ROLE OF ANISOTROPY IN EXCITON TRANSITIONS

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Some features of exciton transitions in a metal model which assumes Fermi surfaces of similar shape for electrons and holes are studied. It is shown in particular that the transition under pressure at T = 0 may involve the appearance of a new period which is not a multiple of the period of the initial lattice.

A few years ago, Keldysh and Kopaev[1] and Kozlov and Maksimov[2] proposed a brilliant model for the metal–dielectric transition corresponding to the production of a bound exciton from an electron and a hole. The model of[1,2] included the assumption of a spherical Fermi surface. The influence of the anisotropy was investigated by Kopaev[3] and by Zittartz[4]. The main conclusion of their results is that anisotropy hinders dielectric pairing. A number of interesting properties that appear in the exciton transition in the presence of anisotropy have gone unnoticed in[3,4]. We investigate here the features of a metal–dielectric transition when the anisotropy parameters are varied, for example under the influence of applied pressure.

We make the following preliminary remarks. The initial premise in[1-6] is that the mechanism producing the transition is the screened Coulomb interaction between the electrons and holes. Such a picture, based on the effective-mass method, is naturally applicable to a material with a small number of carriers. In so far as we know, it is still unclear whether there exist physical objects in which the metal-dielectric transition can be described by the exciton-transition model[1,2]. In particular, in metals of the bismuth group the difference between the electron and hole Fermi surfaces is too large. It seems to us, however, that the question of dielectric pairing is not only of methodological interest alone, since this mechanism may be one of the causes of small gaps and small carrier groups in the electron spectra. One can imagine, for example, the initial metal (neglecting the interaction between the bands) to have two similar large Fermi surfaces of the electron and hole type, spaced half the reciprocal-lattice period apart. The exciton pairing of the two bands leads to a restructuring of the spectrum whereby, generally speaking, anisotropy makes possible the appearance of closed equal-energy surfaces in certain directions. The fact that the number of carriers does not vanish when the spectrum becomes restructured in semimetals in the presence of anisotropy was noted also by Kohn[6].

We describe the lattice by the set of parameters $\{\alpha\}_i$. This set is of course different for different crystalline modifications and is determined by symmetry considerations. Given the external conditions, one of the modifications $\{\alpha\}_{i_0} \neq 0$ is realized. Assume that for $\{\alpha\}_{i_0}$ the Fermi surface has electron and hole Fermi surfaces of identical shape but separated by the vector q_0. The single and double shading in Fig. 1 denotes the multiplicity of the occupation of the electron states in the reciprocal lattice (we disregard spin degeneracy).

The matrix element for the diagram of Fig. 2, which corresponds to scattering of an "electron" from band 1 by a "hole" from band 2, contains the product $G_1(p)G_2(p - q_0)$ of the Green's functions considered in[1,2]. It is easily seen that the existence of a logarithmic contribution from this diagram is not connected with the spherical character of the Fermi surfaces, as was assumed in[1,2], and requires only that the surfaces coincide when they are superimposed.

We shall henceforth attempt, apart from the assumption that the interaction between bands 1 and 2 is weak, not to simplify the models further, our aim being to obtain results that are as general as possible. Following[1,2], we obtain the following integral equation for the determination of the temperature T_{c0}

$$\sigma_{q_0}(\mathbf{p}) = \lambda \left(\sum_0^{\omega/T_{c0}} \frac{2}{n+1/2} \int K(\mathbf{pp}') \frac{dS'}{v_1 + v_2} \sigma_{q_0}(\mathbf{p}') \right), \qquad (1)$$

using in its derivation the fact that the electron spectra in the two bands, reckoned from the normal to the Fermi surfaces, are respectively $\epsilon_1 = v_1 t$ and $\epsilon_2 = -v_2 t$; v_1, v_2, and the interaction $\lambda K(\mathbf{p} \cdot \mathbf{p}')$ depend on the position of the point on the Fermi surface S; $\tilde{\omega}$ is the cutoff of the interaction and is of no importance in what follows ($\tilde{\omega} \sim \epsilon_F$).

