Influence of commensurability on the competition between superconducting and insulating states

K. B. Efetov

L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR
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A quasi-one-dimensional system in which superconducting and insulating states compete is considered. A study is made of the influence of commensurability on the nature of the ground state. It is shown that there is a range of vectors $q_1 < q < q_2$, which represent commensurability and are much smaller than the reciprocal lattice period, in which superconductivity is favored. An insulating state is favored for the other values of q. One of the superconductor–insulator transitions is of the first order and the other is of the second order. Variation of q reveals regions with the superconducting order parameters which vary or are constant in space. A comparison is made with the properties of organic conductor $(TMTSF)_2PF_6$ on the assumption that q varies (although slowly) with pressure.

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1. INTRODUCTION

Recent experiments revealed superconductivity of $(TMTSF)_2PF_6$, which is an organic conductor,[1] At zero pressure and temperatures $T < 15$°K the compound $(TMTSF)_2PF_2$ is an insulator. Increase in pressure reduces the temperature of the transition to the insulating state, so that at 12 kbar and 0.9 °K the transition is to the superconducting state. The properties of the conductor in question are strongly anisotropic. In particular, the conductivity ratios are $\sigma_b/\sigma_a \approx 300$ and $\sigma_b/\sigma_c \sim 3 \cdot 10^4$, where the b axis is directed along stacks of TMTSF molecules. Along the a axis the stacks are in direct contact, whereas along the c axis they are separated by the PF_6 chain.[2]

Subsequent experiments have revealed an unusual pressure dependence of the properties of this conductor. The transition from the insulating to the superconducting states in the vicinity of 9 kbar is clearly of the first order, as indicated by the results of the experiments reported by Andres *et al*.[3] and Greene and Engler,[4] who discovered that superconductivity did not disappear when pressure was reduced to 5 kbar. This transition is abrupt without formation of an intermediate (along the pressure scale) metallic state. Quite unexpectedly a further increase in pressure reduces strongly the superconducting transition temperature. Between 12 and 24 kbar this temperature falls from 0.9 to 0.19 °K. One should point out that the pressures applied in these experiments can alter the interatomic distances by just a few percent.[5] Therefore, it is difficult to explain this behavior by a change in the interaction constants.

We shall propose a model in which small changes in a parameter produce a sequence of insulator–superconductor–insulator transitions. We shall assume that electrons can move in a system of metallic chains and the probability of jumps from one chain (filament) to another is not very high. We shall consider the case when electrons are repelled strongly on a given chain but are attracted between different chains. This strong repulsion alters the statistics so that a description in terms of zero-spin fermions is possible. The attraction between electrons in different chains gives rise to a competition between the superconducting and insulating states. The result is very sensitive to the commensurability of the structure period of the insulator with the period of the normal lattice. We shall consider the case corresponding to the experimental situation in $(TMTSF)_2PF_6$, when a band is quarter-filled and there is also a weak potential with a period equal to twice the lattice period.[6]

This potential doubles the unit cell. If the superstructure period is equal exactly to the unit cell period, such commensurability simply favors the insulating state. However, slight deviations from the exact commensurability may initiate a transition to the superconducting state. Further increase in the noncommensurability again makes the insulating state preferable. The theory proposed below can explain the experimental situation if we assume that the insulating structure period varies (though slowly) with pressure.

2. SELECTION OF THE MODEL

We shall now consider a system of conducting chains. We shall assume that electrons can move along chains or jump from chain to chain. We shall postulate that the probability of such jumps is sufficiently low, so that we can use a quasi-one-dimensional description. The Hamiltonian of the system will be written in the form

$$H = \sum_{i,r,\sigma} [J(\mathbf{r})(a_{i\sigma}^+ a_{i+r\sigma} + a_{i+r\sigma} a_{i\sigma} - 2a_{i\sigma}^+ a_{i\sigma}) - \varepsilon_F a_{i\sigma}^+ a_{i\sigma}\delta_{r,0}]$$
$$+ \frac{1}{2}\gamma_0 \sum_{i,\sigma} a_{i\sigma}^+ a_{i\sigma} a_{i-\sigma}^+ a_{i-\sigma} + \frac{1}{2}{\sum_{i,r,\sigma,\sigma'}}' \gamma(\mathbf{r}) a_{i\sigma}^+ a_{i\sigma} a_{i+r\sigma'}^+ a_{i+r\sigma'} + \sum_{i,\sigma} h(\mathbf{r}) a_{i\sigma}^+ a_{i\sigma}, \quad (1)$$

where $a_{i\sigma}^+(a_{i\sigma})$ are the creation and annihilation operators of an electron at a site i with a spin σ; ε_F is the Fermi energy.

