Non-Fermi-Liquid Behavior in Transport in $(TMTSF)_2PF_6$

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It has recently been suggested that the highly anisotropic organic conductor $(TMTSF)_2PF_6$ is marginally a three-dimensional Fermi liquid which can be destabilized by a small field in the intermediate conducting **b** direction, H_b. This field would cause the interplane transport to become incoherent. We examine the 3D Fermi surface and find it coherent for $H_b = 0$ and incoherent or nonexistent for small H_b. At similar H_b the magnetoresistances become power laws in the field applied perpendicular to the conducting planes ($\rho_a \propto H_\perp^{1/2}$ and $\rho_c \propto H_\perp^{3/2}$). These are 2D, non-Fermi-liquid properties.

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The Bechgaard salts, $(TMTSF)_2X$ (where TMTSF is tetramethyltetraselenefulvaline [1]), are highly anisotropic organic conductors with a rich phase diagram exhibiting spin density wave (SDW), metallic, superconducting, and field induced SDW or quantum Hall phases at low temperatures. The transitions between these phases depend on pressure, temperature, magnetic field, and the anion of the salt (labeled X above). Here we concentrate on the "normal" metallic phase which exists in the PF_6 salt at pressures exceeding ~6 kbar and above the critical field for superconductivity ($H \sim 0.2$ T along the **c** axis) and below the field induced SDW (FISDW) threshold field ($H \sim 4$ T along **c**). The crystal has highly conducting chains along the **a** direction which couple in the **b** direction to form conducting planes. The weakest coupling is perpendicular to the planes along the **c** direction. The bandwidths along the crystal axes are $4t_a{:}4t_b{:}4t_c \approx 1{:}0.1{:}0.003$ eV. The Fermi surfaces consist of slightly warped parallel sheets at $k_x \sim \pm k_F$. For rotation of the magnetic field in the $\mathbf{b}^*\mathbf{c}^*$ plane (perpendicular to the highly conducting **a** axis) Lebed predicted resonances in physical properties at particular angles [2]. At these angles the electron motion across the Fermi sheets has a rational ratio of the frequencies for crossing the Brillouin zone in the $\mathbf{b}^*$ and $\mathbf{c}^*$ directions. These "magic angle" resonances have been observed in the magnetoresistance of the ClO_4 and PF_6 salts [3–7]. While a semiclassical explanation may be appropriate for the behavior seen in ClO_4, an alternative explanation has been proposed for PF_6. The magnetoresistance in PF_6 is unusual in a classical context and the dips at these magic angles are sharper and more striking than in ClO_4. Strong, Clarke, and Anderson [8,9] suggest that there is coherent hopping between the conducting planes only at the magic angles. Slight angular deviations lead to an "incoherence" transition with the 3D Fermi liquid becoming a decoupled set of 2D non-Fermi-liquids. In this Letter we perform an experimental test of this model.

According to the Strong, Clarke, and Anderson model, two-dimensional sheets of $(TMTSF)_2PF_6$ would act as non-Fermi-liquids resulting from electron interactions. However, the interplane coupling, t_c, is marginally large enough to couple the sheets and lead to a 3D Fermi liquid. Introducing a small field in the **b** direction, H_b, dephases the interplane tunneling leading to an effective t_c which is below the threshold for Fermi-liquid behavior. The result should be the reestablishment of decoupled, 2D, non-Fermi-liquid behavior. In this 2D system the transport properties should depend solely on the component of the magnetic field perpendicular to the planes, $H_\perp$. Experiments on the PF_6 salt show that ρ_{xx} and ρ_{zz} vary as $H_\perp^{1/2}$ and $H_\perp^{3/2}$, respectively. These peculiar power laws are indicative of nonclassical, non-Fermi-liquid behavior.

We recently developed a technique to measure the Fermi surface of quasi-one-dimensional metals and have applied this technique to measure $(TMTSF)_2ClO_4$ [10]. We probe the **c** axis magnetoresistance during an **ac** rotation (perpendicular to the magic angle rotation direction described above). There is a sharp structure observed in the angular dependence of ρ_{zz}, particularly near $H \parallel \mathbf{a}$. The effects are quantitatively well described by a simple averaging of the velocity along **c** by the quasiclassical orbits resulting from the magnetic field. These experiments provide the best measurements for the band parameters in this salt. Here we attempt the same experiment for the PF_6 salt with H_b zero and nonzero. According to the coherence-incoherence model, for an **ac** rotation with $H_b = 0$ there should be coherent transport along **c** and, therefore, a Fermi surface, and we should observe the ρ_{zz} resonances near $H \parallel \mathbf{a}$. There should be a small, nonuniversal, non-power-law dependence observed for the resistance as a function of the magnitude and direction of the applied field. With H_b above some small threshold, there should be no coherent motion along **c**, no observable Fermi surface along **c**, and the **ac** resonance should disappear. We should then find a universal power law for $\rho_{zz}(H_\perp)$. This is indeed what we observe.

