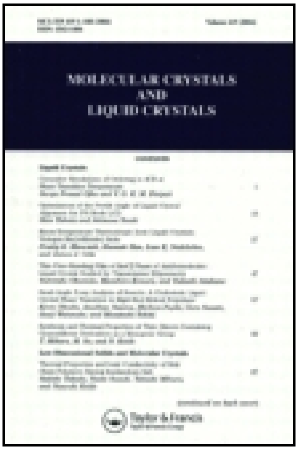

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MAGNETOTRANSPORT IN $(TMTSF)_2PF_6$ AND $(TMTSF)_2ClO_4$ UNDER PRESSURE*

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The Fermi surface of $(TMTSF)_2PF_6$ in the metallic state (under pressure) is shown to be two-dimensional at low temperature. This conclusion is valid even for fields well below the threshold for Shubnikov-de Haas oscillations. In order to explain our observations, it is necessary to assume the persistence of strong correlation effects into the metallic state and a relatively small bandwidth anisotropy $t_a/t_b \sim 10$. Such a Fermi surface cannot support a wide regime of one-dimensional superconducting fluctuations. Data are presented on the angle, temperature, and pressure dependence of the oscillation threshold field in $(TMTSF)_2PF_6$, as well as an apparently analogous effect in $(TMTSF)_2ClO_4$. We believe the threshold to be associated with a disorder-order transition of the spin system, which may also be related to the persistence of spin-density waves.

We have reported previously the observation of Shubnikov-de Haas (SdH) oscillations and an anisotropic magnetoresistance (MR) in $(TMTSF)_2PF_6$ under pressure.[1] We inferred from these that the Fermi surface is two-dimensional and compensated, consisting of electron and hole tubelets running along the crystal c-axis. The existence of a threshold field for the oscillations was also reported.

In the first part of this report we shall show from the MR anisotropy and magnitude that the two-dimensional Fermi surface picture is valid even for fields well below the oscillation threshold. These results can be explained only by a partial nesting of the Fermi surface due to a small value of the bandwidth anisotropy $t_a/t_b \sim 10$, together with significant correlation effects. This large transverse coupling is incompatible with a broad regime of one-dimensional superconducting fluctuations, such as proposed by Jerome and co-workers.[2]

In the latter part of this report we will discuss the threshold field. Data on its angle, temperature, and pressure dependence will be presented and used to infer its association with a transition of the electronic spin system, as observed in the proton spin-lattice relaxation rate.[3] Finally, we will show preliminary data on an apparently analogous effect observed in $(TMTSF)_2ClO_4$ near ambient pressure, which leads us to believe that the threshold anomaly reflects a physical process common in the superconducting $(TMTSF)_2X$ compounds.

EXPERIMENTAL DETAILS

The measurement system is the same as described previously.[1] We merely emphasize here that all data were obtained with a linear four-probe resistivity arrangement along the highly conducting a-axis, and with the magnetic field oriented arbitrarily in a plane approximately ($\pm$ 5°) normal to the current direction.

MAGNETORESISTANCE ANISOTROPY

Fig. 1 shows a "rotation diagram" of the MR of a $(TMTSF)_2PF_6$ crystal at 4.0 K under 6.9 kbar pressure for H = 90 kOe. The corresponding diagram for the same crystal at 1.1 K, where oscillations were seen, is qualitatively the same. Although the magnet is freely rotatable, the sweeps usually were limited to 190°, as here, because the $(TMTSF)_2PF_6$ diagrams always showed 180° symmetry - a reflection of the centrosymmetric property of the crystal structure.

