag{1}$$

Here σ and q are the index of polarization and the momentum of the phonon, r the coordinate of the electron, V the normalized volume, and γ the matrix elements of the interaction. Inasmuch as the Hamiltonian chosen in this fashion strictly conserves the number of phonons, its eigenstates can be classified by the number of phonons.

1. SPECTRUM OF BOUND STATES. THE MODEL HAMILTONIAN

In order to obtain a clear solution of the problem of bound states, we first consider the simplest model, in which the electron interactions with one branch of phonons and, what is more important, γ depends only on the absolute value of the momentum transfer $K = q - q'$:

$$\gamma(q, q') \equiv \gamma(K). \tag{1.1}$$

Then, limiting ourselves to states with one phonon, and changing over to a configurational representation with respect to the phonon coordinates Q, we obtain the interaction in the following form:

$$H_{int} = \frac{1}{V} \sum_K \gamma(K) e^{iKR} \equiv \gamma(R), \tag{1.2}$$

where $R = r - Q$ is the relative coordinate of the electron and phonon.

Assuming that the dispersion of the phonons is unimportant, we have $\omega(q) = \omega_0$, i.e., by actually neglecting the kinetic energy of the phonon, we have the following Schrödinger equation for the electron + phonon system in a magnetic field:

$$\left[\frac{1}{2m}\left(\mathbf{p}-\frac{e}{2c}[\mathbf{H}\mathbf{r}]\right)^2+\hbar\omega_0+\gamma(|\mathbf{r}-\mathbf{Q}|)\right]\Psi(\mathbf{r},\mathbf{Q})=E\Psi(\mathbf{r},\mathbf{Q}),$$
$$(1.3)^*$$

where m is the effective mass of the electron and an axial gauge is chosen for the vector potential. It is convenient to make the gauge transformation

$$\Psi(\mathbf{r},\mathbf{Q})=\exp\left\{-ie[\mathbf{H}\mathbf{Q}]\mathbf{r}/2c\right\}\Psi'(\mathbf{r},\mathbf{Q}),\qquad(1.4)$$

which actually means the choice of the initial origin of the vector potential at the point where there is an immobile phonon. Then Eq. (1.3) is transformed into

$$\left[\frac{1}{2m}\left(\mathbf{P}-\frac{e}{2c}[\mathbf{H}\mathbf{R}]\right)^2+\hbar\omega_0+\gamma(R)\right]\Psi'(\mathbf{r},\mathbf{Q})=E\Psi'(\mathbf{r},\mathbf{Q}),\quad(1.5)$$

where $\mathbf{P}$ is the momentum conjugate to $\mathbf{R}$. It is now seen that the Hamiltonian does not contain $\mathbf{Q}$ and the solution can be chosen in the form

$$\Psi'(\mathbf{r},\mathbf{Q})=\psi(\mathbf{R})\chi(\mathbf{Q}),\qquad(1.6)$$

where χ is an arbitrary function. Thus, the transformation (1.4) allows us to use the translational symmetry and to separate the motion of the mass center, which is identical in the given case with the position of a phonon having infinite mass.

We now regard $\gamma(\mathbf{R})$ as a perturbation, sufficiently small that it does not mix the Landau levels $E_n = \hbar\omega_c(n+\frac{1}{2})$. Furthermore, because of axial symmetry, γ does not mix states with different values of the projection of the angular momentum M, in which these levels are degenerate. We can therefore assume that

$$\psi(\mathbf{R})=\psi_{nM}(\mathbf{R}_\perp)\varphi(Z),\qquad(1.7)$$

where ψ_{nM} is the Landau function and $\mathbf{R}_\perp$ and Z are the transverse and longitudinal (relative to $\mathbf{H}$) components of $\mathbf{R}$. Substituting (1.7) and (1.6) in (1.5), multiplying by $\psi'_{nM}(\mathbf{R}_\perp)$ and integrating over $\mathbf{R}_\perp$, we find the following equation for φ:

