Simple fit for magic-angle magnetoresistance in $(TMTSF)_2PF_6$

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The angular-dependent magnetoresistance in the Bechgaard salts $(TMTSF)_2X$ ($X=PF_6, ClO_4$, etc.) is characterized by sharp dips when the field is aligned along lattice vectors. Although magic-angle effects were originally predicted by Lebed, they remain largely unexplained. Here we present detailed interplane transport data and show that a simple model involving conductivity contributions from transfers between neighboring chains, which are individually destroyed by a perpendicular field, fits most *c*-axis data remarkably well. The fitting illustrates what can be explained by classical transport and what cannot—a dominant term from hopping perpendicular to the current direction. Combined with the observation that the fitting also ''works'' for *a*-axis data, this suggests a different interpretation: The quasi-one-dimensional system is insulating and any unnesting transfer makes it metallic.

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The Bechgaard salts were the first organic superconductors and are among the most remarkable electronic materials yet discovered.[1] A single crystal of $(TMTSF)_2PF_6$ is metallic, semiconducting, insulating, superconducting (triplet?[2]), antiferromagnetic, spin and charge density wave, and exhibits a cascade of first-order field-induced spin density wave (FISDW) transitions, with a bulk three-dimensional quantum-Hall effect as temperature, pressure, and magnetic field are varied. Much of its unusual properties and ground states result from the chainlike structure and quasi-one-dimensional Fermi surface consisting of two warped unconnected planes about $+/-k_{Fa}$.[3] Such a Fermi surface was expected to exhibit an uninteresting magnetoresistance—small for transport along the chains and large and unsaturating for open-orbit electron trajectories along the *b* and *c* axes. Experimentally, there are striking angular-dependent effects with large dips in the magnetoresistance at ''magic angles.''[4] Equally striking, the temperature dependence is ''metallic'' ($dR/dT>0$) for fields aligned at the magic angles and ''nonmetallic'' ($dR/dT<0$) at other orientations.[5,6] The magnetoresistance at high fields depends on the orientation of field relative to the crystal axis rather than the current direction.

Before the experiments, Lebed[7] predicted magnetoresistance peaks when periodic motion along k_b and k_c was commensurate. These trajectories occur when the magnetic field is applied along the crystal lattice vectors in the *b*-*c* plane (i.e., field pointing between pairs of conducting chains). Experimentally, Lebed magic-angle effects have been seen as dips rather than peaks in magnetoresistance.[4] In an attempt to explain these phenomena many theories have been proposed,[8–10] none of which adequately describes the data. Some address the scattering rates in the presence of fields and dimensional crossover,[9] others concern a detailed description of the Fermi surface,[10] and one suggests a transition from Fermi liquid to non-Fermi liquid as field off the magic angles decouples the layers.[11]

In this paper, we present a detailed study of the angular-dependent *c*-axis magnetoresistance as a function of temperature and field and a simple analytic fitting scheme that does a quite reasonable job of reproducing the experimental observations. The fit takes the conductivity as the sum of contributions from hopping between chains. These contributions are killed by the application of a field perpendicular to the hopping directions. For the *c*-axis conductivity this is similar to a classical result[8] but requires the addition of an anomalous contribution from hopping perpendicular to the current. As a description of the transport in all directions, it suggests that the quasi-one-dimensional system is insulating in the absence of interchain transfer (even without a density-wave distortion) and any tunneling that kills the nesting leads to a metallic state.