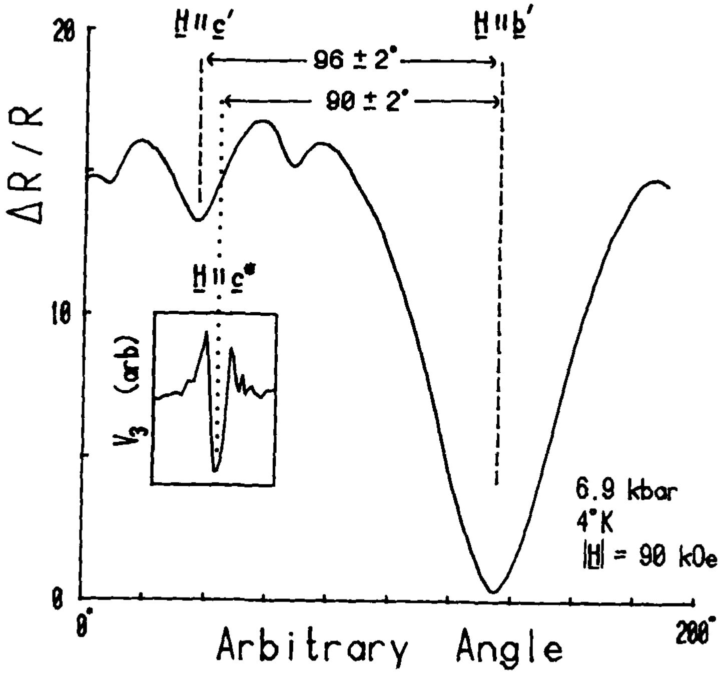

FIGURE 1 Rotation diagram of the relative a-axis transverse magnetoresistance ($\Delta R/R = R(H)/R(0)-1$) for a $(TMTSF)_2PF_6$ crystal. The origin of the angle axis is arbitrary with respect to the crystal structure. Inset is a portion of a rotation diagram of the Shubnikov-de Haas oscillations on the same sample at 1.1° K (see text).

Note first that the two main features of the diagram lie 96° (84° if you rotate to the other side of the crystal) apart, matching very nicely the angular separation of the b' and c' axes (the projections of the triclinic b and c axes, respectively, on the plane normal to the a axis). Moreover, the SdH oscillations for the same specimen at 1.1 K center not on c', but on c* (the direction normal to both a and b'). This is indicated in the inset to Fig. 1: V_3 is the sample response at 150 Hz to an incremental 50 Hz, 0.5 kOe modulation field (H_o = 96 kOe) with a 100μA d.c. current input; it is plotted on the same angle axis as the rest of the figure (the field modulation technique permits enhancement of the oscillations relative to the more slowly varying MR background). Assuming closed orbits generated by couplings only in the a-b plane, c* is precisely the direction on which the oscillations should center.

A rotation diagram with large (divergent) MR in most directions, such as we have in Fig. 1, is the signature of a compensated Fermi surface.[5] If the Fermi surface were uncompensated, we would expect the MR to saturate in most directions. The saturating MR for H ~ ∥ b' arises only because the current direction is normal to the open orbit direction.

Finally, note that the magnitude of the relative MR for H ~ ∥ c* is about 16 at 4.0 K. At 1.1 K in the same field this number goes up to about 50. Rudimentary theory, with $\Delta R/R \sim (\omega_c \tau)^2$, where ω_c is the cyclotron frequency and τ is an isotropic scattering time implies $\omega_c\tau \sim 7(4$ at 4.0 K). This yields, assuming $m^* = 1$, $\tau \sim 10^{-12}$ sec, a value reasonably consistent with the d.c. conductivity.[2] More importantly, it implies, together with the fact that the qualitative shape of the rotation diagrams is always the same (although gradually diminishing in magnitude as the field is lowered), that we are in the "high field" regime even for fields, below (or temperatures above) the onset value for oscillations. This is only possible if there are still closed orbits at relatively low fields. Hence, the threshold for oscillations does not signal a change in the character of the Fermi surface from quasi-one-dimensional to two-dimensional.