$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dZ^2}+\gamma_{nM}(Z)\right]\varphi(Z)=W\varphi(Z),\qquad(1.8)$$

where the role of the potential is played by the quantity

$$\gamma_{nM}(Z)=\int\gamma(R)|\psi_{nM}(\mathbf{R}_\perp)|^2 d\mathbf{R}_\perp\qquad(1.9)$$

and W is the energy measured from threshold:

$$W=E-E_n-\hbar\omega_0\qquad(1.10)$$

The equation for φ is the Schrödinger equation with a shallow one-dimensional well,[4] and therefore has a single bound state (for each nM) with binding energy

$$|W_{nM}|=\frac{m}{2\hbar^2}\left[\int\gamma(R)|\psi_{nM}(\mathbf{R}_\perp)|^2 d\mathbf{R}\right]^2\qquad(1.11)$$

and the wave functions

$$\varphi_{nM}(Z)\sim\exp(-\varkappa_{nM}|Z|),\quad\varkappa_{nM}=(2m|W_{nM}|)^{\frac{1}{2}}.\qquad(1.12)$$

Of course, such a state exists only when the potential γ_{nM} is attractive, i.e., the integral in (1.11) is negative. It is seen from (1.11) that the binding energy depends on the Landau level n at which the electron (in its motion relative to the phonon) is located, and on the of the relative-motion angular momentum M, i.e., the interaction as it were removes the degeneracy in M

*$[\mathbf{H}\mathbf{r}]\equiv\mathbf{H}\times\mathbf{r}$.

existing in the free electron. Each level of the bound state is infinitely degenerate in view of the arbitrariness of the choice of the function $\chi(\mathbf{Q})$. It is seen from the solution thus found that the spatial separation of the electron and the phonon across the field is determined by the magnetic length $\lambda=(c\hbar/eH)^{1/2}$ and its order does not depend on the binding force, while the separation along the field is determined by the length $\varkappa_{nM}^{-1}$ and increases with weakening of the binding force.

We now consider the various types of interaction. We also limit ourselves to the consideration of the lowest Landau level n = 0, where

$$|\psi_{0M}(\mathbf{R}_\perp)|^2=(2\pi\lambda^2 2^M M!)^{-1}\left(\frac{R_\perp}{\lambda}\right)^{2M}\exp\left\{-\frac{1}{2}\left(\frac{R_\perp}{\lambda}\right)^2\right\}.\quad(1.13)$$

Let $\gamma(R)$ be described by the amplitude γ_0 with radius of action a. For the short range potential $a\ll\lambda$, which can simulate the deformation electron-phonon interaction, we can set

$$\gamma(R)=-\gamma_0 a^3\delta(R),\quad\gamma(K)=-\gamma_0 a^3,\qquad(1.14)$$

and then, computing the integral in (1.11), we find

$$|W_{0M}|=\frac{m\gamma_0^2 a^6}{8\pi^2\hbar^2\lambda^4}\delta_{M0},\qquad(1.15)$$

i.e., only bound states with zero angular momentum arise, and $|W_{00}|\sim H^2$. Actually, if we compute the integral in (1.11), without assuming the potential to be delta-shaped, but simply short range, then this integral will be proportional not to $|\psi_{0M}(0)|^2\sim\delta_{M0}$, but to $|\psi_{0M}(a)|^2\sim(a/\lambda)^{2M}$. This means that, there also exist bound states for $M\neq0$, but the binding energy for them contains an extra order of smallness and depends strongly on the magnetic field:

$$|W_{0M}|\approx|W_{00}|(a/\lambda)^{2M}\sim H^{2+M}.\qquad(1.16)$$

For the long-range potential $\lambda\ll a\ll\varkappa^{-1}$, we obviously have

$$\int\gamma(R)|\psi_{0M}(\mathbf{R}_\perp)|^2 d\mathbf{R}\approx\int_{-\infty}^{+\infty}dZ\,\gamma(R)|_{\mathbf{R}_\perp=0}\approx\gamma_0 a.\qquad(1.17)$$

In this case,

$$|W_{0M}|=m\gamma_0^2 a^2/\hbar^2,\qquad(1.18)$$

so that the binding energy is almost independent of M and H.

In order to model the polarization electron-phonon interaction (which does not have a finite radius of action a), we set

$$\gamma(K)=-\gamma_0/K.\qquad(1.19)$$

This yields

$$\gamma(R)=-\frac{\gamma_0}{2\pi^2 R^2},\quad|W_{0M}|=\frac{m\gamma_0^2}{16\pi^2\lambda^2}\left[\frac{\Gamma(M+\frac{1}{2})}{\Gamma(M+1)}\right]^2.\quad(1.20)$$

In this case, the dependence of the binding energy on field is the same for all M, i.e., $|W_{0M}|\sim H$. For $M\gg1$ with increasing M, the last factor in (1.20) decreases like M^{-1} from the value π at M = 0; therefore, the levels cluster about the decay threshold with increasing angular momentum.

