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Further Magnetotransport Studies of $(TMTSF)_2PF_6$

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FURTHER MAGNETOTRANSPORT STUDIES OF $(TMTSF)_2PF_6$

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Abstract We report new magnetotransport studies on $(TMTSF)_2PF_6$. Hall Effect measurements at 7 kbar confirm earlier magnetoresistance measurements indicating the occurrence of magnetic quantum oscillations with a threshold field. The Hall resistance shows step-like structure similar to that in $(TMTSF)_2ClO_4$, although the step magnitudes are not consistent with the quantum Hall effect. The Hall voltage changes sign near 100 kOe, also similar to an effect reported for the ClO_4 salt, suggesting that the explanation in terms of anion ordering given for that effect is incorrect. Data taken near 6.3 kbar, in a regime where the SDW transition temperature is suppressed by pressure to less than 4K, show a strong increase of the transition temperature in a magnetic field.

INTRODUCTION

The 2:1 salt of tetramethyltetraselenafulvalene (TMTSF)[1] and hexafluorophosphate (PF_6), $(TMTSF)_2PF_6$, was the first organic compound discovered to superconduct.[2] Since then there has been a general effort to understand the physics of the TMTSF family of organic conductors. The current state of this effort is described in many of the papers of these proceedings. We will concentrate here on magnetotransport in the PF_6 and ClO_4 salts.

Prior magnetoresistance study of $(TMTSF)_2PF_6$ under pressure

$(TMTSF)_2PF_6$ at ambient pressure undergoes a metal-insulator (MI) transition at T_{MI}=12K[1]; the ground state has been found to be a spin density wave (SDW).[3] Pressure suppresses T_{MI} until, above 6.5 kbar, the system is metallic with a superconducting ground state.[2] An early magnetoresistance study in this regime uncovered the existence of a threshold field above which the resistance increases steeply and "wiggles".[4] NMR studies proved the occurrence of a phase transition at the threshold and uncovered its anomalous temperature dependence.[5] The wiggles were seen to be periodic versus inverse field, as expected for the Shubnikov-de Haas effect, with a frequency of 760 kG, from which were inferred closed orbits on the Fermi surface enclosing 1% of the Brillouin zone cross section. The observed magnetoresistance anisotropy implied compensation of the Fermi surface. Both the threshold field and frequency depend on field only through its c-axis component. Finally, no field-hysteretic effects were observed such as might suggest a first-order transition.

These results were interpreted as implying a Fermi surface nested at high fields by a SDW (of undetermined nature) to produce compensation and small closed orbits oriented along the crystal c axis.[6] Because of the many anomalous aspects of the data, it was not assumed that the wiggles observed represent an "ordinary" Shubnikov-de Haas effect, only that their origin lay in an essentially two-dimensional Fermi surface as described above.

Magnetotransport studies of $(TMTSF)_2ClO_4$

Because of the requirement of high pressures in the PF_6 studies, most subsequent work has concentrated on the ambient-pressure superconductor $(TMTSF)_2ClO_4$.[6-11] The relevant findings are summarized as follows: There is a threshold field with temperature and angular dependences similar to those of $(TMTSF)_2PF_6$. While there are "wiggles" above the threshold, they are not well described as periodic in inverse field. There are hysteretic effects associated with some of the features. At very high fields (100 kOe), new wiggles gradually appear which are periodic in inverse field (frequency 2.7 MG).

The most important findings come from the Hall effect: The Hall resistance is very small below the threshold. Above the threshold, the Hall resistance increases dramatically, more or less in a series of steps or plateaus. The amplitudes of the plateaus are of about the

right size for the quantum Hall effect (QHE),[12] but are very temperature dependent; moreover, the relative amplitudes of neighboring steps are usually not as expected for the QHE, nor does the magnetoresistance decrease toward zero between steps as expected.

The experimental situation is far too complex (and incomplete) to be overly confident in any particular interpretation which has been suggested. There does seem to be some consensus however, that the Hall effect steps reflect a (series of) phase transition(s) reducing the number of carriers (hence the hysteresis) coupled with a complex version of the quantum Hall effect.[9,10]

The data on $(TMTSF)_2PF_6$ we will present in this paper reveal several new areas of similarity between the PF_6 and ClO_4 salts, particularly the finding of steps in the Hall effect. But our data also serve to reemphasize the fact that there are significant differences: the absence of any hysteresis associated with the Hall steps and their periodicity with inverse field. Thus we remain in the situation where new information generates more questions than answers.

