"Hot Spots," Magic Angles, and Magnetoresistance in Quasi-1D Metals

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A simple quasiclassical model is presented to explain the large, highly anisotropic magnetoresistance and striking magnetoresistance dips at magic angles in the Bechgaard salts, $(TMTSF)_2X$. In the presence of a magnetic field the electrons are swept along the open orbit sheets of the Fermi surface into "hot spots" where the scattering rate is high. At magic angles the commensurate motion allows some fraction of the electrons to avoid the hot spots. A striplike hot region seems appropriate for $(TMTSF)_2PF_6$ and suggests a strong role for electron interactions.

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As the first organic superconductors, the Bechgaard salts $(TMTSF)_2X$ attracted a considerable amount of attention [1]. They are considered quasi-one-dimensional (quasi-1D) metals with bandwidths in the ratio $4t_a$:$4t_b$:$4t_c \sim 1$:0.1:0.003 eV and an open-orbit Fermi surface (FS) which sometimes nests sufficiently well to allow density-wave instabilities. Despite being quasi-1D, the magnetoresistance is high and a host of unusual resonances and transitions have been observed in moderate to high fields. These include field-induced spin-density-wave (FISDW) transitions, $1/H$ oscillations in transport, thermodynamic, and magnetic properties [2], and observation of the quantum Hall effect [3]. Recently, a series of experiments have discovered another striking effect—strong "magic angle" dips in resistance for specific field orientations [4–7].

The Lorentz force mandates that an electron move on a constant-energy surface on a plane perpendicular to the magnetic field. (We consider magnetic fields in the plane perpendicular to the highly conducting **a** axis.) On the warped sheets of the quasi-1D FS the motion describes a path with almost constant velocity but different components along the two axes. Therefore the frequency with which the Brillouin zone (BZ) is traversed in the two directions is usually different. Lebed [8] noted that when the frequencies are commensurate, when their ratio is rational, the effective dimensionality of the system is reduced. In a series of papers he predicted that there should be interesting effects on the FISDW transition temperature at these commensurate values, obtained by suitably orienting the field. Similar conclusions were drawn by Chen and Maki [9]. Lebed and Bak [10] suggested that the commensurability effects renormalize the electron-electron and impurity scattering and should be observed in the magnetoresistance in the normal state of these materials. Experimentally, structure at the magic angles has been seen by Boebinger *et al.*, Osada *et al.*, and Naughton *et al.* in $(TMTSF)_2ClO_4$ [4–7] and recently in $(TMTSF)_2PF_6$ [7]. However, the observed magnetoresistance shows dips at the magic angles while the theory predicts peaks.

In this Letter we suggest a simple quasiclassical model which describes the unusually high magnetoresistance, the overall anisotropy, and the magic angle effects for transport along the highly conducting direction. The resistivity along this direction is controlled by the scattering rate between the two FS sheets at $\sim \pm k_F$. If there is a "hot spot" where τ_x^{-1} is very high, the resistance is determined by how fast electrons find this spot [11,12]. Electrons can diffuse to the hot spot; with a field they are swept to it. At unmagic angles the electron trajectories are incommensurate and come arbitrarily close to any point on the FS. Thus each electron is eventually swept to the hot spot. For magic angles the commensurate motion retraces itself and does not cover the FS. Thus a fraction of the electrons never encounter the hot spot.

To observe "magic angle" magnetoresistance dips it is not necessary to have "hot spots" *per se*, it is merely necessary to have some variation of the intersheet scattering rate at different points on the FS, $\tau_x^{-1}(k_y,k_z)$. The zero-field conductivity is determined by the average scattering *time*. For unmagic angles at high field each electron samples the entire FS so the conductivity for $B \to \infty$ is determined by the average scattering *rate*. At magic angles it is between these values. Since the average scattering time is always greater than the reciprocal of the average scattering rate, the high-field conductivity is always lower than the low-field conductivity. Thus with any distribution of scattering rates there is always positive magnetoresistance and magic angle dips. Below we treat two examples which are algebraically easy.