It must be noted that the levels of the bound states, which arise from the lowest Landau level n = 0, lie below the emission threshold of the optical phonon and therefore, in the absence of dispersion, are strictly

stationary if we do not take into account the scattering by impurities or acoustic phonons. This consideration does not apply to the levels which arise from the upper Landau zones $n > 0$, so that these levels are quasi-stationary. The amount of broadening of these levels Γ depends on the relation between $H_{int}^{(1)}$ and $H_{int}^{(2)}$. If these interactions are of the same order, then $|W_0 M| \sim \beta^4$ and $\Gamma \sim \beta^2$, i.e., $\Gamma \gg |W|$. If only $H_{int}^{(2)}$ is significant, then $|W| \sim \gamma^2$ and $\Gamma \sim \gamma^2$, i.e., $\Gamma \approx |W|$. The value of γ is computed in the Appendix, for nonpolarization electron-phonon interaction for two-atom crystals of symmetry T_d. Formula (A.5) shows that in this case γ is an imaginary function, odd relative to permutation of q and q', in contrast with the real and even function (1.19). It is therefore necessary to pursue our analysis somewhat further, which is done in the next section.

2. THE SPECTRUM OF BOUND STATES. GENERAL CASE

If the matrix element γ depends materially on both momenta q and q', then the Schrödinger equation in coordinate representation becomes integral in Q and, in contrast with (1.3), it cannot be reduced to an equation with the difference argument R. In this case it is more convenient to use the momentum representation for the phonons. Again, neglecting the dispersions of the phonons and limiting ourselves to states with a single phonon, we obtain the following Schrödinger equation:

$$\left[\frac{1}{2m}\left(p - \frac{e}{c}A(r)\right)^2 + \hbar\omega_0\right]\Psi_\sigma(q, r)$$
$$+ \sum_{\sigma'}\int\frac{dq'}{(2\pi)^3}\gamma_{\sigma\sigma'}(q, q')e^{-i(q-q')r}\Psi_{\sigma'}(q', r) = E\Psi_\sigma(q, r). \quad (2.1)$$

We assume as above that the interaction is weak and does not mix Landau levels with different n. Being interested only in such bound states in which the electron is located at the level $n = 0$, we can represent

$$\Psi_\sigma(q, r) = \int dk_x \psi_{k_x}(r_\perp)\varphi_\sigma(k_x|q, z), \quad (2.2)$$

where $\psi_{k_x}(r_\perp)$ is the transverse part of the wave function of the level $n = 0$ in the Landau gauge. In the general case considered here, when there is no axial symmetry, this gauge is more convenient, since it allows us to use the translational symmetry. Then

$$\frac{1}{2m}\left(p - \frac{e}{c}A\right)^2\Psi_\sigma(q, r) = \left(-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2} + E_\bullet\right)\Psi_\sigma(q, r). \quad (2.3)$$

Substituting (2.2) and (2.3) in (2.1), multiplying by $\psi_{k_x}^*(r_\perp)$ and integrating over $r_\perp$, we find the set of equations for the functions φ. If we use the expression for the integral over $r_\perp$:

$$\int dr_\perp \psi_{k_x'}{}^*(r_\perp)\exp[-i(q_\perp - q_\perp')r_\perp]\psi_{k_x}(r_\perp) = \delta(k_x' - k_x + q_x' - q_x)$$
$$\times \exp\left\{\frac{i}{2}\lambda^2(q_y - q_y')(k_x + k_x') - \frac{\lambda^2}{4}|q_\perp - q_\perp'|^2\right\}, \quad (2.4)$$

then the system takes the following form:

$$-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\varphi_\sigma(k_x|q, z) + \sum_{\sigma'}\int\frac{dq'}{(2\pi)^3}\gamma_{\sigma\sigma'}(q, q')\exp\left\{i\lambda^2(q_y - q_y')k_x\right.$$
$$\left.+ \frac{i}{2}\lambda^2(q_x - q_x')(q_y - q_y')\right\}\exp\left\{-\frac{\lambda^2}{4}|q_\perp - q_\perp'|^2 - i(q_\parallel - q_\parallel')z\right\}.$$
$$\times \varphi_{\sigma'}(k_x + q_x - q_x'|q', z) = W\varphi_\sigma(k_x|q, z). \quad (2.5)$$