Equations of the type (1) with kernel $K(\mathbf{p} \cdot \mathbf{p}')$ were investigated by Pokrovskiĭ[7] in connection with the anisotropic generalization of superconductivity theory. The formal results of[7] are directly applicable to our problem. In particular, in the weak-coupling limit it follows from (1) that T_{c0} is exponentially small. We note also that the dependence of the dielectric gap $\sigma(\mathbf{p})$ on the direction is always determined by a certain function $\chi(\mathbf{p})$, viz., $\sigma(\mathbf{p}) = \sigma\chi(\mathbf{p})$, where $\chi(\mathbf{p})$ is a suitably normalized solution of the homogeneous equation (1) and σ depends only on the temperature.

The dashed lines in Fig. 1 show the deformation of the Fermi surface when $\{\alpha\}_i \neq 0$. Accordingly, the elec-

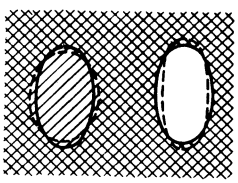

FIG. 1

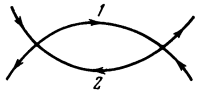

FIG. 2

tron spectra in the first and second bands can be expressed in the form

$$\varepsilon_1 = v_1(t-\eta_1(\mathbf{p})) \text{ and } \varepsilon_2 = -v_2(t-\eta_2(\mathbf{p})),$$

where η_1 and η_2 are small deformations of the Fermi surfaces, reckoned along the normals to the initial surfaces. Obviously, the electroneutrality condition (equality of the numbers of electrons and holes) means that

$$\int dS(\eta_1-\eta_2)=0. \tag{2}$$

A difference between the shapes of the Fermi surfaces decreases the logarithmic contribution to the diagram of Fig. 2, and consequently decreases the temperature T_c of the transition[3,4]. Without giving the details, we write down the general relation from which the transition temperature is determined in the anisotropic case for $\eta_1, \eta_2 \neq 0$:

$$\ln\frac{T_{c0}}{T_c}+\psi\left(\frac{1}{2}\right)-\frac{1}{2}\left[\left\langle\psi\left(\frac{1}{2}+\frac{i(\eta_1-\eta_2)v_1v_2}{2\pi(v_1+v_2)T_c}\right)\right\rangle + \left\langle\psi\left(\frac{1}{2}-\frac{i(\eta_1-\eta_2)v_1v_2}{2\pi(v_1+v_2)T_c}\right)\right\rangle\right]=0, \tag{3}$$

where the angle brackets denote averaging over the Fermi surface:

$$\langle(\ldots)\rangle=\int|\chi(\mathbf{p})|^2\frac{2dS}{v_1+v_2}(\ldots)$$

and the solution $\chi(\mathbf{p})$ of Eq. (1) is assumed to be normalized by the condition

$$\int|\chi(\mathbf{p})|^2\frac{2dS}{v_1+v_2}=1.$$

At small $\eta_1, \eta_2 \ll T_{c0}/v$ we have

$$\frac{\Delta T_c}{T_{c0}}=-\frac{7\zeta(3)}{4\pi^2T_{c0}^2}\left\langle\frac{(\eta_1-\eta_2)^2v_1^2v_2^2}{(v_1+v_2)^2}\right\rangle. \tag{4}$$

At low temperatures we use the representation for the ψ function:

$$\psi(z)=\ln z-\frac{1}{2z}-2\int_0^\infty\frac{t\,dt}{(t^2+z^2)(e^{2\pi t}-1)}.$$

Substituting this expression in (3) and expanding formally as $1/T_c \to \infty$, we obtain

$$\left\langle\ln\frac{|\eta_1-\eta_2|\,2\gamma v_1v_2}{\pi T_{c0}(v_1+v_2)}\right\rangle=\frac{\pi^2T_c^2}{6}\left\langle\left(\frac{v_1+v_2}{v_1v_2}\right)^2\frac{1}{(\eta_1-\eta_2)^2}\right\rangle. \tag{5}$$

We note immediately apropos the right-hand side that the vicinity of the line $\eta_1-\eta_2$ makes no significant contribution to (3) at low temperatures, as can be verified directly. The formally diverging integral in (5) should therefore be taken in the sense of

$$\left\langle(\ldots)\frac{1}{(\eta_1-\eta_2)^2}\right\rangle=\frac{1}{2}\left\langle(\ldots)\left(\frac{1}{(\eta_1-\eta_2-i\delta)^2}+\frac{1}{(\eta_1-\eta_2+i\delta)^2}\right)\right\rangle$$

The fact that the right-hand side of (5) is positive imposes a lower bound on η. In other words, the T_c curve for the exciton transition, as a function of the external parameters $\{\alpha\}_i$, takes the form shown in Fig. 3a. At sufficiently low temperatures the transition may turn out to be of first order with respect to the change in the lattice.