The first term in Eq. (1) describes the jumps of electrons from site to site. In accordance with the quasi-one-dimensional nature of the model, it is assumed that $J(\mathbf{r})$ depends strongly on the direction $\mathbf{r}$,

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where $J(\mathbf{b}) \gg J(\mathbf{a})$, $J(\mathbf{c})$, **b** is the direction along the chains, **a** and **c** are the directions across the chains. The quantities $J(\mathbf{a})$ and $J(\mathbf{c})$ can generally be different. The second term in Eq. (1) describes the repulsion of electrons at one site and the third term represents the interaction of electrons at different sites. The last term in Eq. (1) allows for the periodic fields. In accordance with the experimental data on $(TMTSF)_2PF_6$ in Ref. 6 we shall assume that the period $h(\mathbf{r})$ is double the lattice period. For simplicity, we shall retain only the first harmonic in the last term:

$$h(\mathbf{r}) = h_0 \cos(\pi x/a), \tag{2}$$

where a is the lattice period along the conducting chains. The other harmonics are unimportant.

We shall consider below the case of strong repulsion at one center

$$\gamma_0 \gg J(\mathbf{r}), \gamma(\mathbf{r}), h(\mathbf{r}). \tag{3}$$

This strong repulsion at one center has the effect that configurations with two electrons at one site are unlikely. These configurations can be excluded from our analysis by adopting a description in terms of zero-spin fermions. In this case the configurations which are unfavorable from the energy point of view are eliminated by vanishing of the wave functions of these fermions because of the Pauli principle. Allowance for the strong repulsion by a change in the particle statistics was used by Girardeau[7] to describe a Bose gas. The corresponding formulas for a Fermi gas with a strong repulsion at one site were obtained by Ovchinnikov.[8] We shall adopt a similar procedure in the case of the Hamiltonian (1) subject to the conditions (2) and (3); this gives

$$H = \sum_{i,\mathbf{r}} J(\mathbf{r})\,(c_i^+ c_{i+\mathbf{r}} + c_{i+\mathbf{r}}^+ c_i - 2c_i^+ c_i) - \tilde{\varepsilon}_F \sum_{i,\mathbf{r}} c_i^+ c_i + \sum_{i,\mathbf{r}} \gamma(\mathbf{r})\, c_i^+ c_i c_{i+\mathbf{r}}^+ c_{i+\mathbf{r}} + \sum_i h(\mathbf{i})\, c_i^+ c_i, \tag{4}$$

where c_i^+ and c_i are the creation and annihilation operators of zero-spin fermions at a site **i**. The equivalence of the Hamiltonians (1) and (4) implies identity to the eigenstates in the limit of infinitely strong repulsion. The system is then characterized by a strong spin degeneracy which is lifted only if we include the higher orders in J/γ_0.

In the case of zero-spin fermions the dependence of the Fermi momentum on the density differs somewhat from the corresponding dependence in the presence of spin. Simple calculations give

$$2\tilde{p}_F = 2\pi\rho_0/a, \tag{5}$$

where $\tilde{p}_F$ is the Fermi momentum of zero-spin fermions; ρ_0 is the number of fermions per one site (which is identical with the number of electrons); a is the lattice period. The value of the Fermi momentum p_F given by Eq. (5) is, for a given density, twice the Fermi momentum calculated for noninteracting electrons with finite spin.

We shall carry out further calculations for the effective Hamiltonian (4) by considering the interaction of fermions as a perturbation. At sufficiently low temperatures and for low probabilities of jumps from chain to chain we have logarithmically diverging corrections corresponding to the Cooper and Peierls diagrams. The Peierls singularities appear also for sufficiently high probabilities of jumps if the Fermi surface has regions which coincide in the case of parallel transport. The Cooper singularities are insensitive to these jumps.

Summation of logarithmic diagrams can be carried out in the parquet approximation, exactly as this was done by Gor'kov and Dzyaloshinskiĭ.[9] Following their work, we shall separate the interaction constants γ_1^{ik} and γ_2^{ik} corresponding to the forward and backward scattering, where i and k label the chains (filaments). The equations for these vertices will be written in the form

$$\frac{d\gamma_1^{ik}}{d\xi} = -\gamma_1^{ik}\gamma_2^{ik} + \gamma_1^{ik}\gamma_2^{ii} - \frac{1}{2}\sum_l \gamma_1^{il}\gamma_1^{lk}, \qquad \frac{d\gamma_2^{ik}}{d\xi} = -\frac{1}{2}(\gamma_1^{ik})^2, \quad \xi = \ln\frac{\varepsilon_0}{T}. \tag{6}$$

The system (6) differs from the corresponding equations for electrons with finite spin[9] only by the presence of a factor 1/2 in the third term on the right-hand side of the first equation. If there is exactly half an electron per one site, another vertex γ_3 describing jumps appears in the system. However, even in the absence of jumps the system (6) is fairly complex. As in Ref. 9, only a qualitative analysis can be carried out for arbitrary initial conditions. Nevertheless, one may say that for specific initial conditions there should be a competition between the superconducting and insulating states. Electron jumps from chain to chain are important for the existence of such competition. In the absence of jumps the appearance of superconducting solutions is unlikely, since they are described by unstable "standing poles."[9] Clearly, only jumps can make these poles stable. The parquet equations were investigated by Prigodin and Firsov[10] allowing for jumps.

