New phases in organic superconductors

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(Submitted 3 May 1990)
Pis'ma Zh. Eksp. Teor. Fiz. **51**, No. 11, 583–585 (10 June 1990)

Umklapp processes in the quasi-1D conductors $(TMTSF)_2X$ cause qualitative changes in the phase diagrams of these conductors in a magnetic field. The result is the coexistence of spin density waves characterized by different wave vectors.

The organic superconductors $(TMTSF)_2X$, where $X = ClO_4$, PF_6, have a complicated phase diagram in a magnetic field. The primary distinctive feature of the phase diagram is a cascade of transitions between different spin-density-wave (SDW) subphases.[1-3] There is a related quantum Hall effect.[4,5]

According to Ref. 6, the explanation of the metal-SDW phase transition lies in a "one-dimensionalization" of the electron spectrum in a magnetic field and the appearance of an instability in the "Peierls channels." The insulating subphases which arise in the process are described by an order parameter $\Delta(r)$ which has the form of two plane waves with a quantized value of the longitudinal wave vector:[7,8]

$$\Delta(r) = \Delta_n \exp(ip_x x + i\pi y/b + i\pi z/c) + \text{c.c.}, \tag{1}$$

$$p_x = 2n\omega_c/v_F + 2p_F. \tag{1'}$$

Here ω_c is the cyclotron frequency of the motion of the electrons along unclosed orbits of the quasi-1D electron spectrum,

$$\epsilon(\mathbf{p}) = \pm v_F(p_x \mp p_F) + 2t_b\cos(p_y b) + 2t'_b\cos(2p_y b) + 2t_c\cos(p_z c) \tag{2}$$

in a traverse magnetic field $\mathbf{H}\|\mathbf{Z}$; $\mathbf{p}_F$ and $\mathbf{v}_F$ are the "Fermi" momentum and velocity; t_b, t'_b, and t_c are the overlap integrals of the wave functions across the chains; and n is an integer.

A more detailed study of the $(TMTSF)_2XClO_4$ phase diagram resulted in the discovery of a fine structure of SDW subphases.[9] The explanation[10] of this structure as a consequence of fractional values of n in (1′) does not appear to have a really solid basis, since these states do not correspond to an energy minimum.

In our opinion, a fundamental shortcoming of the theory of an SDW induced by a magnetic field as it exists today is that each subphase is described by means of single value of n in Eq. (1′).

Below we show that umklapp processes, which have heretofore been ignored, lead to the simultaneous existence of SDWs with different values of n. The region near the transition temperature is analyzed separately; in that region, there is the possibility that a hierarchy of subphases can be established and that the most important subphases can be singled out. As a result, the phase diagram would break up into regions of two types, in which either four plane waves or eight exist simultaneously (Fig. 1).

Let us consider the second-order terms in the expansion of the free energy. The existence of a twofold commensurability along the chains, $2p_F = \pi/a^*$, where a^* is the lattice constant, leads to a coupling of the plane waves with $n = n_0$ and $n = -n_0$ from Eq. (1′). The temperature of the transition to the SDW state is determined by the vanishing of the determinant in the matrix equation for the vector order parameter $(\Delta_{n_0}, \Delta^*_{-n_0})$:

$$\begin{vmatrix} 1 - g_2\ln(\Omega/t'_b) - g_z\ln(\omega_c/T)J^2_{n_0}(\lambda), & g_3\ln(\Omega/t'_b) + g_3\ln(\omega_c/T)J^2_{n_0}(\lambda) \\ g_3\ln(\Omega/t'_b) + g_3\ln(\omega_c/T)J^2_{n_0}(\lambda), & 1 - g'_2\ln(\Omega/t'_b) - g'_2\ln(\omega_c/T)J^2_{n_0}(\lambda) \end{vmatrix} \begin{vmatrix} \Delta_{n_0} \\ \Delta^*_{-n_0} \end{vmatrix} = 0, \tag{3}$$

where g_2, g'_2, and g_3 are the electron–electron coupling constants corresponding to ordinary scattering and scattering with an umklapp process, $J_{n_0}(\lambda)$ is the Bessel function of index n_0, $\lambda = 8t'_b/\omega_c$, and Ω is a "cutoff energy."

In the derivation of the matrix elements in (3), use was made of the results of Ref. 6, where the Green's functions in the magnetic field were calculated.

