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Field scaling and exponential temperature dependence of the magnetoresistance in (TMTSF)₂PF₆

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The magnetoresistance of the quasi-one-dimensional organic conductor $(TMTSF)_2PF_6$ is studied under a hydrostatic pressure of about 0.8 GPa in magnetic fields up to 12 T perpendicular to the conducting a-b planes in the temperature range of 5 to 35 K. Both in the best conducting a and second best conducting b directions, the magnetoresistance follows a power law $\Delta R/R = (B/B_0)^{3/2}$. The a-b plane anisotropy is field independent. The scaling field B_0 follows an exponential temperature dependence $B_0 \propto \exp(T/T_0)$ with $T_0 = 10$ K. These findings are discussed in terms of recent theories of the magnetoresistance in $(TMTSF)_2PF_6$. [S0163-1829(99)51536-1]

It has been long suspected that the reason underlying the peculiar behavior of certain "metals" is that Fermi liquid (FL) theory is inappropriate to describe their elementary excitations. A FL in three dimensions (3D), however, is notoriously stable (except for the superconducting instability), whereas in 1D there is a well-understood non-FL ground state called a Tomonaga-Luttinger liquid. Although this latter state is equally notorious for its unstable nature, there is a consensus that reduced dimensionality is an essential ingredient of the non-FL problem. The quest for documenting non-FL behavior therefore focuses strongly interacting highly anisotropic metals. One of the prime candidates is the quasi-1D organic conductor (TMTSF)₂PF₆ [bis(tetramethyltetraselenafulvalene)-hexafluorophosphate] whose transport properties in a magnetic field—the subject of the present study—are indeed very peculiar.^{2,3}

The anisotropy of (TMTSF)₂PF₆ is well characterized by the room-temperature conductivities along the conducting chains (a direction) and perpendicular to the chains (b and c directions): σ_{aa} : σ_{bb} : σ_{cc} = 90:1:(1/700) with σ_{aa} \approx 1000 Ω^{-1} cm⁻¹ (Ref. 4). The conductivities can be related to the electron hopping integrals to obtain a set of characteristic energies; approximate values for those energies are: t_a =3000 K, t_b =250 K, and t_c =7 K. These values hint for the possibility of two dimensional crossovers during a cool-down from room temperature. A good test in this regard is the temperature dependence of the conductivities in different spatial directions. As a guideline, temperatureindependent conductivity ratios in all spatial directions suggest coherent electronic transport and a FL state. Reversing the argument, different temperature dependencies in different directions is a case for incoherently coupled non-FL's. In $(TMTSF)_2PF_6$ both the a-b and a-c anisotropies are temperature independent below about 80 K until the transition to a spin-density wave (SDW) insulator state is reached⁴ at T_{SDW} = 12 K. Moreover, the resistivities are well described by a power law T^{α} with the exponent α close to 2, and this behavior seems to be consistent with FL theory.⁵

The behavior in a magnetic field perpendicular to the conducting a-b planes, on the other hand, is very difficult to explain in the framework of FL theory.^{2,3} These effects are best investigated if a hydrostatic pressure is applied to suppress T_{SDW} . In the present paper, we apply a pressure of p \approx 0.8 GPa for which $T_{\rm SDW} \approx$ 5 K (in zero magnetic field). The temperature dependence of the resistance R_a (current parallel to a) is shown in Fig. 1 for zero magnetic field as well as in magnetic fields B = 6 T and 12 T perpendicular to the a-b planes. Above 40 K, the magnetoresistance is weak as expected for a metal with an open Fermi surface. At lower temperatures, however, a very strong magnetoresistance develops. As a result, the metallic temperature dependence crosses over to an insulating behavior dR/dT < 0 with a resistance minimum occurring at a field-dependent temperature $T_{\rm min}$. In our case, $T_{\rm min}$ = 19.5 K in 6 T and 27 K in 12 T. Decreasing the temperature even further, the high-field resistance saturates at $T_{\rm sat} \approx 8$ K and varies little until the SDW transition is reached.

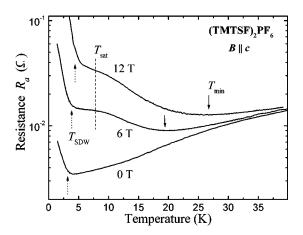


FIG. 1. Resistance R_a in the a direction as a function of temperature without magnetic field and in fields of 6 and 12 T (sample no. 1, p = 0.75 GPa). The meaning of the characteristic temperatures indicated in the figure is explained in the text.

More insight has been gained from the investigation of the magnetoresistance as a function of the orientation of the magnetic field. Chashechkina and Chaikin⁶ have shown that above $T_{\rm sat}$, the resistance depends only on the component of the magnetic field perpendicular to the a-b plane, a behavior characteristic of 2D transport. Below $T_{\rm sat}$, however, an interesting 3D picture emerges: If B is oriented along a real-space lattice vector of the crystal ("commensurate direction"), then the resistance decreases with decreasing temperature. If, however, B is along an incommensurate direction, then B continues to increase below $T_{\rm sat}$.