We now make the substitution

$$\varphi_\sigma(k_x|q, z) = \exp(i\lambda^2 q_y k_x)\chi_\sigma(k_x + q_x|q, z), \quad (2.6)$$

which also corresponds to

$$\varphi_{\sigma'}(k_x + q_x - q_x'|q', z) = \exp\{i\lambda^2 q_y'(k_x + q_x - q_x')\}\chi_{\sigma'}(k_x + q_x|q', z). \quad (2.7)$$

It is now seen that the functions χ enter into the equation with the same first argument $k_x + q_x$, on which, moreover, the kernel does not depend. Disregarding this unimportant dependence for the present, we obtain the following set of equations for the function $\chi_\sigma(q, z)$:

$$-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\chi_\sigma(q, z) + \sum_{\sigma'}\int\frac{dq'}{(2\pi)^3}\gamma_{\sigma\sigma'}(q, q') \quad (2.8)$$
$$\times \exp\left\{-\frac{\lambda^2}{4}|q_\perp - q_\perp'|^2 + \frac{i}{2}\lambda^2(q_x - q_x')(q_y + q_y')\right\}\cdot$$
$$\times \exp\{-i(q_\parallel - q_\parallel')z\}\chi_{\sigma'}(q', z) = W\chi_\sigma(q, z).$$

The differential equation in z can be regarded as the Schrödinger equation for a shallow one-dimensional well. Therefore, for $W < 0$, we set

$$\chi_\sigma(q, z) = \chi_\sigma(q)e^{-\kappa|z|}, \quad \hbar^2\kappa^2/2m = |W|. \quad (2.9)$$

Integrating, as in the general case[4], over the region $|z| < \kappa^{-1}$ near $z = 0$, we find the set of equations for the functions $\chi_\sigma(q)$:

$$\frac{\hbar^2}{m}\kappa\chi_\sigma(q_\perp, q_\parallel) + \sum_{\sigma'}\int\frac{dq_\perp'}{(2\pi)^2}L_{\sigma\sigma'}(q_\parallel; q_\perp q_\perp')\chi_{\sigma'}(q_\perp', q_\parallel) = 0, \quad (2.10)$$
$$L_{\sigma\sigma'}(q_\parallel; q_\perp q_\perp') = \gamma(q_\perp q_\parallel, q_\perp' q_\parallel)\exp\left\{-\frac{\lambda^2}{4}|q_\perp - q_\perp'|^2\right\}$$
$$\times \exp\left\{\frac{i}{2}\lambda^2(q_x - q_x')(q_y + q_y')\right\}. \quad (2.11)$$

In this set of equations, $q_\parallel$ is a parameter. The quantity κ should be found from the set (2.10) as the eigenvalue (with accuracy to within a factor) of the integral operator L; here κ depends parametrically on $q_\parallel$. To each eigenvalue $\kappa > 0$ there corresponds a bound state. The parameter $q_\parallel$ determines the total longitudinal momentum of the electron + phonon system, since the longitudinal momentum of the electron in a shallow well is of the order of $|2mW|^{1/2}$ and is small. The dependence of κ on $q_\parallel$ thus determines the dependence of the binding energy on the total longitudinal momentum. We note that the dispersion of this nature is lacking in the case of the model potential, for which γ depends only on the momentum transfer $q - q'$.

Each bound state is degenerate in the x component of the total momentum $k_x + q_x$ of the elementary excitation, because, as is clear from the derivation, the functions $\chi_\sigma(q, z)$ can be multiplied by an arbitrary function of $k_x + q_x$. This degeneracy has the same nature as the degeneracy of the Landau levels of the free electron in k_x. We emphasize that the spectrum is not invariant relative to inversion of the longitudinal momentum of the excitation, since both time inversion and space inversion are absent from the problem.