In Fig. 1 we show the data and fit for the *c*-axis resistance of $(TMTSF)_2PF_6$ at a pressure of 9.2 kbar as a function of angle for several different fields.[6] The *c* axis is the least conducting and the magnetoresistance in this direction is the easiest to analyze in terms of classical theory. Assuming an

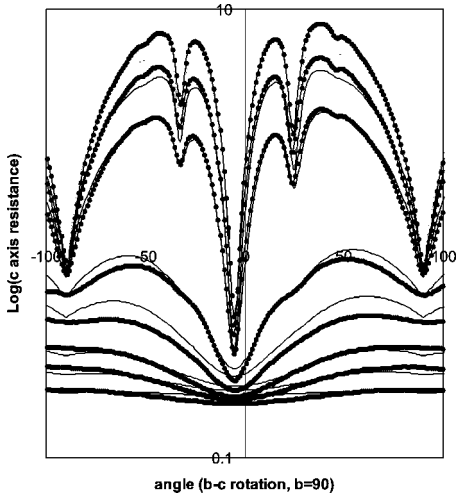

FIG. 1. Angular dependence of the magnetoresistance of $(TMTSF)_2PF_6$ at a pressure of 9.2 kbar and a temperature of 1.2 K. The dotted curves are the experimental data taken in fields of 0.1,0.2,0.3,0.6,1.0,4.0,6.0,7.8 T in order from bottom to top. The light solid lines are a fit to the data using Eq. (2). The data were fit at 4 T and other curves are the result of changing the field.

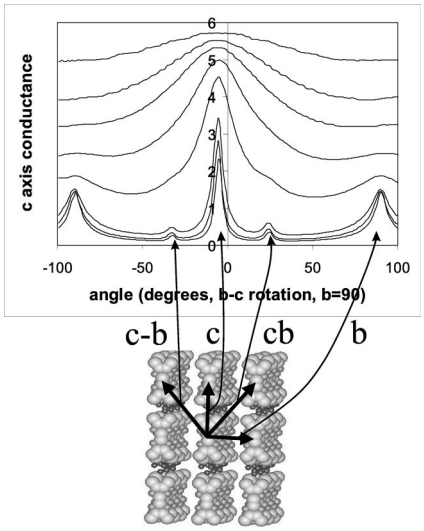

FIG. 2. Data of Fig. 1 are replotted as conductivity. At high field conductivity remains only when the field is aligned along the directions connecting the quasi-one-dimensional chains. The cartoon shows the TMTSF chains and the directions of field alignment for the conductivity peaks.

approximate orthorhombic band structure of the form $\epsilon(k) = 2t_a\cos(k_x a)+2t_b\cos(k_y b)+2t_c\cos(k_z c)$, we can use a variety of techniques to arrive at the answer than the resistivity should vary as $\rho_c=\rho_{c0}[1+(\omega_{\perp c}\tau)^2]$ or $\delta\rho_c/\rho_{c0} \propto(\omega_c\tau[\cos(\theta)])^2$ where θ is the angle with the c axis and $\omega_c=eHv_Fc/h$ is the frequency with which an electron traverses the Brillouin zone in a magnetic field. Intuitively the Lorentz force, $\boldsymbol{F}=\boldsymbol{v}\times\boldsymbol{B}=\boldsymbol{j}\times\boldsymbol{B}/ne$ is zero for field parallel to current and maximum for field perpendicular to the current. For low fields (toward the bottom of Fig. 1) that is precisely the behavior seen. However, as the field is increased the dips begin to appear at the magic angles and the ''background'' takes on a very non-$\cos^2$ shape with a pronounced minima when the field is perpendicular to the current. Note that the positions of the dips do not shift with field. The fact that the θ dependence is not symmetric results from the triclinic (as opposed to orthorhombic) crystal structure.

In order to understand (or at least fit) the data of Fig. 1 we can take a sum of resistivities (as for a scattering rate model) or a sum of conductivities (as when there are contributions from different regions in k space or different channels). In Fig. 2 we plot the conductivity instead of the resistivity. The plot shows that conductivity is the appropriate quantity. With each conductivity peak we can associate a hopping direction between the chains as indicated by the cartoon and arrows. For high fields the conductivity only persists for alignment with the magic angles. We see a sharp drop in the individual conductivity components as the field is rotated away from the hopping direction. The zero-field conductivity is the sum of the contribution from all hopping directions. The form that we chose for the fit is

$$\sigma_c=\tau(T)\left(\left\{\sum_i \frac{t_i^2}{\left[1+\left(\frac{H_{\perp i}}{H_{i0}}\right)^{\alpha}\right]}\right\}+\frac{t_{b'}^2}{\left[1+\left(\frac{H_{\perp b'}}{H_{b'0}}\right)^{\alpha'}\right]}\right). \tag{1}$$