A dispersion relation for the $(TMTSF)_2X$ salts is

$$E(\mathbf{k}) = \hbar v_F(|k_x| - k_F) - 2t_b \cos(k_y b) - 2t_c \cos(k_z c) \tag{1}$$

with $v_F \approx 2\times10^8$ cm/sec. The FS is shown schematically in Fig. 1. It consists of two warped sheets with no intersections. The conductivity in the x direction is given by

$$\sigma_{xx} = 2\frac{e^2}{h} v_F \int \frac{\tau_x(k_y,k_z)\,dS_F}{(2\pi)^2}, \tag{2}$$

where $\int dS_F$ is a surface integral on the Fermi sheet and $v_x(k_y,k_z) = v_F$. The conductivity is simply related to the average scattering time.

In the presence of a magnetic field perpendicular to **a** ($\hat{\mathbf{x}}$) and tilted from **c** ($\hat{\mathbf{z}}$) by θ, Fig. 1, the electron motion projected into the plane is uniform:

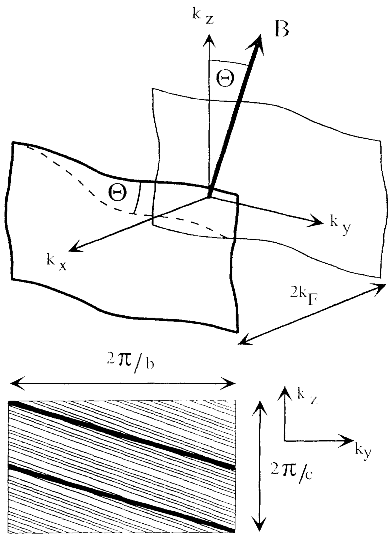

FIG. 1. Top: Schematic Fermi surface for the $(TMTSF)_2X$ salts. With the magnetic field at an angle θ from the k_z axis, an electron traverses the Fermi surface at the angle θ to the k_y axis as shown by the dashed line. Bottom: For an unmagic angle the projection of the electron orbit in the k_y-k_z plane is schematized by the thin line; the orbit covers the entire area. At a magic angle the trajectory retraces its path (thick line illustrates the case $\tan\theta = pb/qc$, $p=1$, $q=2$).

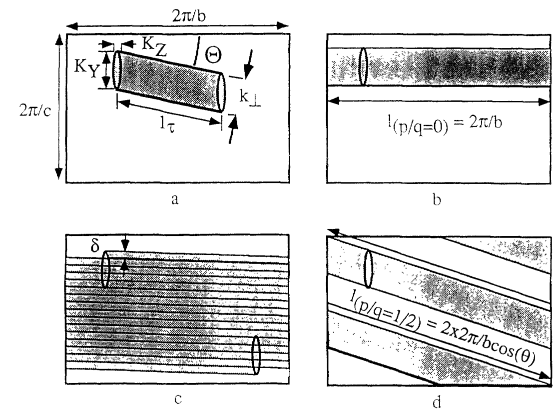

FIG. 2. (a) The fraction of the electrons swept through an ellipsoidal area of dimensions $K_Z K_Y$ is illustrated. It can be represented by the area "painted" by the ellipse in a stroke of length l_τ and width $k_\perp$ (shaded area), where l_τ is the distance the electron travels before being scattered. (b) For a magic angle ($\theta=0$ is shown) the paint strokes overlap and the painted area remains finite in infinite field. (c) For a near magic angle the paint strokes almost overlap, but each umklapp translation by a reciprocal lattice vector introduces a shift δ and the entire surface is eventually painted. However, the time to paint the entire surface is much longer than for angles far from magic. (d) The painted area for $(c/b)\tan\theta = p/q = \frac{1}{2}$ is illustrated.

$$\frac{\partial \mathbf{k}}{\partial t} = \frac{e v_F B}{\hbar c}(\cos\theta\hat{\mathbf{y}} - \sin\theta\hat{\mathbf{z}}) . \tag{3}$$