We are faced with the fact, however, that the Fermi surface predicted by one-electron theory is quasi-one-dimensional.[6] There is nowhere near enough b-axis coupling to close the Fermi surface, no matter which estimate one uses.[7] Indeed, even if the Fermi surface could be closed by a much larger t_b than is presently supposed, there would still be no way in one-electron theory to reconcile the resultant large-area orbits with the very low SdH frequency that is observed (corresponding to 1% of the Brillouin Zone cross-section).

If, however, there is near-perfect nesting of the Fermi surface, closed orbits and a low SdH frequency would be expected.[8] For example, Horovitz, et al.[9] have claimed that the persistence of spin-density waves into the metallic state, as first proposed by Greene and Engler,[7] leads to compensated closed orbits of area $\sim(t_b/t_a)^2$. The relatively small anisotropy estimated from this, $t_a/t_b \sim 10$, is totally inconsistent with a picture of wide-range quasi-one-dimensional superconducting fluctuations.[10] It is clear that any reasonable model capable of generating a closed two-dimensional Fermi surface must include enough transverse coupling to depress tremendously the range in which fluctuations are significant.

THRESHOLD FIELD

We turn now to a consideration of the threshold field for oscillations (H_T). This phenomenon constitutes in itself a very interesting but puzzling riddle, and yet appears to be intimately related to the same physical processes governing the more widely studied effects.

The first thing to note about the anomaly at H_T is that it is very sharp. This may be seen in Ref. 1 (Fig. 2). It is shown more graphically here in Fig. 2. We have reproduced raw data on the oscillations of a $(TMTSF)_2PF_6$ sample. Plotted is the third harmonic modulation response of a sample (as in the inset to Fig. 1) versus magnetic field at several temperatures. The anomaly associated with H_T is indicated by the arrows; the other peaks in the picture are SdH oscillations. As the temperature was increased from 1.08 to 1.66 K, H_T increased from 67 to 80 kOe, swallowing up two oscillations in

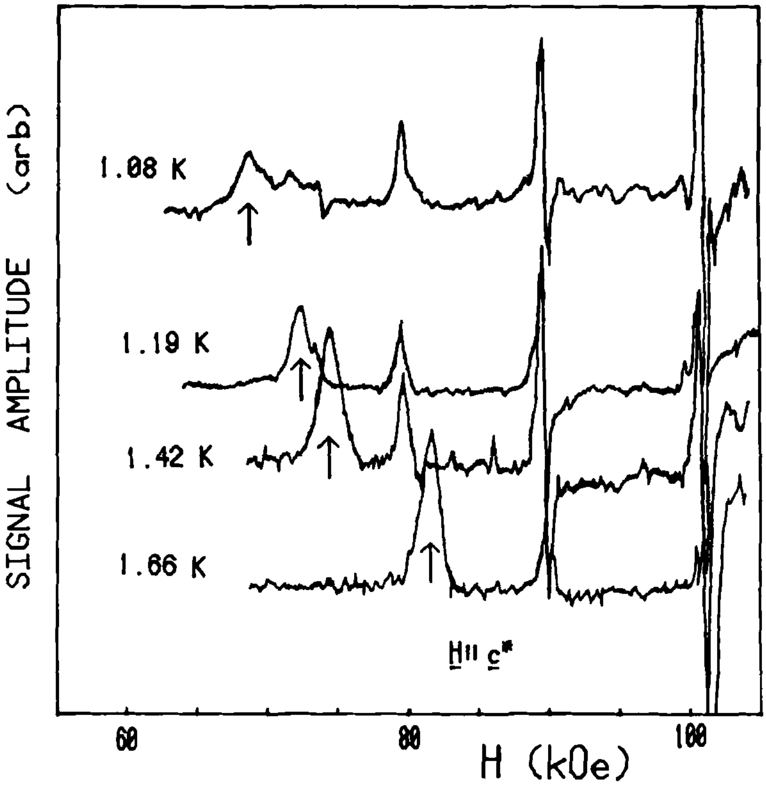

FIGURE 2 Raw data showing the threshold anomaly (indicated by arrows) in a $(TMTSF)_2PF_6$ crystal at several temperatures under 6.9 kbar pressure. The other peaks are Shubnikov-de Haas oscillations (see text).

the process. Thus, not only is the anomaly at H_T narrow ($\Delta H/H \sim 1/40$), but the oscillations vanish abruptly as they are passed by H_T. Both of these properties are included in describing the anomaly at H_T as "sharp."