The basic idea is that each contribution is killed by a perpendicular field and the simplest thing to choose was a Lorentzian-type cutoff. Qualitatively even an exponential cutoff mimics the data, (so the exponents α and α' should not be taken too seriously) but the plots shown in Fig. 1 were made explicitly with

$$\sigma_c=\tau(T)\left(\left\{\sum_i \frac{t_i^2}{1+a_i[H_{\perp i}\tau_i(T)]^2}\right\}+\frac{t_{b'}^2}{1+a_{b'}[H_{\perp b'}\tau_{b'}(T)]^1}\right). \tag{2}$$

The parameters (nine independent ones) were fit to the data at 4 T. The other plots were made by changing the field. (The ratios of the parameters for the transfer integrals are $t_c:t_{cb}:t_{c-b}:t_{c2b}:t_{c-2b}:t_b=1:0.25:0.33:0.05:0.05:0.7$, the ratios of the a_i's are $a_c:a_{cb}:a_{c-b}:a_{c2b}:a_{c-2b}:a_b=3.5:4:4:2:2:2.$) All but the final term in Eq. (2) are to be found in Osada's model that comes from classical Boltzmann transport. He assumes that there are terms in the band structure $\Sigma_{n,m}2t_{nb,mc}\cos(k_bnb+k_cmc)$ from hopping in different interchain directions. The warping they produce on the Fermi surface contributes to the conductivity and is destroyed by a field as $(\omega_\perp\tau)^2$ similar to the elementary Lorentz-force result. However, there is no conductivity contribution to the c-axis conductivity from the warping along b. The interplane terms in Eq. (2) are approximately what would be expected from calculations, but the intraplane term is very low compared to that expected for b-axis hopping (i.e., $t_c:t_b\approx1:30$ rather than $t_c:t_b\approx1:0.7$). This intraplane term is completely anomalous. It is difficult to imagine a way in which intraplane transfer can strongly effect conductivity in the interplane direction. The destruction by perpendicular field of this term is also different from the interplane terms (H^1 rather than H^2).

An in-plane energy that is comparable to the anomalous term in the fit is the unnesting transfer $t_{b'}$. A half-filled tight-binding band has perfect nesting and would undergo a density-wave instability. The perfect nesting is destroyed by moving off half filling (the present case is quarter-filled dimerized) but it is convenient to retain the simpler band structure and filling and treat the unnested condition by adding a term $2t_{b'}\cos(2k_yb)$ to the dispersion relation. The FISDW transitions are the result of a perpendicular field eliminating this unnesting.[12]

To proceed further we wanted to see if this fit would also work for the temperature dependences previously found.[6] We chose the simplest approximation, that there was a single-