In the first case we consider a "wormhole" on the FS—a region in which an electron is immediately transferred by $\approx 2k_F$ to the other Fermi sheet, $\tau_x = 0$ for $\mathbf{k}$ in the ellipse of dimensions K_Z, K_Y as shown in Fig. 2(a). We take the scattering at all other points to be a constant τ. From Eq. (3) the path length traversed by an electron before it scatters is then $l_\tau \equiv e v_F B\tau/\hbar c$. If the field is tilted at an angle θ, then the electrons in area $A = l_\tau \times k_\perp$ are swept into the wormhole. Here $k_\perp = (K_Y^2\cos^2\theta + K_Z^2 \sin^2\theta)^{1/2}$. If the path length l_τ is longer than the dimension of the BZ then upon translation by a reciprocal lattice vector, the area may overlap itself. In particular for magic angles, where

$$\tan\theta = \frac{p}{q}\frac{2\pi/c}{2\pi/b} ,$$

the area exactly overlays itself and no further area is swept to the wormhole with increasing field. This is illustrated for $p/q = 0$ and $p/q = \frac{1}{2}$ in Fig. 2.

We want to find the area swept into the wormhole at an angle slightly away from a magic angle. An electron trajectory starts overlapping itself when it has gone a distance $l_{p/q} = (2\pi/b)q/\cos\theta$ [Figs. 2(b) and 2(d)]. For $l_\tau < l_{p/q}$ the area swept into the wormhole is $l_\tau \times k_\perp$. Equivalently we can imagine the time-reversed process where the wormhole paints an equal area of the BZ by retracing the electrons path. For $l_\tau > l_{p/q}$ the painted areas overlap. The area just before overlap is $A_1 = l_{p/q} \times k_\perp$. The additional area painted is $A_2 = (l_{p/q} - l_\tau) \times \delta$ where $\delta = (2\pi/b)q\tan\theta - (2\pi/c)p$ is the perpendicular mismatch distance for a near magic angle, Fig. 2(c). $\delta = 0$ at the magic angles. If the mismatch is greater than $k_\perp$ then the additional painted area is proportional to $k_\perp$. Below we take $w = \min(k_\perp, \delta)$. A_0 is the unpainted area. The areas painted by this process are

$$\begin{aligned} &A_0 = \frac{(2\pi)^2}{b \times c} - A_1 - A_2 , \\ &l_\tau < l_{p/q} A_1 = l_\tau \times k_\perp, \quad A_2 = 0 , \\ &l_\tau > l_{p/q} A_1 = l_{p/q} \times k_\perp, \quad A_2 = (l_\tau - l_{p/q}) \times w ; \end{aligned} \tag{4}$$

entire area covered:

$$A_1 = l_{p/q} \times k_\perp, \quad A_2 = \frac{(2\pi)^2}{b \times c} - A_1 .$$

In Fig. 3 we show the total area painted (the fraction of the FS swept into the wormhole) for magic angles spanning $p,q = -4$ to 4. In this and the following figure we have generalized the above results slightly by putting in the nonorthorhombic unit cell and the actual lattice parameters. The triclinic nature of the cell is what is responsible for the asymmetry.

The conductivity $\sigma_{xx} \propto \sum_i A_i \tau_i$. The average scattering time for each region is the average time to be swept to the wormhole. For $l_\tau < l_{p/q}$ we have $\tau_0 = \tau$, $\tau_1 = \tau/2$, for $l_\tau > l_{p/q}$, $\tau_0 = \tau$, $\tau_1 = l_{p/q}/(2ev_F B/\hbar c)$, $\tau_2 = \tau_1 + \tau/2$, and if the entire area is covered we have $\tau_1 = l_{p/q}(2ev_F B/\hbar c)$, $\tau_2 = \tau_1 + (A_2/\delta)/(2ev_F B/\hbar c)$. For small Hall conductivity as in the metallic state of the Bechgaard salts the resistance is the inverse conductivity. The resistance for the wormhole model is shown in Fig. 4(a). The cosinelike