The possibility of competition between the superconducting and insulating states may be studied more clearly by considering only the ladder diagrams of the Cooper and Peierls types. If electrons at different sites attract one another, poles appear in the superconducting and insulating channels. At sufficiently low temperatures we may expect averages $\langle c_i^+(p) c_k^+(-p)\rangle$ and $\langle c_i^+(p) c_k(p - 2\tilde{p}_F)\rangle$, where the indices i and k label the chains, and p is the momentum along the chains. The question which of these states is preferred for given interactions cannot be answered by employing the parquet or self-consistent field approximations. We shall not solve this difficult problem but we shall study a system using a free-energy functional written in the Ginzburg–Landau form and deduced from the above phenomenological considerations.

3. PHENOMENOLOGICAL DESCRIPTION WITH THE AID OF THE GINZBURG–LANDAU FREE ENERGY

It follows from the considerations in the preceding section that a state formed at low temperatures can

be superconducting or insulating. A slight change in the initial parameters, such as the probability of jumps from chain to chain and the density–density interaction, can result in a transition from one state to the other. This situation will be described using the Ginzburg–Landau free energy

$$F=\int\Big[-(A_1|\Delta|^2+A_2|\varkappa|^2)+{}^1/_2B(|\Delta|^2+|\varkappa|^2)^2 + \sum_{i=1}^{3}C_i(|\nabla_i\Delta|^2+|\nabla_i\varkappa|^2)-(\tilde V(x)\varkappa^*+\tilde V^*(x)\varkappa)\Big]d\mathbf{r}, \qquad \tilde V(x)=V_0e^{iqx}. \tag{7}$$

The free energy of Eq. (7) includes two complex order parameters Δ and $\varkappa$, which determine the superconducting and insulating gaps, respectively. In terms of zero-spin fermions, considered in the preceding section, the existence of the order parameters Δ and $\varkappa$ corresponds to pairing described by $\langle c_i(p)c_k(-p)\rangle$ and $\langle c_i^+(p)c_k(p-2\tilde p_F)\rangle$. Using Eq. (6), we can find the order of magnitude of the quantities Δ and $\varkappa$:

$$\Delta=|\gamma|^{-1}\langle c_i(p)c_k(-p)\rangle, \quad \varkappa=|\gamma|^{-1}\langle c_i^+(p)c_k(p-2\tilde p_F)\rangle, \tag{8}$$

where γ is the characteristic magnitude of the interaction of electrons located in different chains and at different sites.

We shall assume that the quantities A_1 and A_2 are very close to one another, so that the following inequality is satisfied:

$$|A_1-A_2|/A_1\ll 1. \tag{9}$$

We shall subsequently assume that the difference between the possibilities of formation of the insulating superconducting states is entirely due to the difference between A_1 and A_2. Consequently, the second and third terms are invariant under the substitution $\Delta\rightleftarrows\varkappa$. This invariance simplified our study of the system but it is not essential to our reasoning.

In principle, the free energy of Eq. (7) may contain a term of the $fB|\Delta|^2|\varkappa|^2$ type. The presence of this term permits simultaneous existence (even in the absence of commensurability effects) of finite order parameters Δ and $\varkappa$, provided f is sufficiently large. However, if

$$f\leqslant(A_1-A_2)/2A_1 \tag{9a}$$

this intermediate phase does not appear. All the calculations given below will be made for the $f=0$ case. When the inequality (9a) is satisfied, the results are not affected qualitatively by the assumption that $f=0$.

The third term contains the sums over all the three directions in a crystal. In the case of a quasi-one-dimensional system we have,

$$C_1\gg C_2, C_3, \tag{10}$$

where C_1 is the direction along the chains, whereas C_2 and C_3 are the two directions which are perpendicular to the conducting chains. The orders of magnitude of the coefficients $A_{1,2}$, B, and C are given by

$$A_{1,2}\sim\frac{T_c-T}{T_c}N(0)\equiv\tau N(0), \qquad B\sim\frac{1}{T_c^2}N(0), \quad C_1\sim\frac{v^2}{T_c^2}N(0). \tag{11}$$

In Eq. (11), we have $T_c\sim\varepsilon_0e^{-1/|\gamma|N(0)}$, where ε_0 is the cutoff energy, $N(0)$ is the density of states, and v is the velocity on the Fermi surface. The coefficients C_2 and C_3 in Eq. (7) depend in a complex manner on the jumps from chain to chain, and on the interaction between different chains. The explicit form of these coefficients is not important to us.

The last term is associated with commensurability effects. In a situation such as that in $(TMTSF)_2PF_6$, when the external field period $h(\mathbf{r})$ is equal to twice the lattice period and the band occupancy is close to 1/4, we find from Eqs. (5) and (8) the correspondence

$$\sum_i h(i)c_i^+c_i=\sum_{p,Q}h(Q)c_{p+Q}^+c_p\to V_0(e^{iqx}\varkappa+e^{-iqx}\varkappa^*), \qquad q=\frac{\pi}{a}(1-2\rho_0), \quad V_0\sim\frac{h_0}{|\gamma|}. \tag{12}$$