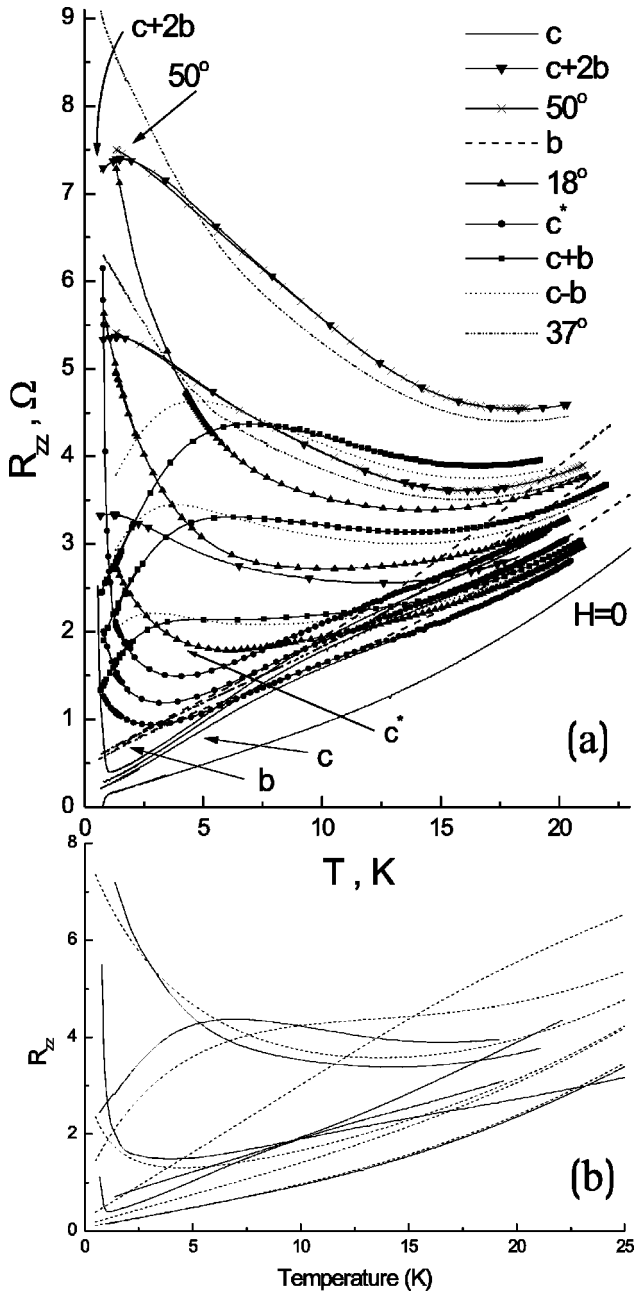

FIG. 3. (a) Temperature dependence of the resistance at 4, 6, and 7.8 T for different field orientations. (b) Solid lines are 7.8 T data, from (a) dashed lines are the result of Eq. (2).

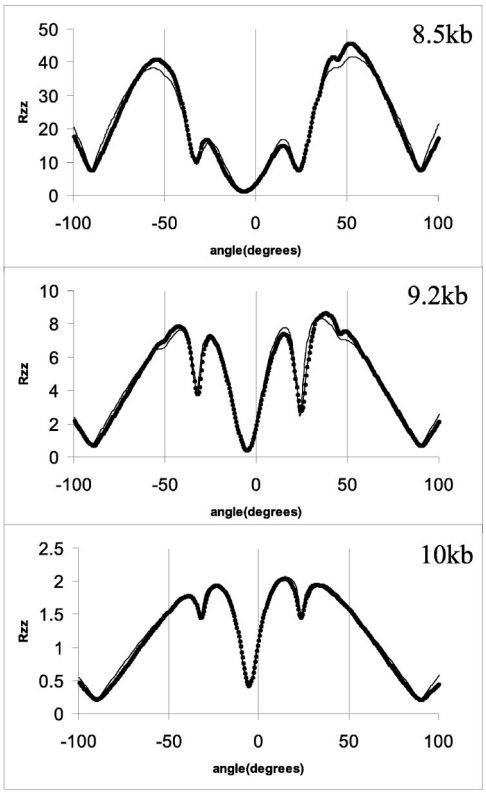

FIG. 4. Angular dependence of the magnetoresistance at 1.2 K and 7.8 T for three different pressures. The data are fit with Eq. (2). Although there are slight modifications to other variables, the main difference in the fits is the change in the $t_{b'}$ term from 0.4 to 0.7 to 1.6 for 8.5, 9.2, and 10 Kb, respectively.

scattering time. We then fit the model at zero field to the measured zero-field resistance and extracted the temperature dependence of τ. This value was then used with the fixed parameters from the 4 T, 1.2 K fit above to produce the dashed lines in Fig. 3. The solid lines are the experimental data. Although the fit is not quantitative (being off by more than a factor of 2 in places), all of the essential features are there. In particular, all magic-angle traces look metallic and decrease at low temperature, while off the magic angles the resistance increases at low temperature. The reason is clear, the conductivity at the magic angles is increasing like τ while in high field off the magic angle it is decreasing like $1/\tau$.