Let us discuss this question in greater detail. Neglecting the interaction between the electrons and phonons, the system energy consists of electron and lattice contributions. We shall consider only the electronic part, or more accurately its variation as a result of changes in the parameters $\{\alpha\}_i$, assuming that the lattice part is automatically specified when $\{\alpha\}_i$ are given. Using Landau's theory of Fermi liquids, we can express the change of the thermodynamic potential $\delta\Omega_e$ as a quadratic functional with respect to the variations of the occupation numbers of the electrons and holes near the Fermi surface (we omit the spin indices, T = 0):

$$\delta\Omega_e=\sum_i\left[\int\varepsilon_{0i}\delta n_i(\mathbf{p})\,d\tau+\frac{1}{2}\int\int f_i(\mathbf{pp}')\delta n_i(\mathbf{p})\,\delta n_i(\mathbf{p}')\,d\tau\,d\tau'\right]. \tag{6}$$

The quantity $\delta\Omega_e/\delta n_i(\mathbf{p}) = \epsilon_i(\mathbf{p})$ has the meaning of the excitation energy reckoned from the chemical potential:

$$\varepsilon_i(\mathbf{p})=\varepsilon_{0i}(\mathbf{p})+c_{ik}(\mathbf{p})\alpha_k+\int f_i(\mathbf{pp}')\delta n_i(\mathbf{p}')\,d\tau'. \tag{7}$$

The first term in (7) equals $v_1|t|$ or $v_2|t|$ for electrons or holes, respectively; the term linear in α_k corresponds to the distortion of the spectrum as a result of the external change of the lattice, and the integral effects takes into account the variation of the self-consistent field and depends therefore on the electron interaction. In accordance with our assumption that the interaction between bands 1 and 2 is small, we omit terms of the f_{12} type from (6) and (7). Allowance for the functions f_i changes the general picture little.

The condition $\epsilon_i(\mathbf{p}) = 0$ determines the equation of the Fermi surface:

$$v_i\eta_i(\mathbf{p})+c_{ik}(\mathbf{p})\alpha_k+(2\pi)^{-3}\int f_i(\mathbf{pp}')\eta_i(\mathbf{p}')\,dS'=0. \tag{8}$$

Equation (8) coincides, as it should, with the condition that $\delta\Omega_e$ be minimal following variation of the shape of the Fermi surface at given $\{\alpha\}_i$, and determines the dependence of $\eta_i(\mathbf{p})$ on $\{\alpha\}_i$. Using (8), we obtain

$$\delta\Omega_e=-\frac{1}{2}\sum_i\left[\int v_i\eta_i^2\frac{dS}{(2\pi)^3}+\int\int f_i(\mathbf{pp}')\eta_i(\mathbf{p})\eta_i(\mathbf{p}')\frac{dS\,dS'}{(2\pi)^6}\right]. \tag{9}$$

This expression should be compared with the energy of the state when pairing is taken into account. Detailed calculations are possible in a concrete model. We shall therefore confine ourselves here to a qualitative discussion of the properties of the thermodynamic quantities in the exciton phase. These are determined by the size of the dielectric gap σ, which in turn is determined by integrals of the off-diagonal thermodynamic functions, which in our case take the form (cf. the equations in[2] and[7]):

$$G_{12}\sim\frac{\chi(\mathbf{p})\sigma}{\omega^2+i\omega(\varepsilon_1+\varepsilon_2)-\varepsilon_1\varepsilon_2+|\chi(\mathbf{p})|^2\sigma^2},\qquad \omega=(2n+1)\pi T. \tag{10}$$