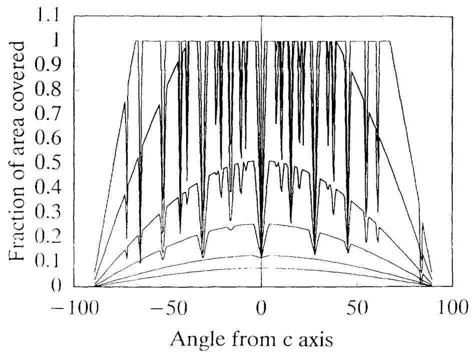

FIG. 3. The fraction of the Fermi surface swept into an ellipse with dimensions $K_Y/(2\pi/c)=0.12$, $K_Z/(2\pi/b)=0.01$, for increasing field with $\omega_c(0)\tau_{yz}\equiv ev_F bB\tau_{yz}/hc=20$ (top) to 10, 4, 2, 1, 0.6 (bottom).

angular dependence is from the ellipsoidal wormhole. A circular wormhole would give a flat background. The magic angle dips develop as the field is increased. They first appear when the orbit first retraces itself, i.e., when $ev_F B\tau/\hbar c$ first equals $l_{p/q}$. The magnetoresistance saturates at the magic angles. The magnetoresistance is nonsaturating and approaches a linear dependence on field at the unmagic angles.

Another algebraically simple model is a hot strip ex-

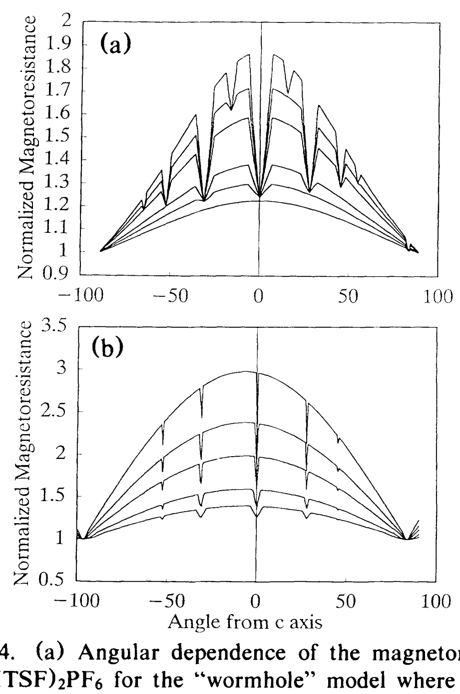

FIG. 4. (a) Angular dependence of the magnetoresistance for $(TMTSF)_2PF_6$ for the "wormhole" model where electrons entering the ellipse defined by $K_Y/(2\pi/c)=0.2$, $K_Y/(2\pi/c)=0.001$ are instantly scattered. The fields are 10, 9, 8, 6, 5, and 4 T from top to bottom and $\tau_x=\tau_{yz}=6\times10^{-12}$. (b) Magnetoresistance for a model where scattering is from a hot strip across the Fermi surface parallel to k_z. The strip width is $K_Z/(2\pi/b)=0.3$ and the hot part $[K_Y/(2\pi/c)=0.75]$ scatters 10 times faster than the warm part. The fields are 10, 7, 5, 3, and 2 T and $\tau_{yz}=5\times10^{-11}$.

tending across the BZ parallel to the c^* axis. A large region on the strip, of length K_Z, is even hotter. When an electron traverses the strip it has a probability ϵ_1 of being scattered in the hotter region and ϵ_2 in the warm part. The scattering rate for electrons is proportional to ϵ and the rate at which they encounter the belt. For $\theta=0$, orbits are parallel to the $\hat{y}$ axis. The frequency with which electrons cross the BZ is $\omega_c=ev_F bB/hc$. For $\theta=0$, the area of the FS crossing the hotter part of the strip is $a_1=(2\pi/b)K_Z$ and the scattering rate is $\tau_1^{-1}=\epsilon_1\omega_c$. Similarly the cooler part has $a_2=(2\pi/b)(2\pi/c-K_Z)$ and $\tau_2^{-1}=\epsilon_2\omega_c$. The conductivity in each region is the product of area and scattering time.