Let us now study in more detail the case of interaction with polarized phonons in two-atom crystals without a center of inversion, for which the matrix elements γ are found in the Appendix. We first consider the longitudinal vibrations. Then, in accord with (A.11),

$$L(q_\parallel; q_\perp q_\perp') = L^*(q_\parallel; q_\perp' q_\perp), \quad (2.12)$$

i.e., the kernel is Hermitian for each $q_{\|}$. It is seen from (A.14) that the kernel is odd:

$$L(-q_{\|}; -\mathbf{q}_{\perp}, -\mathbf{q}_{\perp}') = -L(q_{\|}; \mathbf{q}_{\perp}\mathbf{q}_{\perp}'). \qquad (2.13)$$

It is further seen from (A.5) and (A.7) that L in general does not vanish for $q_{\perp}, q_{\perp}' \to \infty$ if $|q - q_{\perp}'| = \text{const}$. Therefore, the kernel L does not have a Fredholm behavior at infinity and, although judging from its behavior on the diagonal as $|q_{\perp} - q_{\perp}'| \to 0$ it is a kernel with a weak singularity,[5] one can make only limited deductions on the spectrum of this kernel. Inasmuch as the kernel is not degenerate and is Hermitian in accord with (2.12), it has for each q an infinite number of eigenfunctions, and among the corresponding eigenvalues there are always some that are not equal to zero. The bound states correspond to positive $\kappa(q_{\|})$ > 0. In this connection it is very significant that it follows from the oddness of the kernel (cf. (2.13)) that if $\kappa(q_{\|})$ is the eigenvalue of the equation with parameter $q_{\|}$ corresponding to the eigenfunction $\chi(\mathbf{q}_{\perp}, q_{\|})$, then $-\kappa(q_{\|})$ is the eigenvalue of the equation with parameter $-q_{\|}$, which corresponds to the eigenfunction $\chi(-\mathbf{q}_{\perp}, -q_{\|})$. It is then clear that there exist positive κ corresponding to bound states; in particular, such states with $q_{\|} = 0$ always exist.

Unfortunately, because of the non-Fredholm character of the kernel, it is not possible to draw definite conclusions on the number of different eigenvalues and on the structure of the spectrum. Since Eq. (2.10) is two-dimensional (in the plane of $\mathbf{q}_{\perp}$), its eigenfunctions should be determined by two quantum numbers. Therefore the spectrum of values of κ can in principle be both discrete and continuous (in the latter case, κ varies continuously as a function of the additional quantum number).

The dependence of κ on H can be found from considerations of dimensionality. It is seen from (A.5) and (A.7) that γ^l is a homogeneous function of minus first degree in q. Therefore, if we transform in (2.10) to the nondimensional momentum λ_q and divide the equation by γ_0^l, it then becomes clear that the quantity $\hbar^2 \kappa \lambda / m \lambda_0^l$ must be the eigenvalue of the nondimensional kernel, which contains the single parameter $\lambda q_{\|}$. It then follows that the characteristic scale of momentum in the dispersion law is λ^{-1} and the characteristic binding energy is

$$|W| \approx m(\gamma_0')^2 / \hbar^2 \lambda^2. \qquad (2.14)$$

Comparison of (2.14) and Eq. (1.20), obtained by means of the model potential (1.19), shows that both the order of magnitude and the dependence of the binding energy on H are in agreement. However, the mechanism of producing this result is different in the two cases. The transition to the model potential is obtained if we formally set $P^l = \text{const}$ in place of (A.10). Then Eq. (2.10), after separation of the angular variable and completing of the Laguerre transformation,[6] is solved exactly and leads to the results of the previous section. Here it is important that the effective Hamiltonian is obtained as the matrix element that is diagonal in M. On the other hand, for an odd potential (A.10), the matrix elements diagonal over M are annihilated and the binding appears due to the elements that are nondiagonal in M, i.e., due to the mixing of states with different M.

It is not difficult to see that completely analogous arguments are applicable to interactions with transverse phonons. It is only necessary to transpose σ along with the transposition of $q_{\perp}$, and along with the substitution $q_{\perp} \to -q_{\perp}$: to make the substitution $\sigma \to -\sigma$. The homogeneity of the kernel obviously occurs if, in addition to (A.13), we assume that the polarization vectors $e_{\sigma q}$ are identical for all parallel q of a single direction.