It is assumed that $q\ll\pi/a$, which corresponds to an occupancy close to 1/4. Equation (12) is valid if the external field is sufficiently weak, so that the following inequality is satisfied:

$$h_0\ll\gamma T_cN(0)\tau^{3/2}. \tag{13}$$

We can calculate all the thermodynamic quantities using the expression for the free-energy functional (7). We shall assume that C_2 and C_3 are not very small and the temperature T is not very close to the critical value. The physical quantities are then found from the condition for a minimum of the free energy and the contribution of fluctuations is small. Simple estimates demonstrate that the criterion of smallness of the contribution of fluctuations is the inequality

$$\tau\gg\frac{C_1^2}{C_2C_3}\Big(\frac{T_c^2}{v^3N(0)}\Big)^2. \tag{14}$$

The inequality (14) shows that the contribution of fluctuations is small in a wide range of parameters. Moreover, the criterion of smallness of fluctuations is not in conflict with the hypothesis of the possibility of insulating pairing of the Peierls type, since this requires only the existence of parts on the Fermi surface which can be made to coincide by parallel transfer.

It is convenient to introduce a vector Σ with the following four components:

$$\Sigma_1=\varkappa', \quad \Sigma_2=\varkappa'', \quad \Sigma_3=\Delta', \quad \Sigma_4=\Delta'', \tag{15}$$

where $\varkappa'$, Δ' and $\varkappa''$, Δ'' are the real and imaginary parts of the insulating and superconducting order parameters, respectively. When the inequalities (9), (13), and (14) are obeyed, the modulus of the vector Σ is determined exactly by the condition for a minimum of the free energy of Eq. (7) and it is given by

$$\Sigma^2=A_1/B. \tag{16}$$

If the condition (16) is satisfied and the vector Σ is written in the form

$$\Sigma=\mathbf{S}(A_1/B)^{1/2}, \tag{17}$$

where $\mathbf{S}$ is a four-component unit vector defined by

$$S_1=\cos\theta\cos\chi,\quad S_2=\cos\theta\sin\chi,$$
$$S_3=\sin\theta\cos\varphi,\quad S_4=\sin\theta\sin\varphi, \tag{18}$$

we can reduce the free-energy function to

$$F=-\frac{A^2}{2B}+\frac{A_1}{B}\int\Big[\frac{1}{2}\sum_{i=1}^{3}C_i((\nabla_i\theta)^2+\cos^2\theta(\nabla_i\chi)^2+\sin^2\theta(\nabla_i\varphi)^2) - V\cos(qx+\chi)\cos\theta+\frac{\alpha}{2}\sin^2\theta\Big]d\mathbf{r}, \tag{19}$$

$$\alpha=A_1-A_2,\quad V=V_0\left(\frac{B}{A_1}\right)^{1/2}\sim\frac{h_0}{\gamma T_0\tau^{1/2}}. \tag{20}$$

The free energy of Eq. (19) can be rewritten in a simpler form by introducing dimensionless variables

$$x=u(C_1/V)^{1/2},\quad \bar{q}=q(C_1/V)^{1/2},\quad \beta=\alpha/V,$$
$$y=v(C_2/V)^{1/2},\quad z=t(C_3/V)^{1/2}. \tag{21}$$

Substituting Eq. (21) into Eq. (20) and dropping the unimportant constant terms, we obtain

$$F=K_0\int\Big[\frac{1}{2}(\nabla\theta)^2+\frac{1}{2}(\nabla\chi)^2\cos^2\theta+\frac{1}{2}(\nabla\varphi)^2\sin^2\theta - \cos(\bar{q}u+\chi)\cos\theta+\frac{\beta}{2}\sin^2\theta\Big]du\,dv\,dt, \tag{22}$$

$$K_0=\frac{A_1}{B}\left(\frac{C_1C_2C_3}{V}\right)^{1/2}.$$

It is clear from Eq. (22) that a minimum of the free energy is obtained for $\varphi=\text{const}$ and for values of θ and χ dependent on just one coordinate u. Variation of the energy F of Eq. (22) with respect to θ and χ yields the following equations for an extremum

$$-\frac{d^2\theta}{du^2}-\frac{1}{2}\left(\frac{d\chi}{du}\right)^2\sin 2\theta+\cos(\bar{q}u+\chi)\sin\theta+\frac{\beta}{2}\sin 2\theta=0,$$
$$-\frac{d^2\chi}{du^2}\cos\theta+\sin(\chi+\bar{q}u)+2\sin\theta\left(\frac{d\theta}{du}\frac{d\chi}{du}\right)=0. \tag{23}$$

These equations should be supplemented by the condition of vanishing of the gradients on the boundary:

$$\nabla\theta=\cos^2\theta\nabla\chi=0,\quad x\in L. \tag{24}$$

Equations (23) and (24) together with the conditions for an energy minimum describe completely the behavior of the system in question provided we are sufficiently far from a transition point. We shall show below that this behavior depends strongly on the quantity $\bar{q}$. If the solution of Eq. (23) is $\theta=0$, the system has insulating properties. If $\theta\neq 0$, there is nonzero component of the superconducting order parameter and this clearly means that superconductivity appears in the system. In the next two sections we shall study the solutions of Eqs. (23) and (24) for different values of $\bar{q}$.