In Fig. 4 we show the angular dependence of the magnetoresistance of $(TMTSF)_2PF_6$ for three different pressures along with model fits. Although the curves look very different the fits are again extraordinary. The main thing that has been changed with pressure is the anomalous fit parameter $t_{b'}$ that increases with pressure. A similar fit could be done for $(TMTSF)_2ClO_4$ with a yet smaller value of $t_{b'}$.

The "c"-axis transport described above is, therefore, consistent with a contribution from classical-band effects but with an additional anomalous contribution from a term that is destroyed by a field perpendicular to the b-axis. When compared with the other terms this anomalous contribution is of the order of unnested bandwidth $t_{b'}$ and varies with pressure as might be expected for such a term. The effect of a magnetic field perpendicular to the plane is well known in the studies of the FISDW. As $H_{\perp a,b}$ is increased, the susceptibility toward a density wave increases and at a threshold there is a transition to a spin density wave.[12] Higher fields lead to a cascade of transitions associated with quantum-Hall states. In fact, in our data, we observe the FISDW for $H \sim ||c$ at 1 K at 7.8 T (see Fig. 3). However, we observe the same transition temperature for $H||c$ (a magic angle) and for ($H||c$*

$=H_{\perp a,b}$). In the former case the resistance is decreasing before the FISDW, while in the latter it is increasing. In both cases the FISDW is sharp, indicating that the resistance increase for the nonmagic angle is not simply a fluctuation effect from the approaching FISDW.

The Lebed magic-angle dips are also observed for transport along the *a* (Ref. 4) and *b* (Ref. 6) directions. Along the *a* direction there is no classical reason for the large dips nor does the field rotation in the perpendicular b*-c* plane kill any hopping term parallel to the current. However, the model written in Eqs. (1) and (2) works surprisingly well as is for *a*-axis transport and the fits become almost perfect if the anomalous term is instead taken as $t_{b'}^2/[1+(H_{\perp b'}\tau)^{1/2}]$. Since, the conductivity fits a sum of terms corresponding to hopping from one chain even for transport along the chain, an explanation cannot be in terms of classical transport.

Instead we consider the following scenario. In the absence of any interchain hopping the system is insulating, with a gap and no states at the Fermi energy. When interchain hopping is turned on, states appear at ϵ_F (in proportion to the square of the interchain hopping?). In fact the quarter-filled dimerized chains should be correlated insulators and several models suggest that the effect of interchain transfer is precisely to introduce states at ϵ_F rather than to close the gap.[13] Our data tend to support this picture. However, the relevant transfer in the *b* direction appears to be $t_{b'}$ rather than t_b. Thus, either the one-dimensional correlated insulating state persists in two dimensions as long as there is perfect nesting or t_b is renormalized by a factor of 60.[13]

In conclusion, we have demonstrated that the *c*-axis magnetotransport is well modeled by a sum of conductivity contributions from transfers along directions perpendicular to the quasi-one-dimensional chains. Each of these contributions is progressively killed by application of a field perpendicular to the tunneling direction. Similar modeling holds for *a*-axis transport. Most of the conductivity components for the *c*-axis (interplane) transport are expected from classical models, however, there is an anomalous in plane contribution that changes with pressure and dominates the shape of the angular-dependent magnetoresis tance. If this contribution were understood, the problem could be viewed as largely solved. On the other hand, since the modeling ''works'' for all directions it suggests a more interesting scenario: The interacting nested two-dimensional quarter-filled band system is insulating and gapped (even in the absence of a density-wave distortion). The effect of any hopping that competes with the two-dimensional nesting adds conducting states inside the gap. Each of these contributions is killed by a perpendicular magnetic field.

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