3. DISCUSSION OF THE RESULTS

We begin with a general remark. Bound states that arise as the result of quadratic interaction $H_{int}^{(2)}$ have certain differences from those which arise from $H_{int}^{(1)}$, and have been considered in[1,2]. It can be understood in the following way, that a phonon participates in them that was previously in the crystal, while in the case considered earlier, a phonon participates which could be emitted by the electron itself. A reflection of this circumstance is found in the fact that the bound states which arise from linear interaction are found as poles of the single-particle (electron) Green's function,[1,2] while the bound states considered in the present work are found only as the poles of a two-particle (electron + phonon) Green's function. In other words, these bound states do not have any singularities in the electron density of the states for $T = 0$. A great many parameters of states of both types, such, for example, as the dependence of the binding energy on the magnetic field, also differ significantly from one another.

The fundamental question which remains to be discussed is the magnitude of the effect. We begin with unpolarized optical phonons. The potential energy of the electron in the field of a deformed lattice can be written as

$$U(\mathbf{r}) = U^{(1)}(\mathbf{r}) + U^{(2)}(\mathbf{r}) + \ldots = D_1(\mathbf{r})\zeta + \zeta \hat{D}_2(\mathbf{r})\zeta + \ldots, \qquad (3.1)$$

where $\zeta = u/d$ is the dimensionless displacement of the nuclei. Generally speaking, the coefficients have atomic orders of magnitude: $D_1, D_2 \approx D \approx e^2/d \approx 10$ eV. However, D_2 can be anomalously large if there are nearby bands at a distance Δ: it then follows from ordinary perturbation theory that $D_2 \approx D^2/\Delta$. As has been noted in the Appendix, in an estimate of the contribution of the term $U^{(1)}$, it is convenient to exclude, by a canonical transformation, those terms which are linear in ζ. Inasmuch as the energy changes by $\hbar\omega_0$ in a change in the number of phonons by unity, we have in place of $U^{(1)}$ the operator $\zeta \tilde{D}\zeta$ with $\tilde{D} \approx D^2/\hbar\omega_0$. Therefore, $U^{(2)}$ should be taken into account along with $U^{(1)}$ only in the presence of nearby bands with $\Delta \approx \hbar\omega_0$. It must be kept in mind, however, that frequently $U^{(1)} = 0$ from symmetry considerations, while $U^{(2)} \neq 0$ always; under such conditions, the role of $U^{(2)}$ naturally increases.

We now proceed to polarized phonons. As shown in the Appendix, the transverse vibrations, which do not interact with the electrons in the linear approximation, create a macroscopic electric field in the nonlinear approximation, the potential of which field is determined

by Eq. (A.4). As a result, bound states of the electron are produced with transverse optical phonons, lying substantially below the bound states with the participation of longitudinal phonons. It is well known[7] that the additional term $\Phi(E, w)$ determines the nonlinear optical characteristics of the medium in the range of frequencies below the electron transitions. Using experimental data on the coefficient of frequency doubling and the electro-optical effect, we could in principle determine the constants A, B, C, D. Such an attempt was made for CdTe in[8]. However, the indeterminacy in the signs of some of the experimentally determined coefficients, and also the current low level of accuracy of the experiment allow us to determine these constants only with an error of several times the size. Inasmuch as they enter into the binding energy in quadratic form, the possible error in W is unavoidably large. At the same time, from symmetry considerations, they can be estimated as the inverse atomic field $(e/d^2)^{-1}$, which, for $d \approx 5 \times 10^{-8}$ cm, gives A, B, C, D $\approx 10^{-10}$ m/V $\approx 3 \times 10^{-6}$ cgs units and correspond in order of magnitude with the experimental data for nonlinear susceptibility[9] of semiconducting crystals. If we assume $m = 0.1\, m_0$, $H = 100$ kOe, $\hbar\omega_0 = 0.03$ eV, we obtain $W = 10^{-3}\,\hbar\omega_0$ which is clearly outside the possibilities of observation. However, it must be kept in mind that such an estimate is extremely rough;[1] at the same time, an increase of the constant γ_0 by an order of magnitude, which is entirely possible for a "lucky" crystal, gives $W \approx 10^{-1}\,\hbar\omega_0$, which is easily observable. Therefore, we can hope that the bound states can be observed for favorable values of the parameters.