4. INSULATOR–SUPERCONDUCTOR TRANSITION

So far, we have made no assumptions about the sign of β in Eqs. (22) and (23). In principle, this sign can be positive or negative. However, we shall confine our discussion to the more interesting case $\beta>0$, which corresponds to a situation such that the insulating state is preferred in the absence of commensurability effects. We shall begin with low values of $\bar{q}$. If the superstructure period is identical with the normal lattice period, then $\bar{q}=0$. In this case, the solutions of Eqs. (23) and (24) minimizing the free energy are obvious:

$$\theta=0\quad \chi=0. \tag{25}$$

For nonzero values of $\bar{q}$ the solution begins to depend on the coordinates. It is clear from Eqs. (23) and (24) that the solution with $\theta=0$ always exists. However, in a certain range of $\bar{q}$ this solution does not correspond to the energy minimum. We can show this by expanding the free energy (22) in terms of small deviations θ_1 and χ_1 from the solution $\theta_0=0$ and $\chi_0(u)$ of the system (23). If

$$\theta_0=0, \tag{26}$$

the system (23) reduces to the well-known pendulum equation. This equation describes also a Josephson junction in a magnetic field and many other physical phenomena. If we use the results of Ref. 11, we find that

$$\chi_0=-\bar{q}u \text{ if } \bar{q}_0<4/\pi. \tag{26a}$$

Formally, the solution (26a) does not satisfy the boundary conditions (24) and, therefore, it is valid only sufficiently far from the boundaries. The influence of the boundaries is unimportant if a sample is sufficiently large and we can then use the solution (26a). Expanding the energy (22) near the extremum (26) we obtain

$$F_1=\frac{K_0}{2}\int[(\nabla\theta_1)^2+(\beta+1-\bar{q}^2)\theta_1^2+(\nabla\chi_1)^2+\chi_1^2]du\,dv\,dt. \tag{27}$$

It follows from Eq. (27) that if

$$\bar{q}^2>\bar{q}_c^2=1+\beta, \tag{28}$$

the solution represented by Eqs. (26) and (26a) does not correspond to a free-energy minimum. Our study of the instability is valid in the case of sufficiently small values of β, such that

$$\beta<(4/\pi)^2-1. \tag{29}$$

We shall assume that the condition (29) is satisfied. Instability of the solution with $\theta=0$ in the case when the inequality (28) is satisfied implies a nonzero value of θ. Near q_c the value of θ should be small. Expanding the system (23) near the solution represented by Eqs. (26) and (26a) and retaining terms up to θ^3, we obtain

$$-\frac{d^2\theta}{\partial u^2}-2\bar{q}_c\frac{d^2\theta^2}{\partial u^2}\theta-2\bar{q}_c(\bar{q}-\bar{q}_c)\theta+\frac{1}{2}\theta^3=0. \tag{30}$$

The second term in Eq. (30) is unimportant if $\theta\ll 1$. Therefore, Eq. (30) is simply the Ginzburg–Landau equation. We can see that if $\bar{q}>\bar{q}_c$, there is a nonzero solution independent of the coordinates:

$$\theta_0^2=4\bar{q}_c(\bar{q}-\bar{q}_c). \tag{31}$$

The solution described by Eq. (31) is valid if $\bar{q}-\bar{q}_c\ll 1$. However, a coordinate-independent solution always exists, although it does not always correspond to the absolute minimum of the free energy. In this solution the dependence of χ on the coordinates is described by Eq. (26a). Substituting Eq. (26a) in Eq. (23), we obtain the equilibrium value of θ:

$$\cos\theta_0=(\bar q^2-\beta)^{-1}. \tag{32}$$

The solution (32) exists if $q>q_c$. As in Eq. (31), it is valid provided we are not sufficiently close to a boundary. If $\bar q$ is close to $\bar q_c$, when θ_0 is small, Eq. (32) reduces to Eq. (31). For large values of $\bar q$, the angle θ_0 approaches $\pi/2$. We can show that the solution (32) always corresponds to at least a local energy minimum. We shall do this by expanding the expression for the free energy (22) near the solution represented by Eqs. (26a) and (32) and retain terms up to the squares of deviations θ_1 and χ_1:

$$F=F_0+\frac{K_0}{2}\sum_k[|\theta_k|^2(k^2-\bar q^2\cos 2\theta_0+\cos\theta_0+\beta\cos 2\theta_0) \tag{33}$$
$$+|\chi_k|^2(k^2\cos^2\theta_0+\cos\theta_0)+ik\bar q\,\chi_k\theta_k\sin 2\theta_0],$$
$$F_0=\frac{K_0}{2}\left[\beta-\frac{1}{\bar q^2-\beta}\right]. \tag{34}$$

In Eq. (34), F_0 is the free energy on an extremal trajectory corresponding to Eqs. (26a) and (32). In Eq. (33), θ_k and χ_k are the Fourier components of the deviations; θ_0 is the solution of Eq. (32).

