Theory of Unusual Anisotropy of Magnetoresistance in Organic Superconductors

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(Received 17 May 1989)

We suggest that weak electron tunneling between layers is responsible for the experimentally observed dramatic deviations from two-dimensional behavior in the magnetoresistance in the metallic phase of the quasi-two-dimensional conductors $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$. We predict a fine structure due to resonances in open orbits between motion parallel and perpendicular to the planes.

PACS numbers: 72.15.Gd, 74.70.Kn

The organic superconductors $(TMTSF)_2X$, $X=ClO_4$, PF_6, ReO_4, have a very complicated phase diagram in a magnetic field perpendicular to the layers. The main feature is a cascade of phase transitions between different spin-density-wave (SDW) states.[1,2] Theoretically, the transitions, and the accompanying quantum Hall effect, have been explained in terms of a two-dimensional model.[3-7] A single SDW opens a band of gaps, and the cascade is caused by successive locking of these Peierls gaps onto the Fermi vector. The two dimensionality has been confirmed, in particular, by observation of a cosine law for the threshold field for the appearance of the SDW phases.[8]

In the *metallic* phase the relation between experiments and theory is more subtle. Indeed, the electron spectrum near the Fermi wave vectors $\pm p_F$ is known to have a quasi-two-dimensional character

$$\epsilon_{1,2}=\pm v_F(p_x\mp p_F)+2t_b\cos(p_y b\sin\gamma\pm\theta)+2t_c\cos(p_z c^*)\,, \tag{1}$$

where the hopping in the b and c directions are $t_b \approx 100$–200 K and $t_c \approx 5$–10 K, respectively.[9,10] However, measurements of the magnetoresistance anisotropy in $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ show dramatic deviations from a cosine law;[11-14] this must necessarily be a three-dimensional effect. This is quite spectacular in view of the tiny value of t_c. In a previous work,[15] one of us showed that in a tilted magnetic field the electron motion in the (b^*,c^*) plane is quasiperiodic, and the two-dimensional limit is reached only for fields for which the cyclotron frequency $\omega_c=eH/mc$ exceeds the bandwidth, $\omega_c \geq 4t_c$, or $H \geq 20$ T. This corresponds to the limit where the amplitude z_0 along c^* becomes smaller than the lattice parameter c^*.

In this Letter we show that the quasiperiodic motion of electrons with spectrum (1) leads to a complicated angular dependence of the magnetoresistance [Figs. 1(a)–1(c)], similar to experiments. Rational relations between the periods of electron motion in the c^* and b^* directions results in resonances and should lead to a fine structure with peaks in the magnetoresistance at well-defined angles. At these values, the electron orbits are one-dimensional Lissajous figures.

Note that the problem of the magnetoresistance of metals with two open, and slightly modulated, sheets of Fermi surface is not trivial. According to the standard theory of metals, the magnetoresistance of open orbit systems is very small, of order $\rho_0(t_b/t_a)^2$, where ρ_0 is the resistance without a magnetic field. In order to explain the large magnetoresistance, Yamaji[16] and Gor'kov[17] studied the magnetic field dependence of the electron-electron scattering in a strictly two-dimensional model, with the magnetic field in the c^* direction perpendicular to the plane [see Fig. 2(a)]. In addition to generalizing the mechanism to three dimensions, we also consider the interaction of electrons with impurities [Fig. 2(b)]. Both diagrams have been investigated by Gor'kov and Dzyaloshinsky[18] in the absence of a magnetic field. The magnetoresistance is given by the imaginary part of the self-energy of the electron Green's function:

$$\mathrm{Im}\Sigma_a=g^2T\int_0^\pi dq\int_0^\pi dQ\int_0^\infty dx\,\frac{2\pi T/v_F}{\sinh^2(2\pi Tx/v_F)}\left(\frac{2\pi Tx/v_F}{\exp(4\pi Tx/v_F)-1}+\frac{2\pi Tx/v_F-1}{2}\right)\times J_0^2\left[\frac{2\lambda_1}{\cos\varphi}\sin\left(\frac{x\cos\varphi}{x_H}\right)\cos q\right]J_0^2\left[\frac{2\lambda_2}{\sin\varphi}\sin\left(\frac{xc^*\sin\varphi}{bx_H\sin\gamma}\right)\cos Q\right] \tag{2a}$$

and

$$\mathrm{Im}\Sigma_b=\frac{|g|}{\tau_0}\int_0^\pi dq\int_0^\pi dQ\int_d^{v/T}\frac{dx}{x}J_0^2\left[\frac{2\lambda_1}{\cos\varphi}\sin\left(\frac{x\cos\varphi}{x_H}\right)\cos q\right]J_0^2\left[\frac{2\lambda_2}{\sin\varphi}\sin\left(\frac{xc^*\sin\varphi}{bx_H\sin\gamma}\right)\cos Q\right], \tag{2b}$$