EXPERIMENTAL DETAILS

The samples were mounted on 25μm gold wires using evaporated gold and gold paint. Six contacts were applied: two on the ends for current, and two pair arranged perpendicular to the current and to the crystal c axis to serve as Hall probes.

Three samples have been run thus far. In all three we have encountered the problems of currents not exactly along the highly conducting a axis and/or large unnested voltages. Thus, we have been unable to eliminate resistive contributions to the signals from the Hall contacts. Since our superconducting solenoid is rotatable, the Hall data were taken by measuring the voltage versus field at one angle, rotating the magnet 180 degrees, doing the measurement versus field again, then taking the difference. When hysteretic effects were evident, the 180 degree separated traces would both be taken with the field magnitude sweeping the same way. The location of the crystal c axis was determined within 1 degree as an extremum in the angular dependence of the magnetoresistance.[6]

The presence of a resistive component in the signals from the Hall leads, although theoretically cancelled by

the rotation and subtraction procedure, in practice limited the degree of accuracy of the Hall measurements. One reason is that the resistance in high fields is especially sensitive to temperature. A systematic variation of only .02K during the lengthy (about 2 hours) Hall measurement could cause an error of up to 1 milliohm, comparable in size to the smaller Hall steps. The other factor is the sensitive angular dependence of the resistance at high fields. This problem was minimized by working at the extremum in the resistance rotation curve; the extremum is offset from the normal to the b axis, however, by an angle of 6 degrees.[6] If one worked at the angle normal to b (as one theoretically should to have an ideal Hall configuration), then the angular dependence of the resistance combined with the 1 degree accuracy in angle could cause several milliohms error. Note, however, that the present procedure at least allows control over the angle at which one works and a method of determining where one is.

Pressure was applied using helium gas (which solidifies upon cooling).[13] The accuracy in controlling pressure by this technique is .2 kbar, while the resolution is .05 kbar. The high degree of confidence in the pressure thus obtained allowed us to successfully straddle the boundary between SDW and superconducting ground states.

The sample, contained within a high-pressure probe, was cooled by slow lowering into the helium-filled crystat. The entire cool-down procedure took 4-8 hours. Pressure on a sample was changed by warming to 70K to thaw the solid helium and recooling after the pressure adjustment. Temperatures down to 1.04K were obtained by pumping on the liquid helium bath.

RESULTS

Field dependence of the SDW transition

Figure 1a shows the resistance versus temperature in zero field and at 6.3 kbar pressure of the first sample tested. Note the minimum in resistance at 2.25K, which was taken as the transition temperature. Figure 1b shows the magnetoresistance of the same sample at the same pressure for T=3.23K. The transition was taken as the point where the slope increases (the small hump at the transition is peculiar to this sample).

These and similar data were used to generate the transition temperature versus field curves shown in figure

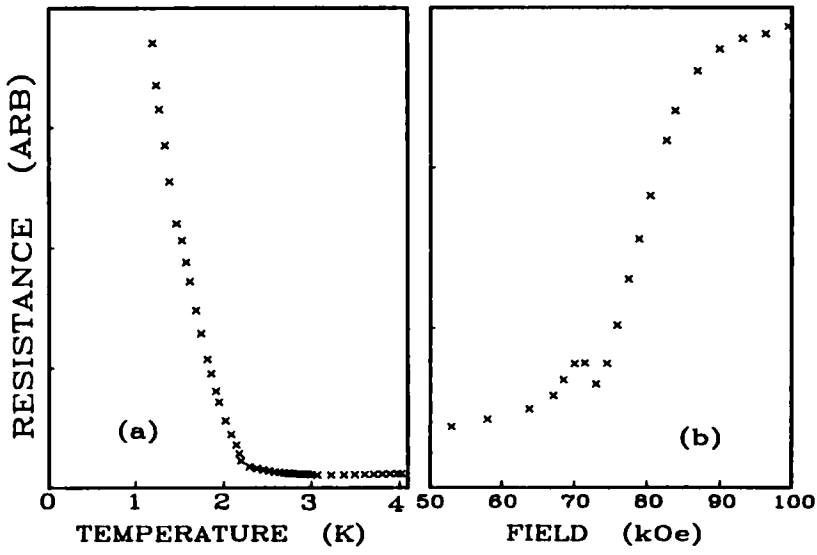

FIGURE 1. Resistive behavior of $(TMTSF)_2PF_6$ (sample 1) at 6.3 kbar (a) versus temperature in zero field, and (b) versus field at 3.23K.