Calculation of the eigenvalues $\lambda_{1,2}$ of the quadratic form (33) yields

$$\lambda_{1,2}=\frac{1}{2}\left\{\frac{1}{\cos\theta_0}+k^2(1+\cos^2\theta_0)\right.$$
$$\left.\pm\left[\left(k^2\sin^2\theta_0+\frac{1}{\cos\theta_0}-2\cos\theta_0\right)^2+4k^2\sin^2 2\theta_0\bar q^2\right]^{1/2}\right\}. \tag{35}$$

A simple analysis of Eq. (35) for moderate values of β shows that both eigenvalues $\lambda_{1,2}$ are always positive. This proves that the solution represented by Eqs. (26a) and (32) always corresponds to at least a local minimum. If the range of small values of $\bar q-\bar q_c$ this minimum is absolute, as can be demonstrated by Eq. (30). We shall show in the next section that, in addition to a solution independent of the coordinates, we can also have solutions which vary in space. If $\bar q$ is sufficiently large, such solutions are preferred for energy reasons. A further increase in $\bar q$ gives rise to a transition back to the insulating state with $\theta=0$.

5. SUPERCONDUCTOR–INSULATOR TRANSITION

In this section we shall consider the case $\beta\ll 1$. We shall show that when this condition is satisfied, the superconductor–insulator transition occurs in the range of large values of the parameter $\bar q\gg 1$. In this range the solution of the system (23) should be found by expanding θ and χ as Fourier series. The ratio of a given term to the preceding one is small if $\bar q$ is large, so that we need retain only the first few terms of each series.

It should be noted that the equations in the system (23) together with the boundary conditions are not affected by the substitutions $\chi\to-\chi$, $u\to-u$. This means that $\theta(u)$ is an even function and $\chi(u)$. Consequently, we shall seek a solution in the form

$$\theta=\bar\theta+\bar a\cos\bar q u+\bar b\cos 2\bar q u+\dots,$$
$$\chi=\bar c\sin\bar q u+\bar d\sin 2\bar q u+\dots. \tag{36}$$

In the main approximation, it is sufficient to include in the system (36) only the zeroth and first harmonics. Substituting Eq. (36) into Eq. (23) and equating the coefficients of the same harmonics, we find that

$$\bar a=-\frac{1}{\bar q^2}\sin\bar\theta,\quad \bar c=-\frac{1}{\bar q^2\cos\bar\theta},$$
$$\sin 2\bar\theta\,(-{}^1/_4\bar q^2\bar c^2+{}^1/_2\beta)-{}^1/_2\bar c\sin\bar\theta+{}^1/_2\bar a\cos\bar\theta=0. \tag{37}$$

Substituting the values of $\bar a$ and $\bar c$ from the first two equations into the third, we obtain the equation for $\bar\theta$

$$\sin 2\bar\theta(\beta-1/2\bar q^2)=0. \tag{38}$$

It follows from Eq. (38) that inclusion of the first harmonic with respect to $1/\bar q^2$ renormalizes the quantity β. Equation (38) then has two solutions:

$$\bar\theta=0,\quad \bar\theta=\pi/2. \tag{39}$$

If $\bar q^2>(2\beta)^{-1}$, an energy minimum corresponds to the first solution, whereas if $\bar q^2<(2\beta)^{-1}$, it corresponds to the second solution. This result means that if $\bar q$ is sufficiently large, the insulating state is again preferred.

The nature of the transition cannot be determined from the first approximation. Calculations in the higher order can be carried out conveniently by substituting the system (36) into the expression for the free energy (22). After integration over space the free energy F becomes a function of $\bar\theta$, $\bar a$, $\bar b$, $\bar c$, and $\bar d$. Variation of F with respect to the parameters $\bar a$, $\bar b$, $\bar c$, and $\bar d$, followed by calculation of the free energy at extremal values, gives

$$F=K_0\Omega\left[\left(\frac{\beta}{2}-\frac{1}{4\bar q^2}-\frac{1}{4\bar q^6}+\frac{\beta}{4\bar q^4}\right)\sin^2\bar\theta\right.$$
$$\left.+\left(\frac{5}{64\bar q^6}-\frac{\beta}{2\bar q^4}\right)\sin^4\bar\theta-\frac{1}{4\bar q^2}+\frac{3}{64\bar q^6}\right], \tag{40}$$

where Ω is the sample volume.

A comparison of the coefficients of the harmonics, which will not be given here explicitly, leads to the conclusion that the approximation employed is valid if

$$1/\bar q^2\cos\bar\theta\ll 1. \tag{41}$$

Therefore, Eq. (40) is valid for values of $\bar\theta$ which are not too close to $\pi/2$. Minimizing Eqs. (40) with respect to $\bar\theta$, we obtain

$$\sin 2\bar\theta\left[\left(\beta-\frac{1}{2\bar q^2}-\frac{1}{2\bar q^6}+\frac{\beta}{2\bar q^4}\right)+\frac{1}{\bar q^4}\left(\frac{5}{16\bar q^2}-2\beta\right)\sin^2\bar\theta\right]=0. \tag{42}$$

Equation (42) always has solutions given by Eq. (39). Strictly speaking, the solution $\bar\theta=\pi/2$ is outside the range of validity of Eqs. (40) and (42). Nevertheless, the exact value of $\bar\theta$ should be close to $\pi/2$. If $\bar q^2$ is sufficiently close to $(2\beta)^{-1}$, we obtain an additional solution:

$$\sin^2\bar\theta=\frac{2}{11\beta^3}\left(\beta-2\beta^3-\frac{1}{2\bar q^2}\right). \tag{43}$$

This solution exists if

$$\bar q_{c1}<\bar q<\bar q_{c2};$$
$$\bar q_{c1}{}^2=1/2\beta+\beta,\quad \bar q_{c2}{}^2=1/2\beta+{}^{15}/_2\beta \tag{44}$$

and corresponds to a maximum.