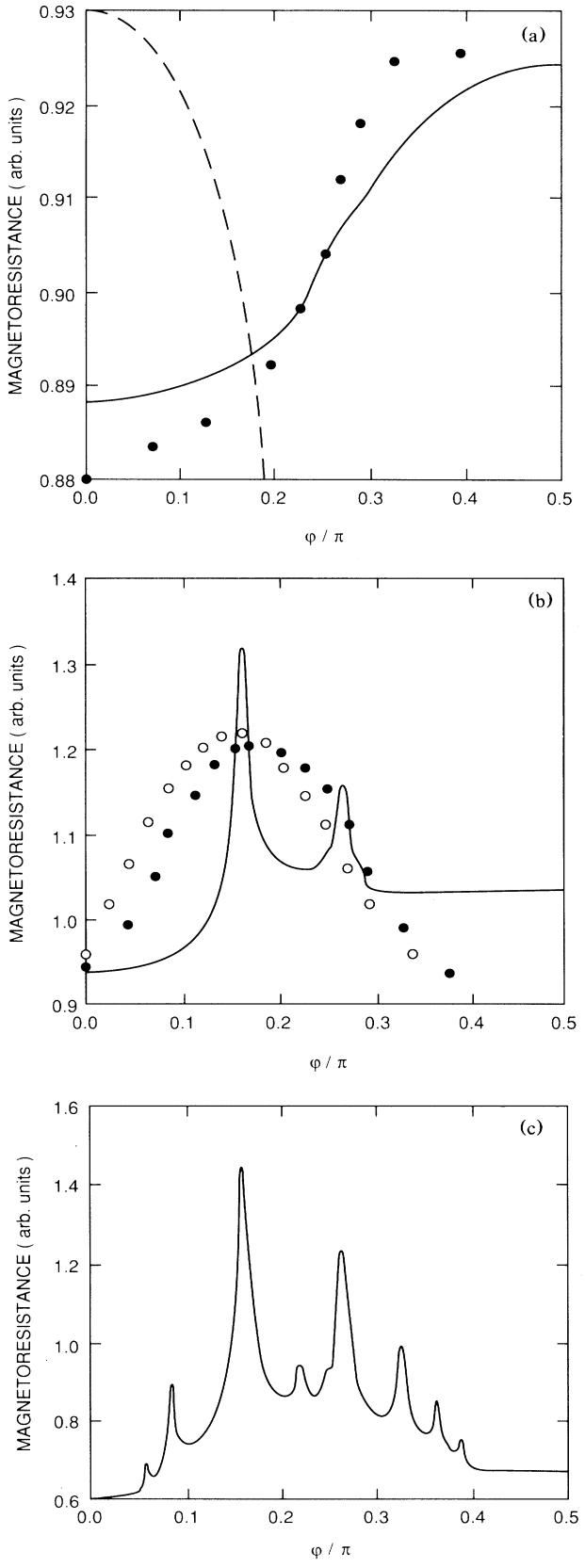

FIG. 1. Angular dependence of magnetoresistance calculated from Eqs. (2) (solid line). (a) $H=2$ T, $T=0.4$ K; the broken line shows the two-dimensional cosine law. The experimental points were measured with $H=0.5$ T and $T=0.5$ K (Ref. 13). (b) $H=4$ T, $T=0.4$ K. Experiments are for $H=5$ T, $T=0.87$ K (filled circles, Ref. 13), and for $H=4$ T, $T=0.34$ K (open circles, Ref. 12). (c) $H=4$ T, $T=0.2$ K.

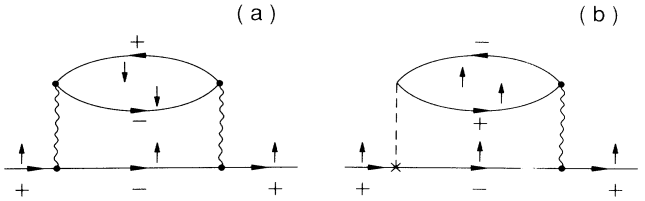

FIG. 2. Feynman diagrams for contributions to the electron self-energy from (a) electron-electron interaction (wavy lines) and (b) defect interaction (broken line).

where $\lambda_1=4t_bc/evHb\sin\gamma$, $\lambda_2=4t_cc/evHc^*$, $x_H=2c/eHb\sin\gamma$; φ is the angle between the magnetic field and the c^* axis, and g is the strength of the electron-electron interaction. We have ignored the possibility of a nonzero phase shift θ in Eq. (1). This would lead to a different oscillating function than the first Bessel function for the electron-electron diagram, Eq. (2a), but the qualitative features remain the same. The defect diagram is not affected. Note that the electron-electron interaction vanishes when $T\rightarrow 0$. On the other hand, the defect scattering increases with decreasing T, so these two processes control the conductivity in wide temperature regimes. The Pauli contribution to the two diagrams cancel; the appropriate spin configurations are shown in Fig. 2 by arrows.