For arbitrary angle the trajectory path length, from $-\pi/b$ to π/b is $l_0=(2\pi/b)/\cos\theta$. The rate at which the hot strip is traversed is $\omega_c(\theta)=\omega_c(0)\cos\theta$. If all other scattering processes are represented by the zero-field value τ^{-1} then we approximate the scattering rate as $\tau_{1,2}^{-1}(\theta)=1/\tau+\epsilon_{1,2}\omega_c(\theta)$. The areas swept by hot and warm parts are as in Eq. (5) with $a_1=A_1+A_2$, $a_2=A_0$. The magnetoresistance in this model is shown in Fig. 4(b). For this model to show significant effects an electron must traverse the BZ many times before scattering. The number of magic angle dips is not governed by the lifetime as in the wormhole model; rather, for large K_Z, the painted area covers the entire FS even for small q. The magnetoresistance is nonsaturating at both magic and unmagic angles and is $\propto\omega_c\propto B$. (A self-consistent solution would limit τ_{yz} by the longitudinal rate and this leads to saturation at higher fields [12].)

The strip model is qualitatively consistent with data on $(TMTSF)_2PF_6$ [7]. It shows a large nonsaturating cosinelike angular magnetoresistance with sharp downward cusps at a few magic angles. The cusps get sharper but their number does not increase with higher field as in the experiment. The actual distribution of scattering rates is undoubtedly not this simple, but the highly anisotropic striplike region, into which most of the FS is swept even at the most commensurate angle is probably not a bad approximation.

What mechanism might produce the dramatic variation in scattering rates? A minimum $2k_F$ is necessary to relax the x-axis conductivity. Impurity scattering might do; phonons are completely ineffective. The most natural origin would be the electron-electron umklapp scattering which is peculiar to the quasi-1D system at half filling. Scattering two electrons backward by $2k_F$ requires balancing by $4k_F$ from the lattice. Since $4k_F$ is a reciprocal lattice vector, it works. On the other hand the constraints put forward by electron-electron umklapp processes are not tremendously restrictive. A more interesting and speculative idea is that the incipient spin-density wave observed at lower pressures or higher magnetic field produces a region near the best nesting vector where the scattering rate is high. Both of these mechanisms have previously been suggested to explain the large magnetoresistance itself [11].

The striplike scattering region suggests an even more speculative idea. If a zone boundary intersects the FS it would do so in a pair of wavy lines along $\mathbf{c}^*$. A reciprocal lattice vector would then directly give the interplane scattering. The strong-coupling version would be a gap along these lines and the formation of two closed-orbit cylinders containing an equal number of electrons and holes. (For one dimension, the large-Coulomb-repulsion half-filled Hubbard model has an insulating gap. Whether some aspects of this gap remain for the quarter-filled case with small dimerization and small transverse bandwidths remains to be treated theoretically.) Most of the calculations above go through but the interpretation would be that at certain regions along k_z there is a higher probability of breakdown between the cylinders. The closed orbit cylinders would also explain the mysterious "fast oscillations" observed in the Bechgaard salts [2] and the magnetization anomalies at the magic angles [6].

Cu shows magic angle effects from electrons swept into the neck orbits in a mechanism similar to the present proposal [13]. In quasi-two-dimensional systems, magic angles are understood in terms of the vanishing of the dispersion along the field direction when an orbit takes in both the belly and the neck [14]. A similar model has been proposed by Osada, Kagoshima, and Miura for the Bechgaard salts [15] with the commensurability leading to the vanishing of the dispersion along the field. Maki [16] has recently calculated strong resonant dips in conductivity. In these latter cases a characteristic behavior is that the magic angle dips are broad and parabolic, whereas the anti-magic-angle peaks are cusplike, the opposite shape of the present model. Moreover, the latter models show large effects along the field direction and only subsidiary effects for ρ_{xx}. The $(TMTSF)_2PF_6$ [7] data show strong downward cusps in ρ_{xx} much more like the predictions of the present model.