The experiments were performed in a micropressure bomb, whose dimensions were 0.76 cm diam × 1.27 cm long [11]. This beryllium-copper pressure clamp can attain

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pressures of 15 kbar and is designed to be placed on a "flipper" probe. This probe has a gear driven rotation mechanism with the rotation axis perpendicular to the probe's long axis. It has the ability for 360°+ degrees of rotation with an accuracy of 0.05°. The sample flipper is immersed in ^{3}He in a cryostat capable of cooling to 0.5 K. The clamp is centered in a 9 T, split-bore, superconducting magnet. The combination of a rotation of the probe by a goniometer (accuracy 0.0025°) and flipper rotation allows complete 4π orientation of the sample relative to the magnetic field. The sample alignment along the **a** axis is greatly aided by the fact that the superconducting critical field is highest for $H \parallel \mathbf{a}$. The **b** axis is also easily located from the fact that the magnetoresistance is smallest (and cusplike vs angle) for $H \parallel \mathbf{b}$.

In the bottom trace of Fig. 1 we show the **c** axis resistance for PF_6 as a function of angle as the sample is rotated about **b*** near **a** at 9.8 kbar, 0.57 K, and 4T. The inset shows a similar plot for anion ordered ClO_4. The behavior of the "background" is opposite in the two salts. ClO_4 has a maximum near $H \parallel \mathbf{a}$ and no magnetoresistance (below the threshold field for the FISDW transition) for $H \parallel \mathbf{c}$. This is the classically expected behavior—no magnetoresistance when the probing current and magnetic field are parallel. With current along **c** in PF_6, the magnetoresistance has a *minimum* for H perpendicular to the current and a *maximum* when the field and current are parallel. This behavior suggests that even with $H_b = 0$, PF_6 is not a conventional metal. Despite the strong difference in the "background" behavior, the $H_b = 0$ data for both salts show structure near $H \parallel \mathbf{a}$. To eliminate the effects of the background and make the peak structure more apparent, we compare $d^2 \ln(\rho_{zz})/d\theta^2$ for both materials in Fig. 2. The θ axis for the ClO_4 data has been scaled for ease of comparison (see [12]). We now see that the structures in the **ac** resonances align remarkably well. The structures are larger (~2 times) in the 7.5 T field trace (second from bottom), but are still clearly visible at the same angular positions in the $H_{ac} = 4$ T trace for PF_6 (second from top). From this we conclude that the **c** axis motion is coherent and there is a Fermi surface that can be accurately measured in PF_6 for $H_b = 0$. We obtain a value of $4t_b = 0.13$ eV for PF_6 at a pressure of 9.8 kbar.

In Figs. 1 and 2 we have also shown the data for $H_b \neq 0$ in PF_6. Remarkably, at $H_b = 0.2$ T the structure associated with the **ac** resonances has completely vanished. Since these resonances originate from coherent quasiclassical trajectories on the Fermi surface of electrons in a field, their absence indicates that either the coherence or the Fermi surface or both are no longer present. We can calculate the **ac** resonances quantitatively, and the results show that the behavior for a small H_b is a slight shift in the positions and amplitudes of the structures but that they remain essentially unchanged. We have actually performed such rotations with $H_b \neq 0$ on ClO_4 [13], and the results agree with the simulations: The positions of the peaks shift slightly, but there is no large decrease in amplitude of the resonance for small H_b. The effects in PF_6 cannot be explained in a classical Fermi liquid context, and it would appear that some nonclassical effect such as incoherence must provide the answer.