Let us consider for simplicity the case T = 0. To

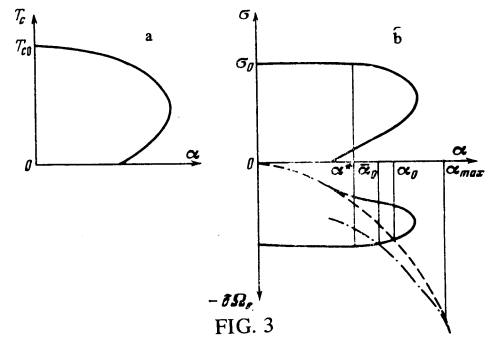

FIG. 3

determine the thermodynamic quantities, we shall therefore need to calculate the integrals of (10) with respect to the frequency and the momentum. We write down the roots in the denominator of (10):

$$\omega_{1,2}=-i\frac{\varepsilon_1+\varepsilon_2}{2}\pm i\left[\left(\frac{\varepsilon_1-\varepsilon_2}{2}\right)^2+|\chi(\mathbf{p})|^2\sigma^2\right]^{1/2}. \qquad (10')$$

The integral of (10) with respect to ω differs from zero if the roots $\omega_{1,2}$ lie in different half-planes, and is equal to

$$2\pi\chi(\mathbf{p})\sigma/((\varepsilon_1-\varepsilon_2)^2+4|\chi(\mathbf{p})|^2\sigma^2)^{1/2}.$$

Substituting here the expressions $\epsilon_1 = v_1(t-\eta_1)+\delta\mu$, and $\epsilon_2 = -v_2(t-\eta_2)+\delta\mu$, for ϵ_1 and ϵ_2, we obtain[1)]

$$2\pi\chi(\mathbf{p})\sigma/\{[(v_1+v_2)t-v_1\eta_1-v_2\eta_2]^2+4|\chi(\mathbf{p})|^2\sigma^2\}^{1/2}. \qquad (11)$$

When integrating with respect to t, it is convenient to make the substitution $u = t-(v_1\eta_1+v_2\eta_2)/(v_1+v_2)$. We see from (11) that the integral with respect to t, and hence the value of σ and all the thermodynamic quantities, does not depend on η so long as the roots ω_1 and ω_2 do not reverse sign in the entire interval of variation of u. Making the substitution $t \to u_1$ in ω_1, we obtain

$$\omega_1=\frac{i}{2}\left\{[(v_1+v_2)^2u^2+4|\chi(\mathbf{p})|^2\sigma^2]^{1/2}-(v_1-v_2)u+2\delta\mu+\frac{v_1v_2(\eta_1-\eta_2)}{v_1+v_2}\right\}.$$

The root ω_1 lies in the upper half-plane for all u, if

$$4|\chi(\mathbf{p})|\sigma>(v_1v_2)^{1/2}\left|\eta_1-\eta_2+2\frac{v_1+v_2}{v_1v_2}\delta\mu\right|. \qquad (13)$$

The condition (13) is violated first at certain points of the Fermi surface. Figure 3b shows schematically the behavior of the dielectric gap and of the thermodynamic potential $\delta\Omega_e$ in the exciton phase; the point α^* corresponds to condition (13). The dashed line corresponds to the quadratic relation (9).

Once condition (13) is violated, carriers of opposite signs appear in certain regions of the reciprocal lattice, inasmuch as the anisotropy can cause the "lower" branch of the energy spectrum in one direction to be higher than the second branch in the other direction. The requirement that the particle number be constant leads to the "electroneutrality" condition, from which $\delta\mu$ is determined. It is easy to verify that the electroneutrality condition reduces to equality of the volumes in phase space, where Im $\omega_1 < 0$ and Im $\omega_2 > 0$.

It follows thus from an examination of the curves of Fig. 3 that at sufficiently low temperatures the transition from the dielectric phase into the metallic phase, following lattice deformation, should be of first order. The actual situation may be more interesting. Let us investigate the stability of the metallic phase relative to formation of a dielectric gap corresponding not to a period q_0, as everywhere in (1) and (3) above, but to a certain $\mathbf{q}_0+\mathbf{q}$, with $\mathbf{q} \ll \mathbf{q}_0$. To this end we consider the matrix element of the diagram of Fig. 2, which contains the Green's function product $G_1(p)G_2(p-q_0-q)$. It is easy to verify that this circumstance leads to substitution $\eta_1-\eta_2 \to \eta_1-\eta_2-\nu q$ in the argument of the ψ-function of Eq. (3), where ν is the cosine of the angle between the vector $\mathbf{q}$ and the vector normal to the Fermi surface at the given point. A corresponding change will take place in the condition that follows from (5) and determines, at T = 0, the limit of the stability of the metallic phase against small fluctuations $\sigma_{q_0+q}(p)$. Recognizing that $\eta_1(\mathbf{p})$ and $\eta_2(\mathbf{p})$ are not changed by the substitution $\mathbf{p} \to -\mathbf{p}$, whereas $\nu(\mathbf{p})$ reverses sign. We can write this condition in the form