We have demonstrated that a quasiclassical model with any variation in the scattering rate on the Fermi surface can produce magic angle dips in the magnetoresistance of quasi-one-dimensional metals. A model which reproduces the essential features of the effects observed in $(TMTSF)_2PF_6$ is one in which the scattering is dominated by a strip parallel to the c^* axis. A possible cause for this strip is strong electron-electron scattering, the presence of a pseudo-gap related to the insipient field-induced spin-density wave, or the combined effect of a strong Coulomb correlation with the dimerization of the TMTSF stack giving rise to true or pseudo closed orbits.

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[1] D. Jerome and H. J. Schulz, Adv. Phys. **31**, 299 (1982); R. L. Greene and P. M. Chaikin, Physica (Amsterdam) **126B**, 431 (1984); in *Low Dimensional Conductors and Superconductors,* edited by D. Jerome and L. G. Caron, NATO Advanced Study Institutes, Ser. B, Vol. 155 (Plenum, New York, 1987); *Proceedings of the International Conference on Synthetic Metals, 1991* [Synth. Met. **43** (1991)]; T. Ishiguro and K. Yamaji, *Organic Superconductors,* Springer Series in Solid State Sciences Vol. 88 (Springer-Verlag, Berlin, 1990).

[2] P. M. Chaikin *et al.*, Phys. Rev. Lett. **51**, 2333 (1983); H. Schwenk *et al.*, Phys. Rev. Lett. **56**, 667 (1986); T. Osada *et al.*, Solid State Commun. **60**, 441 (1986); Physica (Amsterdam) **143B**, 403 (1986); J. P. Ulmet *et al.*, Physica (Amsterdam) **143B**, 400 (1986); X. Yan *et al.*, Phys. Rev. B **36**, 1799 (1987).

[3] S. T. Hannahs *et al.*, Phys. Rev. Lett. **63**, 1988 (1989); J. R. Cooper *et al.*, Phys. Rev. Lett. **63**, 1984 (1989).

[4] G. S. Boebinger *et al.*, Phys. Rev. Lett. **64**, 591 (1990).

[5] T. Osada *et al.*, Phys. Rev. Lett. **66**, 1525 (1991).

[6] M. J. Naughton *et al.*, Phys. Rev. Lett. **67**, 3712 (1991).

[7] W. Kang, S. T. Hannahs, and P. M. Chaikin, preceding Letter, Phys. Rev. Lett. **69**, 2827 (1992).

[8] A. G. Lebed, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 137 (1986) [JETP Lett. **43**, 174 (1986)].

[9] L. Chen and K. Maki, Synth. Met. **29**, F493 (1989).

[10] A. G. Lebed and P. Bak, Phys. Rev. Lett. **63**, 1315 (1989).

[11] C. S. Jacobsen *et al.*, Solid State Commun. **38**, 423 (1981).

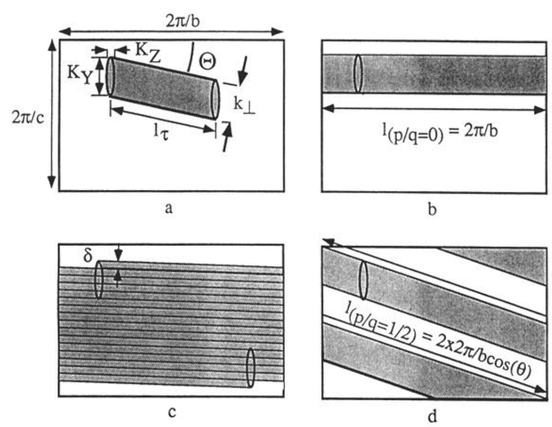
[12] Mu-Yong Choi, P. M. Chaikin, and R. L. Greene, J. Phys. (Paris), Colloq. **44**, C3-1069 (1983).

[13] A. B. Pippard, *Magnetoresistance in Metals* (Cambridge Univ. Press, Cambridge, 1989).

[14] K. Yamaji, J. Phys. Soc. Jpn. **58**, 1520 (1989).

[15] T. Osada, S. Kagoshima, and N. Miura, Phys. Rev. B **46**, 1812 (1992).

[16] K. Maki, Phys. Rev. B **45**, 5111 (1992).



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