Using $|g_2 - g'_2| < g_3$, we consider the terms of fourth order for states with $\Delta_{n_0} \approx \Delta^*_{-n_0}$ (for definiteness, we assume below that the constant g_3 is greater than 0). It is easy to see that these terms relate *vector* order parameters for different values of n_0 from (3). This coupling near the transition temperature turns out to be effective, however, only for states $(\Delta_{n_0}, \Delta^*_{-n_0})$ and $(\Delta_{n_0+1}, \Delta^*_{-n_0-1})$, so regions corresponding to the coexistence of waves with four different values of n from (1′) appear on the phase diagram (Fig. 1).

We write an expansion of the free energy for these states:

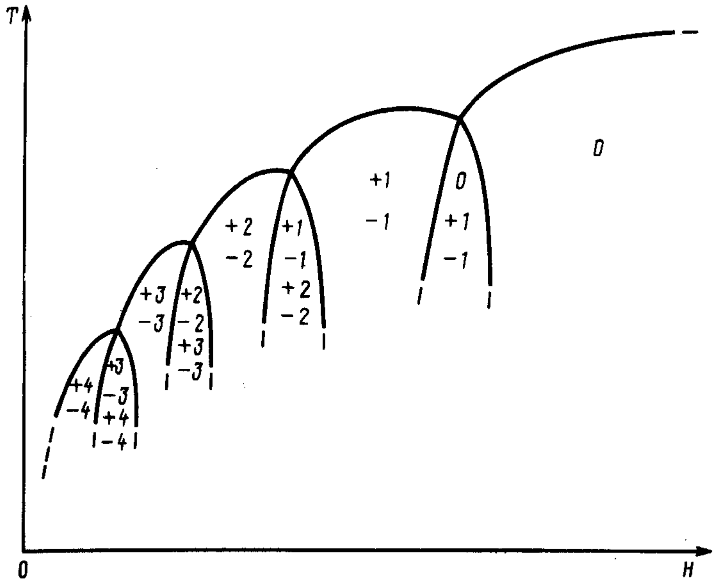

FIG. 1. Fragment of the phase diagram in a magnetic field. The values of the quantum numbers n [see (1′)] for the order parameter of the spin density wave, (1), are given.

$$F=\alpha(g_2+g_3)[(T-T_{n_0})J^2_{n_0}(\lambda)\,|\Delta_{n_0}|^2+(T-T_{n_0+1})J^2_{n_0+1}(\lambda)\,|\Delta_{n_0+1}|^2]$$

$$+\beta(g_2+g_3)^3[6J^4_{n_0}(\lambda)|\Delta_{n_0}|^4+6J^4_{n_0+1}(\lambda)|\Delta_{n_0+1}|^4$$

$$+8J^2_{n_0}(\lambda)J^2_{n_0+1}(\lambda)|\Delta_{n_0}|^2|\Delta_{n_0+1}|^2], \tag{4}$$

The dimensional coefficients α and β, which are common to the different subphases, do not affect the nature of the phase diagram; T_{n_0} is the temperature of the transition to vector state (3).

The "vertex angle" of the region in which plane waves with four different values of n coexist does not depend on the relation between g_2 and g_3. It is determined by minimizing free energy (4); for the phase $(\Delta_{n_0}, \Delta^*_{-n_0}, \Delta_{n_0+1}, \Delta^*_{-n_0-1})$ with $n_0 \neq 0$ it is

$$\frac{2}{3}(T_{n_0+1}-T)<(T_{n_0}-T)<\frac{3}{2}(T_{n_0+1}-T). \tag{5}$$

The case $n_0=0$ is a special one because of the commensurability effect, and the region in which this phase exists is given by a different expression:

$$\frac{2}{3}(T_1-T)<(T_0-T)<(T_1-T) \tag{6}$$

(Fig. 1).

Let us take a brief look at how well these results correspond to the existing experimental data. On the basis of the expansion of the free energy in (4), one can show that the jump in the heat capacity at the transition from the metallic phase to vector state (3) should be smaller by a factor of 1.5 than that in a BCS theory.[7,8] This circumstance agrees well with measurements of the heat capacity in weak magnetic fields.[11]

The branchings which we have found on the phase diagram—branchings of the basic subphases into smaller subphases, which contain eight wave vectors [with four different values of n from (1)] near the transition temperature—agree qualitatively with the experimentally observed breakup of subphases with decreasing temperature.[9]

With regard to the value of the quantum Hall effect in vector states (3), we note that it would again be given by the expression $\rho_{xy} = \hbar/2e^2n_0$, where the sign of n_0 is determined by the component of the vector $(\Delta_{n_0}, \Delta^*_{-n_0})$ which is larger in absolute value.[1)]

I wish to thank L. P. Gor'kov and V. M. Yakovenko for a discussion of this work and for comments.

[1)] I am indebted to V. M. Yakovenko for this nontrivial comment.

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Translated by D. Parsons