Most attempts to explain these findings operate with the conjecture that the magnetic field lowers the effective dimensionality of the system (decouples the planes or chains), and the low-dimensional system, in turn, is unstable against the formation of (or scales towards) an insulating non-FL state. Strong, Clarke, and Anderson⁸ suggest that the system is a collection of coherent 2D planes below $T_{\rm sat}$ at commensurate field directions whereas the c-direction coherence is destroyed by a magnetic field in an incommensurate direction. Behnia et al. 9 explain the existence of T_{min} in the similar compound (TMTSF)₂ClO₄ by the "one-dimensionalizing" effect of the magnetic field perpendicular to the a-b plane. As pointed out by Gor'kov and Lebed, 10 the Landau quantization in this quasi-1D metal for a field parallel to c results in quantized motion along b and a 1D dispersion along a. The "cyclotron gap" for laboratory fields, however, is much smaller than the coupling t_h .

In the present study we focus on the magnetoresistance in the temperature range of 5 K<T<35 K with the field perpendicular to the a-b plane and with the current flowing along the a or b directions. Our original aim—with the "one-dimensionalization" picture in mind—was to infer a characteristic magnetic field separating the high-field "1D" and the low-field "2D" states. We find no such characteristic field as the resistivities follow a power law as a function of magnetic field both in the a and b directions:

$$\rho_{aa(bb)} = \rho_{aa(bb)}^{0} \{ 1 + [B/B_0(T)]^{\beta}] \}. \tag{1}$$

The a-b anisotropy is field independent, i.e., the parameters B_0 and β are the same for the a and b directions. For the exponent we find a temperature-independent value β =1.49 \pm 0.1. The temperature dependence of the scaling field B_0 is well described by an exponential form

$$B_0(T) = B_{00} \exp(T/T_0) \tag{2}$$

with $T_0 \approx 10$ K.

We have measured the magnetoresistance of two (TMTSF)₂PF₆ single crystals. In sample no. 1, four electrode contacts were applied to each of the two opposite *a-c* sides of the crystal allowing measurements with current flowing either in the *a* or *b* directions. In sample no. 2, four contacts on the *a-b* plane allowed measurements with current flowing parallel to the conducting chains only. Hydrostatic pressure was applied to the samples in a Cu-Be pressure clamp with kerosene as pressure transmitting fluid. The pressure values communicated in this article were calculated from the force applied to the pressure cell taking into account the loss of pressure during cool-down inferred from independent calibrating measurements. The crystals were vi-

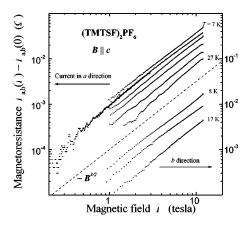


FIG. 2. Magnetoresistance vs magnetic field at temperatures (from top to bottom) 7, 10, 13, 17, 22, and 27 K in the *a* direction (above the dashed line) and 8, 12, and 17 K in the *b* direction (below the dashed line). The dashed line represents the power law $\Delta R \propto B^{3/2}$ (sample no. 1, p = 0.81 GPa).

sually aligned so that the magnetic field was always perpendicular to the conducting a-b planes.

The magnetoresistance in magnetic fields up to 12 T has been obtained from constant field temperature scans (see, e.g., Fig. 1) and constant temperature field scans. The powerlaw field dependence of the resistivity, Eq. (1), is demonstrated in Fig. 2 where the change of the resistance ΔR_a and ΔR_h is plotted against the magnetic field on logarithmic scales at several temperatures. Equation (1) is clearly obeyed above 1 T. Below this field, the relative experimental error in ΔR is high and we omitted these data points from the figure for clarity, except for the results of a 10-K sweep. In this case the magnetic field has been changed very slowly for an increased accuracy of the resistance measurement, and there is no systematic deviation from the power law down to 0.3 T. We stress that Eq. (1) is equally valid below and above the field B^* that results in a T_{\min} equal to the temperature of the field sweep; no change in the field dependence is observed at B^* . For example, in a magnetic field of 3 T, T_{\min} occurs at about 16 K. At this temperature dR/dT is positive ("metallic''), if B < 3 T and negative ("insulating"), if B > 3 T, still the magnetoresistance follows the law $\Delta R \propto B^{3/2}$ over the entire field range from 1 to 12 T.

It is also obvious from Fig. 2 that the power-law exponent is close to 3/2 in all cases. For an accurate determination of the exponent β , we have performed three-parameter fits to the form $R(B) = R(0)[1 + (B/B_0)^{\beta}]$. The results are summarized in Fig. 3 for both samples and—in case of sample 1—for both current directions a and b. Below about 20 K, there is no systematic temperature dependence and the mean value for all results is $\beta = 1.49$ with a statistical standard deviation of 0.10. The results therefore are consistent with the simple fraction $\beta = 3/2$ independent of temperature. Above 20 K a weak systematic increase of the exponent is observed. At these temperatures $\Delta R/R$ is small, and the admixture of a regular B^2 magnetoresistance may lead to an increase of the overall exponent.