The contribution to the interaction operator containing the operators b^+b^+ and bb can be obtained entirely analogously to (A.4). The interaction constants are close to (A.6). These terms should lead to pinning in experiments of the type described in[10]. Therefore, it is expedient to seek the pinning due to the transverse phonons at the frequency $2\omega_t$. Evidently, pinning can also appear at the frequency $\omega_t + \omega_l$, due to crossed interaction terms. The case is also possible in which the linear interaction of the electron with a definite group of phonons is unpolarized, but the quadratic interaction is polarized. This can occur, for example, in atomic crystals, where the charge on the individual atoms arises only as the result of deformation and is of the order of eu/d. In this case, as can be shown, $\gamma_0 \approx \hbar\omega_0 d^2$; this agrees with the estimate obtained above for polarized transverse phonons.

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APPENDIX

THE HAMILTONIAN OF POLARIZATION ELECTRON-PHONON INTERACTION

If the interaction $H_{int}^{(2)}$ has a polarization character, i.e., if it is produced by macroscopic electric fields,

then it can be expressed in terms of the nonlinear optical characteristics of the crystal in a way similar to that for the linear interaction, where it is expressed in terms of the linear optical characteristics. For this purpose, we use the nonlinear generalization of the Born-Huang theory,[11] which adds to the density potential used there (which is quadratic relative to the displacement of the sublattice w and the electric field E) the "anharmonic" terms $\Phi(E, w)$ with higher powers of w and E. For realization of this program, we write $w = w^0 + w'$ and $E = E^0 + E'$, where w^0 and E^0 are the displacement and the corresponding field for $\Phi = 0$ and w' and E' are small corrections which are due to the "anharmonism." It is obvious that w^0 and E^0 contain a time dependence with the frequencies of the longitudinal and transverse oscillations ω_l and ω_t, and w' and E' are their combinations and harmonics. If we introduce the potential φ of the field E', we can then calculate the amplitude of the potential with frequency ω and wave number K, which will have the form

$$\varphi'_{\omega, K} = -i\frac{4\pi}{\varepsilon(\omega)}\left[\frac{\partial}{\partial E}\Phi(E, w) + \frac{b}{\omega^2 - \omega^2}\frac{\partial}{\partial w}\Phi(E, w)\right]^0_{\omega, K}\frac{K}{K^2}. \quad (A.1)$$

Here $\varepsilon(\omega)$ is the dielectric constant which enters in[11] and $b = (\epsilon_0 - \epsilon_\infty)^{1/2}\,\omega_t/2\pi^{1/2}$, the index 0 above the square bracket indicates that it must be calculated for $w = w^0$ and $E = E^0$; the subscripts denote the fact that the corresponding frequency-time component is taken. Here E^0 can be expressed in terms of w^0 according to the linear theory.

The interaction Hamiltonian of interest to us is $H_{int} = e\varphi'(r, t)$. In order to represent it in the general form, we need to expand w^0 in the normal oscillations:

$$w^0 = \sum_{\sigma q} e_{\sigma q} i\left(\frac{\hbar}{2V\omega_\sigma}\right)^{1/2} b_{\sigma q}(t) e^{iqr} + c.c., \quad (A.2)$$

where $e_{\sigma q}$ is the polarization unit vector, and assume the $b_{\sigma q}(t)$ to be operators. In consideration of examples, we limit ourselves to "anharmonism" of third order in crystals of the T_d class. Then, from symmetry considerations,

$$\Phi(w, E) = A\omega_t^6 b^{-3} w_x w_y w_z + B\omega_t^4 b^{-2}(w_x w_y E_z + \ldots) + C\omega_t^2 b^{-1}(w_x E_y E_z + \cdots) + D E_x E_y E_z. \quad (A.3)$$

The dots indicate terms obtained by cyclic permutation of the principal axes x, y, z; The notation of the coefficients is dictated by the simplicity of the final formulas.