This sharpness of the anomaly eliminates the possibility that field-induced breakdown of orbits ("magnetic breakdown") could be responsible for the appearance of the oscillations. That effect would produce a much more gradual field dependence of the oscillation amplitudes than is observed: $\sim [\exp(-H_o/H)]^{n/2}$, where H_o is the "breakdown field," and n is the number of breakdown points associated with an orbit.[11] Moreover, the shape of the MR rotation diagram would change fundamentally in the breakdown region, contrary to what is observed. Finally, the breakdown fields observed in other systems are not temperature dependent, as is the anomaly here.[5]

Fig. 3 shows the dependence of H at constant temperature and pressure on the angular deviation of the field direction away from the c* direction. The curve is the function $H(\theta) = H(0) \sec\theta$, with $H(0) = 60$ kOe. It is clear from this that the anomaly depends on the projection of the field on c*. This same angular dependence was observed for a strong enhancement of the proton spin-lattice relaxation rate.[3] In addition, the same temperature dependence is obtained at 6.9 kbar (the only pressure at which systematic measurements of H_T from the magnetotransport have thus far been made). This is shown in Fig. 4. We are confident, therefore, that these two very different experiments are both seeing the same physical phenomenon.

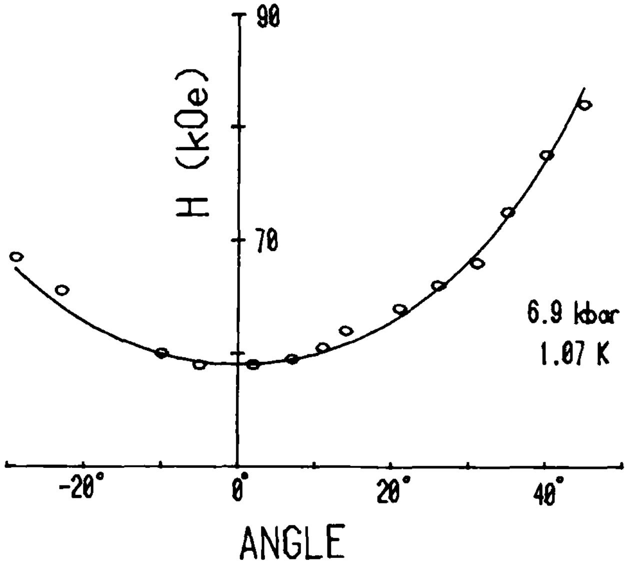

FIGURE 3 Dependence of the magnitude of the threshold anomaly field in $(TMTSF)_2PF_6$ on its orientation relative to the c* axis. The curve is the function $H(\theta) = H(0) \sec\theta$ (see text).