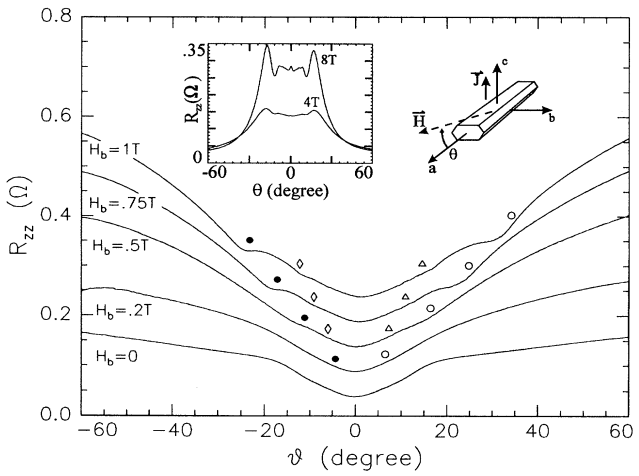

FIG. 1. **c** axis resistance (ρ_{zz}) for $(TMTSF)_2PF_6$ at 9.8 kbar, 0.57 K, and $H_{ac} = 4$ T as a function of angle between the applied field and the crystal **a** axis in the **ac** plane. Data are presented for four rotations with different values of the field along the **b** axis (offset for clarity). The inset shows the resonances observed for similar **ac** rotation for a $(TMTSF)_2ClO_4$ crystal ($H_b = 0$). The minima at the higher H_b are from "magic angle" minima of the Lebed type and are marked by their p/q value: ○ = −1, △ = −2, ◇ = 1, and ● = 2.

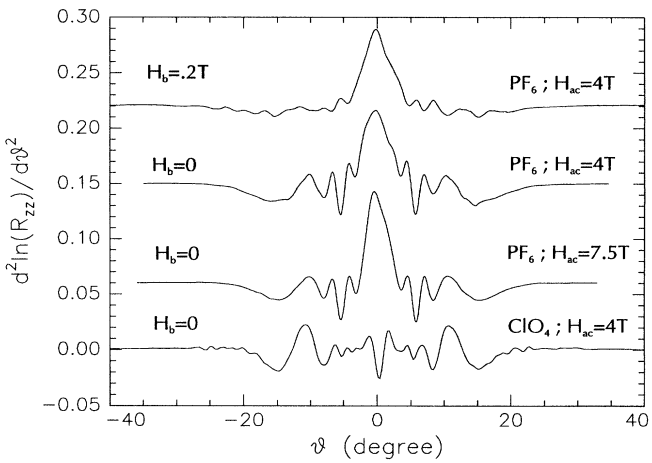

FIG. 2. Second derivative of the **c** axis resistance with respect to θ_{ac} from Fig. 1. Included for comparison is a similar derivative for $(TMTSF)_2ClO_4$ where the horizontal axis has been rescaled [12] for ease of comparison. With $H_b = 0$ the angular features in PF_6 match those for ClO_4 demonstrating for $H_b = 0$ that PF_6 has a Fermi surface extended along k_z. For $H_b = 0.2$ T, though, the features have disappeared implying that PF_6 has become a non-Fermi-liquid. The vertical scales have been adjusted to make the features of similar size: from top to bottom, ×2, ×2, ×1, ÷20.

At higher values of H_b we see from Fig. 1 that the "background" angular dependence of ρ_{zz} has changed and that there is a new set of minima that appears. These are the Lebed magic angle $\mathbf{b}^*\mathbf{c}^*$ dips discussed above. The reason that they appear near $\theta = 0$ in this rotation is that when we perform an **ac** rotation with a fixed H_b the projection of the field in the $\mathbf{b}^*\mathbf{c}^*$ plane changes and produces a highly distorted pseudo $\mathbf{b}^*\mathbf{c}^*$ rotation. For a given $H_b/H_{ac} = \tau$ we can find the angle of the field projection in the **bc** plane, θ_{bc}, in terms of the rotation angle of Figs. 1–3, θ_{ac}, according to $\tan\theta_{bc} = \tau/\sin\theta_{ac}$. We know the positions of the magic angles in the $\mathbf{b}^*\mathbf{c}^*$ plane, and we therefore expect dips at the positions indicated by symbols on the plots in Fig. 1 for a given value of H_b. The rotation is being done in the **ac** plane, but a small (1° or so) misalignment results in a large shift in the observed minima positions. This is most likely responsible for the small disagreement between the symbols and the observed dips. The magic angle effects are clear for the larger values of H_b and are not present at lower H_b. In fact, the magic angle effects and the **ac** resonances *never* coexist. This supports the suggestion that the origin of the magic angle effects in PF_6 is from non-Fermi-liquid behavior; the coherent **c** axis motion indicated by the **ac** resonances is destroyed before the magic angle effects are observed.