2. The presence of hysteresis in the transition is indicated by the pair of dots at each temperature (the higher-field dot is for increasing field magnitude in each case). Note that one of the curves is for sample 1 at 6.3 kbar and the other two are for sample three at 6.1 and 6.3 kbar. It should be remembered that the transition temperature is very sensitive to pressure in this regime, and our absolute pressure accuracy is .2 kbar. Hence it is not surprising that the two curves at 6.3 kbar do not overlay; in fact, it is only through use of the solid helium pressure generation technique that we were able to come as close as we did.

The curves in figure 2 clearly show upward curvature and so it is not surprising that the simple empirical relation $T_c=T_{c0}+AH^2$ should fit the data. It is noteworthy, however, that such a fit, with only one adjustable parameter, A, for each curve, is off by no more than 1% at any point. The curves shown were actually drawn using this fit with $A=1.94\times10^{-4}$ for curve 1-6.3, 1.82×10^{-4} for 3-6.1, and 2.48×10^{-4} for 3-6.3.

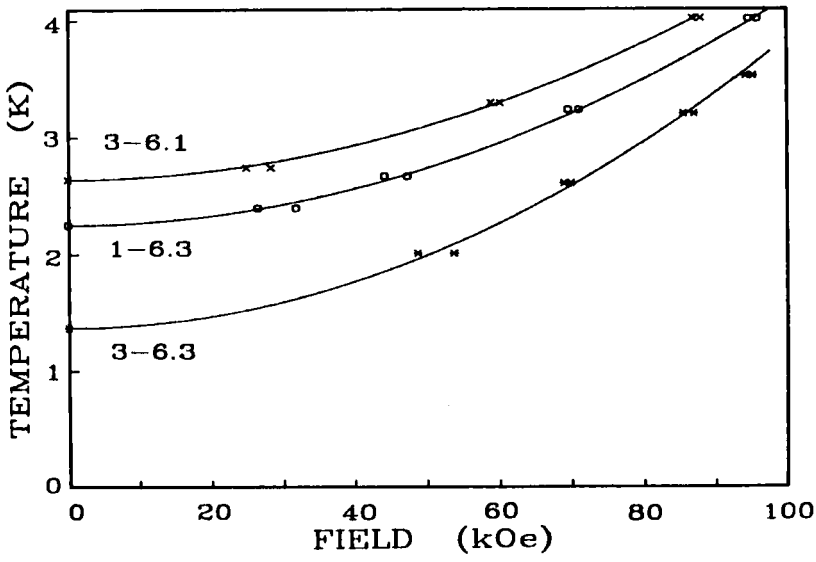

FIGURE 2. Spin density wave transition temperature versus magnetic field for $(TMTSF)_2PF_6$ samples near the critical pressure separating the SDW and superconducting states. "1-6.3" refers to sample 1 at 6.3 kbar, etc.

Hall effect at 7.0 kbar

Representative data for the Hall resistance versus field of a sample at 7.0 kbar and 1.04K are shown in figure 3. The absolute accuracy in the measurement was about 1 mΩ due to the limitations discussed earlier. The sign of the Hall voltage was not determined, the sign used here being chosen arbitrarily. There were no hysteresis effects observable in any of the 7 kbar data.