We first consider interaction with transverse phonons only. For this, w^0 is solenoidal and $E^0 = 0$. We find those terms H_{int} which contain bb^+ and b^+b; evidently, these correspond to the static field φ, i.e., $\omega = 0$. From (A.1) and (A.3), we find

$$\varphi'_{0, K} = -i\frac{4\pi}{\varepsilon_0}\frac{\omega_t^4}{b^2}(A + B)\,[W]^0_{0, K}\frac{K}{K^2}; \quad (A.4)$$

$W_x = w_y w_z$ and so on. Expanding w, which enters into W, with the aid of (A.2), where σ means two possible polarizations of the transverse waves, for example, right and left $(\sigma = \pm)$ and $\omega_\sigma = \omega_t$, and also returning to the coordinate representation, we find $e\varphi'$ in the form (I), where

$$\gamma_{\sigma\sigma'}^t(q, q') = \gamma_0^t|q - q'|^{-1}P_{\sigma\sigma'}^t(q, q'). \quad (A.5)$$

Here the constant is

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[1]How great the error can be in such a formal estimate is easily understood if we note that the analogous estimate for the Fröhlich coupling constant gives $\alpha \approx (e^2/d\,\hbar\omega_0)^{1/2} \approx (M/m_0)^{1/4} \sim 10$, while the experimental values for different crystals give $\alpha \approx 10^{-2}$ to 5.

$$\gamma_\bullet{}^t = \frac{(4\pi)^2 e\hbar\omega_t}{\varepsilon_0(\varepsilon_0 - \varepsilon_\infty)}(A + B) \qquad (A.6)$$

and the polarization factor

$$P_{\sigma\sigma'}^t(\mathbf{q}, \mathbf{q}') = \frac{(\mathbf{q} - \mathbf{q}')_z}{2i|\mathbf{q} - \mathbf{q}'|}(e_{\sigma\mathbf{q}, x} e_{\sigma'\mathbf{q}', y}^\bullet + e_{\sigma\mathbf{q}, y} e_{\sigma'\mathbf{q}', x}^\bullet) + \dots \qquad (A.7)$$

Similar considerations for interaction only with longitudinal phonons gives a formula similar to (A.5), where

$$\gamma_0{}^l = \frac{(4\pi)^2 e\hbar\omega_l}{\varepsilon_0(\varepsilon_0 - \varepsilon_\infty)} \frac{\varepsilon_\infty}{\varepsilon_0}(A_1 + B_1); \qquad (A.8)$$

$$A_1 = A - 2B\frac{\varepsilon_0 - \varepsilon_\infty}{\varepsilon_\infty} + C\left(\frac{\varepsilon_0 - \varepsilon_\infty}{\varepsilon_\infty}\right)^2,$$
$$B_1 = B - 2C\frac{\varepsilon_0 - \varepsilon_\infty}{\varepsilon_\infty} + D\left(\frac{\varepsilon_0 - \varepsilon_\infty}{\varepsilon_\infty}\right)^2; \qquad (A.9)$$

$$P^l(\mathbf{q}, \mathbf{q}') = \frac{(\mathbf{q} - \mathbf{q}')_z}{2i|\mathbf{q} - \mathbf{q}'|}(e_{\mathbf{q},x} e_{\mathbf{q}',y}^\bullet + e_{\mathbf{q},y} e_{\mathbf{q}',x}^\bullet) + \dots \qquad (A.10)$$

The following properties of the polarization factors guarantee the Hermitian nature of the interaction:

$$P^l(\mathbf{q}', \mathbf{q}) = P^l(\mathbf{q}, \mathbf{q}')^*, \qquad P_{\sigma'\sigma}^t(\mathbf{q}', \mathbf{q}) = P_{\sigma\sigma'}^t(\mathbf{q}, \mathbf{q}')^*. \qquad (A.11)$$

It is convenient to choose the polarization axes so that, for longitudinal oscillations,

$$e_{-\mathbf{q}} = -e_\mathbf{q}, \qquad (A.12)$$

and for transverse ones,

$$e_{-\sigma, -\mathbf{q}} = -e_{\sigma\mathbf{q}}, \qquad (A.13)$$

where $-\sigma$ means circular polarization counter to the polarization σ. Then the polarization factors can be transformed in inversion in the following fashion:

$$P^l(-\mathbf{q}, -\mathbf{q}') = -P^l(\mathbf{q}, \mathbf{q}'), \qquad P_{-\sigma,-\sigma'}^t(-\mathbf{q}, -\mathbf{q}') = -P_{\sigma\sigma'}^t(\mathbf{q}, \mathbf{q}'). \qquad (A.14)$$

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