If the conditions of Eq. (44) are satisfied, then both solutions of Eq. (39) represent minima. In the other cases only one of the solutions of Eq. (39) corresponds to a minimum and the minimum at $\bar{\theta}=0$ exists for $\bar{q}>\bar{q}_{c2}$, whereas the minimum at $\theta=\pi/2$ exists for $\bar{q}<\bar{q}_{c1}$. Simultaneous existence of two minima in the range described by Eq. (44) indicates a first-order phase transition. Moreover, there is a minimum described by Eq. (32), which is discussed in the preceding section.

We shall determine which of these three states is preferred by calculating the free energy at the relevant minima. Using Eqs. (34) and (40), we obtain

$$F=\Omega K_0\begin{cases}-1/4\bar{q}^2+3/64\bar{q}^6, & \bar{\theta}=0\\ -1/2\bar{q}^2+\beta/2-1/8\bar{q}^6-\beta/4\bar{q}^4, & \bar{\theta}=\pi/2.\\ -1/2\bar{q}^2+\beta/2-\beta/2\bar{q}^4, & \cos\theta_0=1/(\bar{q}^2-\beta)\end{cases} \tag{45}$$

It follows from Eq. (44) that if $\bar{q}^2<(2\beta)^{-1}$, the strongly preferred state is the one with $\bar{\theta}=\pi/2$. When $\bar{q}$ is increased, the third state with $\cos\theta_0=(\bar{q}^2-\beta)^{-1}$ becomes preferred. This state should occur in the range

$$q_{c3}=1/2\beta<\bar{q}^2<1/2\beta+{}^{19}/_{8}\beta=q_{c4}. \tag{46}$$

A further increase in $\bar{q}$ has the effect that the lowest energy is exhibited by the state with $\bar{\theta}=0$.

It should be noted that if $\bar{q}^2<(2\beta)^{-1}$, the solution of θ corresponding to a minimum depends on the coordinates, because it contains nonzero harmonics of Eq. (36). Therefore, in this range the magnitude of the superconducting gap varies in space.

The existence of local minima may give rise to metastable states and to superconductivity even in the range where the insulating state is preferred for energy reasons.

All the calculations in the preceding two sections are based on the free-energy functional of Eq. (22) which contains only the phases of the order parameters. This approximation is valid if $q\xi(T)\ll 1$, where $\xi(T)\sim v/T_c\tau^{1/2}$ is the size of an electron pair. If $q\sim q_{c2}$, this inequality is comparable with the condition $\beta\ll 1$ and it imposes restrictions on the proximity of the reduced temperatures of the superconducting τ_2 and insulating τ_1 transitions:

$$\delta^2\ll(\tau_1-\tau_2)/\tau_1\ll\delta,\qquad \delta=h_0/\gamma T_c N(0)\tau^{3/2}. \tag{46a}$$

It follows from the system (46a) that when the inequality (13) is obeyed, there is a range in which all the quantitative results obtained above are valid. The qualitative results on the insulator–superconductor-insulator sequence of transitions is valid in a wider range.

6. CONCLUSIONS

It follows from our investigation that proximity to commensurability may favor superconductivity. The phase diagram depends on the vector q which governs this proximity. An increase of q from zero causes a system under discussion to go over the insulating to the superconducting state and back to the insulating state. In the superconducting state there is a transition from a homogeneous superconductivity with a coordinate-independent order parameter to an inhomogeneous superconductivity whose order parameter varies in space. This can be demonstrated by comparing the solution corresponding to a minimum and given by Eq. (31) in the case when $\bar{q}\sim 1$ with the solution given by Eq. (36) when $\bar{q}\gg 1$. Near a transition an inhomogeneous state clearly represents a soliton lattice. Unfortunately, this explicit form of the solitons representing solutions of the system (23) is not known. A further increase in q again leads to the preference for the homogeneous superconductivity, which is followed by a transition to an insulator. The last two transitions are of the first order. The sequence of the transitions is shown schematically in Fig. 1.

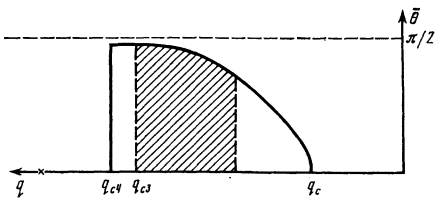

FIG. 1. Dependence of the average value of $\bar{\theta}$ corresponding to a free-energy minimum on the commensurability vector q. In the shaded region the value of θ varies in space. Superconductivity exists in the interval $q_c<q<q_{c4}$.