Using a temperature independent exponent $\beta=3/2$, the temperature dependence of the scaling field B_0 in Eq. (1) is easily determined from a comparison of the temperature dependence of the zero-field and 12-T resistances. Figure 4

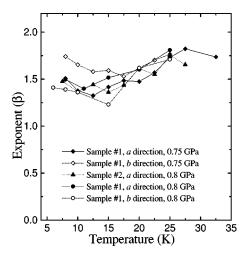


FIG. 3. Temperature dependence of the power law exponent β in Eq. (1) for two samples and two crystalline directions a and b (p=0.81 GPa).

shows $\Delta R/R = [R(12 \text{ T}) - R(0)]/R(0)$ as a function of temperature on linear temperature- and logarithmic resistance scales. The linear dependence in this representation verifies the exponential temperature dependence Eq. (2): $\Delta R/R \propto \exp[-(3/2)T/T_0]$. The deviation from the linear dependence below 8 K is due to the proximity of the SDW transition in 12 T. The results for the characteristic temperature T_0 are the following: Sample no. 1, a direction: 10.4 K, b direction 10.4 K; sample no. 2, a direction: 9.5 K. Therefore the results on both samples and crystalline directions are consistent with the same $T_0 \approx 10$ K.

We begin the discussion by pointing out that Kohler's $law^{11} \Delta R/R = f(B/R)$ together with Eqs. (1) and (2) would imply a zero-field resistance $R \propto B_0 \propto \exp(T/T_0)$, in obvious contradiction with the experimental results. Since this argument is valid at any magnetic field within the range of validity of Eq. (1), any purely kinetic explanation of the magnetoresistance is highly unlikely.

If the high-field resistance reflects precursor fluctuations of a phase transition to an SDW, one expects a divergent power-law behavior as the transition is approached from above. The exponential temperature dependence and the saturation of the magnetoresistance at about $T_0 = 9\,$ K, well above the transition temperature (typically 3 to 5 K in 12 T and close to p_c), are difficult to reconcile with this theory.

Our results raise two phenomenological arguments against the "one-dimensionalization" scenario. First, there should be characteristically different field dependencies in the "2D" low-field state and the "one-dimensionalized" high-field state, contradicting the observed self-similar power-law behavior [Eq. (1)]. Second, by virtue of the "1D nature" of the "one-dimensionalized" phase, it should exhibit a stronger a-b plane anisotropy than the low-field "2D" phase, whereas we observe no change in the a-b plane anisotropy when the magnetic field is turned on. The field- and temperature independent a-b plane anisotropy suggest a coherent electronic transport in the a and b directions. We note here that the qualitatively very similar magnetoresistance in ρ_{cc} reported in Ref. 12 makes it likely that the transport is coherent along all spatial directions even in a magnetic field. If this is so, a 3D FL state seems to be ines-

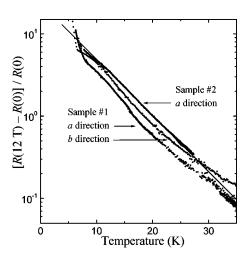


FIG. 4. Magnetoresistance vs temperature in a constant magnetic field $B = 12\,$ T. The straight line represents Eq. (2).

capable. The exponential temperature dependence and the unusual power law exponent of the magnetoresistance, on the other hand, point to a non-FL state. A purely speculative possibility is that the system is a 3D FL in the temperature range investigated, but the FL parameters are tuned by the c component of the magnetic field.

We conclude that the existing theories, including FL theory, do not seem to account for the behavior of the magnetoresistance. To provide some clue for the interpretation of the phenomenon, we make some simple phenomenological considerations. It is important to emphasize that the twoparameter (T_0, B_{00}) fit described by Eqs. (1) and (2) works equally well above and below the resistance minimum T_{\min} as well as around $T_{\rm sat}$ where the magnetoresistance saturates. This finding suggests, that the same mechanism is responsible for the magnetoresistance in the entire temperature range investigated. In particular, we find no difference in the behavior below and above the temperature of the resistance minimum, T_{\min} , or below and above the magnetic field B^* corresponding to T_{\min} . Similarly, Eqs. (1) and (2) do not break down near $T_{\rm sat}$ where the high-field resistance saturates. The saturation occurs because—according to Eq. (2)— B_0 varies little below T_0 . Since T_0 is inferred from a "2D" temperature range where the magnetoresistance depends on the component of the field perpendicular to the a-b plane only, $T_{\rm sat} \approx T_0$ is likely to be an a-b plane property, and $T_0 \approx t_c$ may be a coincidence. ¹³

Finally we note that the exponential temperature dependence of the magnetoresistance with the exponent proportional to the temperature is highly unusual. By a purely formal analogy, this temperature dependence may point towards the thermal excitations of a bosonic degree of freedom. The significance of this observation to the system investigated is, however, unclear.

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