The origin of the peculiar three-dimensional character of the magnetoresistance is quite evident from Eqs. (2a) and (2b): The scattering probability contains a *product* of Bessel functions with arguments related to the electron motion in the b^* and c^* directions, respectively. Thus, in spite of the smallness of the parameter t_c, the motion in the c^* direction is as important as the motion in the b^* direction in determining the resistivity. This unusual effect is due to the independent motion of the electrons in the b^* and c^* directions.

Figures 1(a)–1(c) show the angular dependence of the magnetoresistance for various values of temperature, with φ being the angle between the c^* direction and the magnetic field. The calculations are based on Eq. (2a). There are two main features. The first (and most important) is a general deviation from the two-dimensional cosine law which is shown in the same figure as a dashed line. The discrepancy could hardly be more dramatic. The overall character of the anisotropy of the magnetoresistance is dominated by the quasiperiodic motion of electrons in the (b^*,c^*) plane. The second is a fine

structure of narrow peaks for high fields and low temperatures. These peaks are due to resonances when the electrons have commensurate frequencies in the b^* and c^* directions. Mathematically, the resonances occur when the oscillations of the two Bessel functions in Eqs. (2) are synchronized. The angles where one Bessel function has n oscillations while the other has m oscillations are defined by the condition

$$\tan\varphi = \frac{m}{n}\frac{b}{c^*}\sin\gamma. \quad (3)$$

Note that the maximum of the magnetoresistance is temperature and field dependent. In high fields [Figs. 1(b) and 1(c)] its position is at $\varphi_{\max}=28°$, the first "magic" angle of Eq. (3) with $m=n=1$, but at lower fields [Fig. 1(a)] the maximum decreases and shifts to $\varphi_{\max}=90°$. The maximum never occurs at 0° as expected for the two-dimensional model.[16]

We would like to discuss the experimental situation in more detail. Measurements of $\rho(\phi)$ in $(TMTSF)_2ClO_4$ by Brusetti *et al.*[12] and Murata *et al.*[13] are shown in Fig. 1 for comparison with calculations. First of all, the general features, including the deviations from the cosine law in Fig. 1(a), are fairly well reproduced. The position of the maxima in the experimental curves corresponds to the first magic angle $\phi_{\max}=28°$ of Eq. (2). The broadening of the peak could be attributed to imperfections of the sample. In our opinion, however, it is more likely that the broadening arises from corrections to the energy Eq. (1) mixing the motions along the c and b axes; this can be estimated as $\delta\epsilon \approx t_c t_b/t_a \approx 1$ K.[20] Such additional terms cannot shift the positions of maxima in high fields which are entirely of geometric origin.

In fact, there exist experimental data on the magnetoresistance anisotropy in $(TMTSF)_2PF_6$ under pressure[11] showing two peaks. It is tempting to connect them with resonances in Eq. (3) with $m\neq 1$, $n\neq 1$, although their positions are not in good agreement with Eq. (3).[19]

At lower magnetic fields, $\omega_c < 2\pi T$, the maximum shifts to $\phi_{\max}=\pi/2$ in accordance with theory, Fig. 1(a).

Analysis of Eq. (2) shows a quadratic field dependence away from the magic angles, $\rho(H)\approx H^2$, while for the magic angles it is linear $\rho(H)\approx H$. Boebinger *et al.*[14] find for large angles $\rho\approx H^\alpha$, $1.8<\alpha<2$.

We would like to thank G. Montambaux and G. S. Boebinger for discussing their unpublished experimental data with us, and also L. P. Gor'kov and P. M. Chaikin for very useful discussions. This work was supported by the Division of Materials Science, U.S. Department of Energy, under Contract No. DE-AC02-76CH00016.

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[19]Note that the resistance in $(TMTSF)_2X$ may be more complicated than the one in our model, since we have used a simplified variational principle for the kinetic equation similar to the one used by Yamaji (Ref. 16) [see J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960), p. 275]. Our purpose is to demonstrate that three-dimensional effects in magnetoresistance in organic superconductors are very large, and have some striking qualitative features.

[20]G. Montambaux (private communication).