It is important to stress that all the transitions occur as a result of very small (compared with the reciprocal lattice period) changes in q. We can use Eqs. (11), (20), (21), and (44) to write down readily (in terms of dimensional units) the order of magnitude of the vector q_{c2} corresponding to the last transition to the insulating state:

$$q_{c2}\sim\frac{V}{(C_1\alpha)^{1/2}}\sim p_F\frac{h_0}{\gamma N(0)\varepsilon_F}\frac{1}{\tau_1^{1/2}(\tau_1-\tau_2)^{1/2}}. \tag{47}$$

In Eq. (47), p_F and ε_F represent the Fermi momentum and energy, whereas τ_1 and τ_2 are the reduced temperatures representing the proximity to the temperature of the transitions to the insulating and superconducting states, respectively. The quantity $\gamma N(0)$ is the dimensionless interaction occurring in the exponential function that finds T_c in the system (11).

In the case of the organic conductor $(TMTSF)_2PF_6$ a transition occurs in the vicinity of 15 °K. Therefore, $\gamma N(0)$ should be of the order of unity. If we assume that τ_1 and $\tau_1-\tau_2$ are also of the order of unity, we find that the ratio of q_{c2} to p_F is proportional to the ratio of the external periodic field to the Fermi energy. We have considered above a model with a quarter-filled band in a field h_0 whose period is twice the lattice period. This model was selected to correspond to the properties of $(TMTSF)_2PF_6$. According to Ref. 6, an alternation of periods 3.63 and 3.66 Å occurs along the molecular stacks. This difference between the periods suggests that the ratio h_0/ε_F is of the order of 0.01. It follows from Eq. (47) that the ratio $q_{c2}/p_F\sim 0.01$ is of the same order of magnitude.

The dependence of the nature of the ground state on the commensurability vector q described above explains the alternation of phases in $(TMTSF)_2PF_6$ under pressure if we assume that the pressure can alter the vector q and that at zero pressure the value of q is finite, as denoted by a cross in Fig. 1. If we also assume that an increase in pressure reduces the vector q, then at some pressure the system should go over from the insulating to the superconducting states, and this transition should be of the first order. Metastable superconductivity can then exist well inside the insulating range of conditions. This is in agreement with the experiments reported in Refs. 3–5. A further increase in the pressure should reduce the superconducting gap and at the second critical pressure this gap should vanish.

This behavior is in good qualitative agreement with the experimental results of Refs. 4 and 5, where the pressure dependence of the critical superconducting temperature was determined. Although the transition temperature was not calculated, it was natural to assume that it decreased on reduction in the superconducting order parameter. When the condition

$$\beta \sim \gamma N(0) T_c \tau_1^{1/2} (\tau_1 - \tau_2)/h_0 \ll 1$$

is satisfied, we can expect the superconducting transition temperature T_s found experimentally to be of the same order as T_c. However, if we take the field to be $h_0 \sim 30°K$, as deduced from the observed degree of dimerization and if we also postulate that $\gamma N(0) \sim 1$ as well as τ_1, $\tau_1 - \tau_2 \sim 1$, we find that β is of the order of unity. In this case the maximum value of θ in Fig. 1 should be considerably less than $\pi/2$, which corresponds to a low value of T_s observed experimentally. Superconductivity disappears completely for large values of β.

Clearly, in a substance without band intersection, such as $(TMTSF)_2PF_6$, commensurability cannot depend strongly on pressure. In a purely one-dimensional system free of defects there can be no change in commensurability at all, because this would be in conflict with the Luttinger theorem. However, if the Fermi surface is more complex, then insulating instabilities may appear because of the existence of regions which can be made to coincide by translation by a certain vector **Q** (Ref. 12). In this situation the vector is most likely determined by the properties of the Fermi surface rather than by the lattice. Pressure can distort the Fermi surface and cause changes in the vector **Q** and this can alter slightly the commensurability vector. The pressure dependence of the commensurability vector was observed experimentally for $(TTT)_2I_{3+\delta}$ (Ref. 13), which again does not exhibit band intersection.

The whole of the above discussion is based on a model with a strong repulsion at one center and attraction between different centers. In the zeroth approximation the spin variables corresponding to the strong repulsion are separated from the other variables and the substance exhibits paramagnetic properties. Allowance for the corrections may give rise to an antiferromagnetic ordering with a small antiferromagnetic order parameter. Such ordering was reported in Ref. 14. A strong repulsion gives rise to an insulating pairing with a momentum $4p_F$. In the case of $(TMTSF)_2PF_6$ this momentum is identical with the dimerization field, so that the anomaly in question is difficult to observe. This is in agreement with the results of an experimental study of x-ray scattering,[15] which failed to detect $2p_F$ or $4p_F$ anomalies. The assumption of a strong Coulomb repulsion in $(TMTSF)_2PF_6$ $(TMTSF)_2PF_6$ was made in Ref. 16. The proximity of the system to superconductivity at normal pressure may account for the strong dependence of the resistance on the magnetic field in the vicinity of 20°K (Ref. 1), since superconducting fluctuations should be important in this temperature range. Allowance for fluctuations and a study of thermodynamics should be made in future.

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