Our accuracy limitations at low fields gave a lower bound of .01 cm^3/coulomb to the Hall constant we could measure. Thus, our failure to observe any measurable Hall effect at low fields is consistent with the value .004 cm^3/coulomb obtained for the ClO_4 salt.[10]

Plotted in figure three for reference is the resistance versus field. The threshold is seen to occur at 64 kOe. Analysis of the wiggle positions shows a good fit to periodic behavior with a frequency of 750 kG, in excellent agreement with the earlier data.[4]

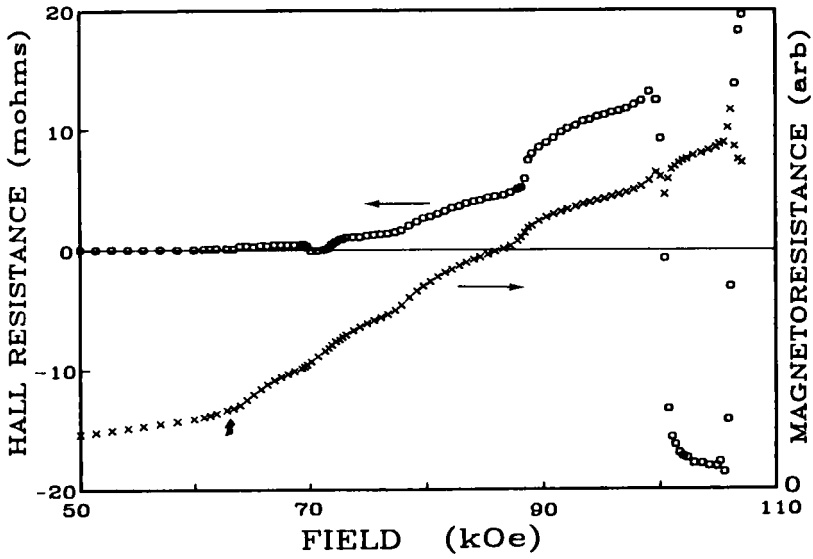

FIGURE 3. Hall resistance and magnetoresistance versus field for $(TMTSF)_2PF_6$ (sample 1) at 1.04K and 7.0 kbar. The threshold field is marked by a heavy arrow.

DISCUSSION

The key feature of the Hall data at 7 kbar is the step-like structure of increasing magnitude as field is increased. We assume the failure of the Hall resistance to saturate at each step to be due to thermal effects. We are able to resolve five steps from figure 2 with magnitudes of .5, 1.3, 4.3, 11.5, and -18.0 milliohms in ascending order. Our accuracy limitations preclude quantitative analysis of the step ratios in terms of the quantum Hall effect. However, the QHE predicts $\rho_H = 120 m\Omega/i$, where i is the number of occupied Landau levels. It does not appear possible that the steps could correspond to reasonable values for i, even given our accuracy limitations.

We noted earlier that the Hall steps in the ClO_4 salt have been interpreted as signaling a series of phase transitions progressively reducing the carrier number. This interpretation seems inapplicable to the PF_6 salt because

of the well-defined frequency associated with the magnetotransport: one would expect a change in carrier number to change the Fermi surface and hence the frequency.

Another striking feature of the 7 kbar data is the fact that the highest-field step is opposite in sign to the preceding steps. We do not understand the origin of this effect. It is noteworthy, however, that a similar effect was seen in ClO_4, but only when cooled at an extremely slow rate (much slower than necessary to produce the so-called relaxed state).[14] The interpretation applied in that instance was that such careful treatment is necessary to produce complete ordering of the ClO_4 anions, ordering which fundamentally alters the nature of the Fermi surface.[15] But there is no evidence for an analogous anion ordering effect in the PF_6 salt, and if there were, our relatively speedy cool-down should have quenched it, so the Fermi surface is presumably of the simpler nature expected in the absence of such ordering. It seems reasonable to propose, therefore, that the sign change is not related to the anion ordering per se, but perhaps to the absence of disorder.

The most significant aspect of the SDW transition temperature data is that the temperature increases with field, contrary to what one might expect thermodynamically, since the field should act to disrupt antiferromagnetic ordering. On the other hand, the behavior is analogous to that of the threshold transition at higher pressures.[5] Significant differences in the two cases, however, lie in the absence of hysteresis at the higher pressures and the absence of wiggles in the resistance at fields above the SDW ordering field.

A further mystery is that the SDW ordering temperature at 12K shows no field dependence.[16] The magnitude of the variation seen here is about the size expected from field effects on the electron spin (Remember the sign is wrong!), $H/T \approx 15$ kOe/K = μ/k_B. One should therefore have seen a 10% effect at ambient pressure in a 100 kOe field.

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