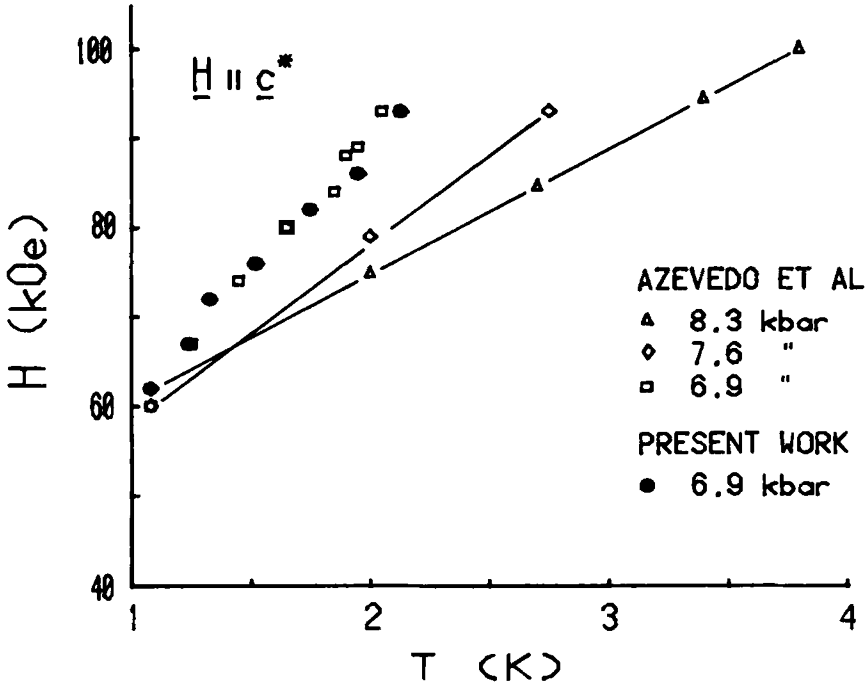

FIGURE 4 Temperature dependence of the threshold anomaly in $(TMTSF)_2PF_6$ at several pressures. Data of Azevedo et al. (Ref. 3) were taken by NMR. The agreement between the NMR and transport results at 6.9 kbar is apparent (see text). The lines are guides to the eye.

There is no independent-electron mechanism we know of which could explain these observations. It therefore seems reasonable to propose that the anomaly is associated with the persistence of spin-density waves into the metallic state. Since we know the Fermi surface to be unaltered at the anomaly, the most likely mechanism would be one involving a disorder-order transition of the electronic spin system: H_T might be the analog in the metallic state of the spin-flop field observed at ambient pressure,[12] or even the point at which the antiferromagnetic ground state is completely quenched. On the other hand, it must be noted that the temperature dependence of H_T is positive, contrary to what would be expected thermodynamically for a system with no fluctuations (of the spins) at all. Thus it appears that spin-density waves and dimensionality are ubiquitous factors in the physics of $(TMTSF)_2PF_6$.

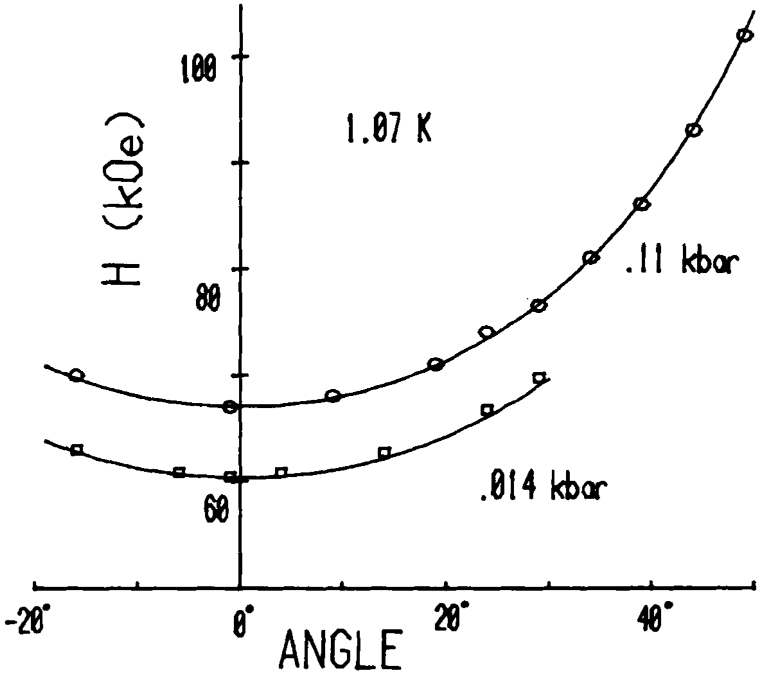

FIGURE 5 Orientation dependence (cf. Fig. 3) of the anomaly in $(TMTSF)_2ClO_4$ for two pressures. The curves represent $H(\theta) = H(0) \sec \theta$.