$$\left\langle \ln\left\{|(\eta_1-\eta_2)^2-v^2q^2|\left(\frac{2v_1v_2\gamma}{\pi T_{c0}(v_1+v_2)}\right)^2\right\}\right\rangle = 0. \qquad (5')$$

from which we see that the stability limit shifts towards larger η. A qualitative study of this relation shows that there should exist a vector q_{max} that maximizes the deformation of the lattice $\{\alpha\}_i$. At smaller deformations, the normal phase is absolutely unstable against fluctuations of $\sigma_{q_0+q_{max}}(\mathbf{p})$. Figure 4 shows schematically the stability limit of the normal phase in the (α, q) plane.

We now turn again to the question of the singularities of the transition at lower temperatures. At the given conditions, the normal phase is in a definite crystalline modification characterized by the set of parameters $\{\alpha\}_{i_0}$ corresponding to the given symmetry. By varying the pressure, we change the deformation, i.e., we change the quantity α in Fig. 3b (the sign of $\partial\alpha/\partial P$ is of course unknown). Then, as shown above, when α decreases the metallic phase is stable against small perturbations only when $\alpha > \alpha_{max}$, and at the point α_{max} we have absolute instability against infinitesimally small perturbations of $\sigma_{q_0+q_{max}}(\mathbf{p})$. Two possibilities are then conceivable. In the former case α_{max} is smaller than α_0 (the point of intersection of the solid and dashed curves of Fig. 3b). Here α_0 determines the true metastability limit of the normal phase, and the transition from the metallic into the exciton phase is of first order. The second possibility, $\alpha_{max} > \alpha_0$, is shown in Fig. 3b; in this case the transition is of second order, and the resultant new phase is characterized by a certain new reciprocal-lattice vector $\mathbf{q}_0+\mathbf{q}_{max}$. The dash-dot line in Fig. 3b shows the behavior of the electronic part of the energy of the new phase. It is impossible to state in general whether $\alpha_{max} > \alpha_0$ or $\alpha_{max} < \alpha_0$. In the simplest models it seems that $\alpha_{max} > \alpha_0$ always. This takes place, for example, for a spherical Fermi surface at $|\eta_1-\eta_2| = \text{const}$[2)]. The new phase, when produced, is characterized by rather complicated properties. The appearance of finite $\sigma_{q_0+q_{max}}(p)$ near α_{max} means that, in addition to doubling of the period, there appear in the new phase long-wave oscillations of the electron and (or) spin densities ("singlet" and "triplet" pairing according to the terminology of Kozlov and

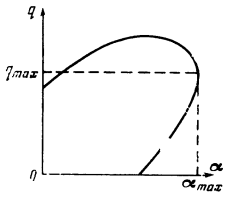

FIG. 4

[1)] $\delta\mu$ is the change of the chemical potential in the new phase.

[2)] The case $\eta_1-\eta_2$ = const denotes, in accord with (2), that the numbers of the electrons and holes are different. This case was considered by Rice [8] in connection with the magnetic properties of chromium alloys.

Maksimov[2]). In the formal sense, $q_{max} \neq 0$ corresponds to helicons of some sort, just as in the case of antiferromagnets (see, e.g., the paper by Dzyaloshinskiĭ[9]). It is important, however, that the electron-density oscillations inevitably restructure the lattice and cause production of a certain resultant period of the structure. We therefore propose that upon further change the transition from the new phase into the ordinary exciton phase occurs jumpwise at the point $\overline{\alpha}_0$ shown in Fig. 3b.

In conclusion, we note the mathematical analogy between the question considered above and the problem of the properties of the superconducting state in the presence of an exchange field[10].

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