The second part of our test of the incoherence transition is whether the field dependence of the magnetoresistance changes over to a power law in $H_\perp$ as H_b is turned on (and interplane coupling turns off). In Fig. 3 we show $\ln(\rho_{zz})$ vs $\ln(H_\perp)$ for the different values of H_b from the data in Fig. 1. We see that for $H_b = 0$ the magnetoresistance increases with field but does not follow a power law in this field region. Recall that the classical behavior that one should expect for the magnetoresistance should involve some function of H^2 which decreases with angle as the field and current become parallel. Here we see that even the $H_b = 0$ behavior is anomalous and nonclassical. As H_b is increased we see that ρ_{zz} tends to the $H_\perp^{3/2}$ dependence found previously for $\mathbf{b}^*\mathbf{c}^*$ rotations. The fact that $H_\perp^{3/2}$ is the correct scaling is seen by the comparison of an actual $\mathbf{b}^*\mathbf{c}^*$ rotation in this figure, where again we have plotted $\ln(\rho_{zz})$ vs $\ln(H_\perp)$. This reemphasizes that above a threshold value of H_b the magnetoresistance only depends on $H_\perp$, and PF_6 behaves as a non-Fermi-liquid.

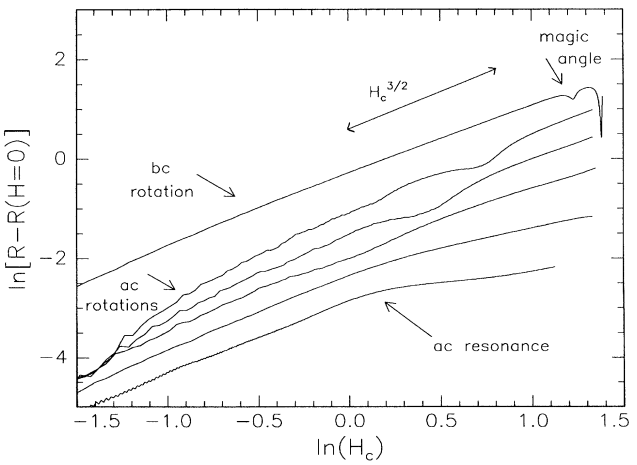

FIG. 3. Log-log plot of the c-axis resistance for $(TMTSF)_2PF_6$ as a function of $H_\perp$ for several values of H_b (from Fig. 1). As H_b increases past ≈0.5 T the resistance at high $H_\perp$ follows a power law with exponent 3/2. Included are data from a $\mathbf{b}^*\mathbf{c}^*$ rotation plotted on the same scales which also show the 3/2 power law. Traces have been offset for clarity.

Why is there such a drastic difference in behavior between the two Bechgaard salts ClO_4 and PF_6? The former seems to be a reasonable stable Fermi liquid [14]. PF_6 is only marginally a Fermi liquid and is easily transformed into a non-Fermi-liquid by a small field. The differences between these salts are most often attributed to the anion ordering transition which occurs in the ClO_4 salt at 24 K. PF_6 has a single set of Fermi surface sheets which are centered on $\pm\pi/2a$, and has a single, half-filled band (more correctly, the band is quarter filled and dimerized along **a**). We expect that electron-electron interactions will have the largest effects on a half-filled band. The anion ordering transition in ClO_4 doubles the unit cell in the **b** direction with the result that ClO_4 has two sets of Fermi surfaces (two bands) neither of which is centered at half filling. The presence of a field along **c** may also one dimensionalize the Fermi surface and lead to a strongly correlated, one-dimensional, half-filled band in PF_6. This may explain the unusually large magnetoresistance for fields along **c** ($H_b = 0$) [14].

We have conducted an experimental test of the model of Strong, Clarke, and Anderson in the quasi-one-dimensional conductor $(TMTSF)_2PF_6$. Our results indicate that below a threshold field along the **b** direction PF_6 is marginally a Fermi liquid with a Fermi surface extended in the k_z direction. Comparison of the results seen for field rotating in the **ac** plane in PF_6 to those in ClO_4 support this conclusion. For fields above ~0.2 T along **b**, however, the **ac** resonance is destroyed and PF_6 behaves as like a 2D, non-Fermi-liquid with the magnetoresistance following $H_\perp^{3/2}$.

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