If $(TMTSF)_2PF_6$ were the only system to exhibit this anomaly, the line of reasoning above would be greatly weakened. However, there is reason to believe that this is not the case. We have begun to make measurements on $(TMTSF)_2ClO_4$. Although it is not clear yet whether or not there are oscillations, we have observed a very similar anomaly in the one sample thus far examined. The same angular dependence is seen, as is shown for two different pressures in Fig. 5. Note that the field magnitudes involved are also about the same (> 60 kOe). The temperature dependence of the anomaly at different pressures is shown in Fig. 6, and is again seen to be

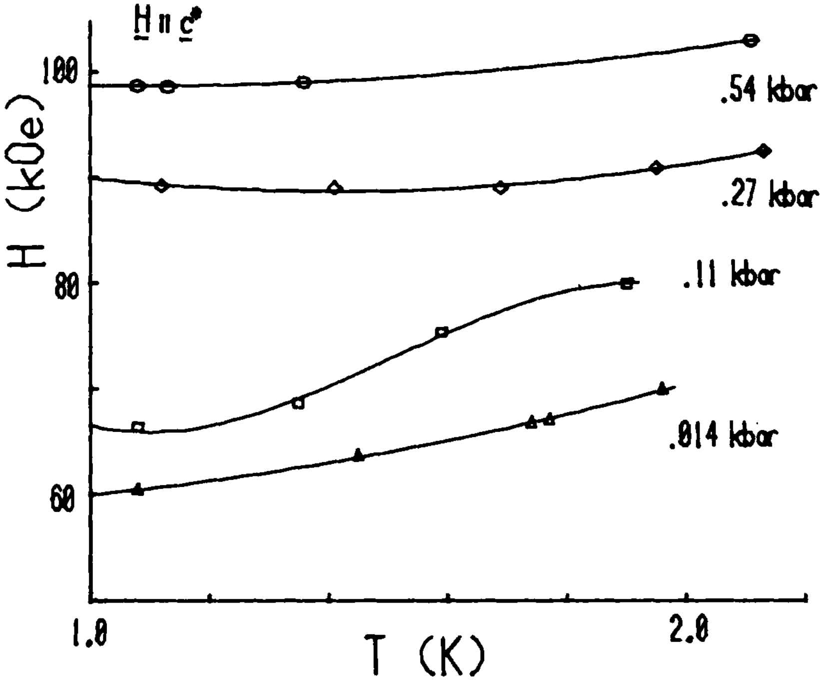

FIGURE 6 Transport data on the temperature dependence of the anomaly in $(TMTSF)_2ClO_4$ at several pressures. The lines are guides to the eye.

positive. The most striking thing, however, is the difference in the pressures at which the effect is observed for field magnitudes we can obtain: 1/2 kbar or less. It appears that the anomaly is tied to the metal-insulator transition in some sense, perhaps by a narrow domain where fluctuations are significant.

SUMMARY

We have shown from the magnetotransport that the Fermi surface of $(TMTSF)_2PF_6$ is effectively closed two-dimensionally and is compensated in the metallic state at low temperature. These results are inconsistent with a broad regime of one-dimensional superconducting fluctuations.

Independent electron theory is seen to be inadequate to explain either the Fermi surface or the threshold field for oscillations, thereby implying that correlation effects are still important in the metallic state. Finally, an anomaly has been observed in the magnetotransport of $(TMTSF)_2ClO_4$ which is analogous to the threshold field in $(TMTSF)_2PF_6$.

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