Anomalous Magnetoresistance Anisotropy in Metallic and Spin-Density-Wave Phases of the Quasi-One-Dimensional Organic Conductor (TMTSF)$_2$ClO$_4$

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We report new features of the transverse magnetoresistance (MR) anisotropy in (TMTSF)$_2$ClO$_4$ in both the metallic phase and the field-induced spin-density-wave (FISDW) phases. In the metallic phase, the MR varies as $\Delta R_{xx} \propto H^{\alpha}$, where $\alpha < 2$ and α decreases as the magnetic field is tilted from the c^* axis. In the FISDW states, there is a dramatic enhancement of the MR anomalies at the FISDW phase transitions as the magnetic field is tilted, with particular angles for stronger and weaker enhancement. This is clear experimental evidence that the FISDW phases are not simply 2D in nature.

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Some quasi-one-dimensional organic conductors of the Bechgaard salt family exhibit dramatic behavior when a magnetic field is applied perpendicular to their most conducting planes.[1] In the most studied of these compounds, the tetramethyltetraselenafulvalene perchlorate [(TMTSF)$_2$ClO$_4$], the magnetic field induces a series of at least eight field-induced spin-density-wave (FISDW) phases between 3 and 27 T.[2-7] Each transition between FISDW phases can be detected by a transverse magnetoresistance (MR) anomaly as shown by the arrows in Fig. 1. In each FISDW phase, the Hall resistance exhibits a plateau.[3,4,8,9] More recent thermodynamical measurements, such as magnetization[5] and specific heat,[6] have confirmed the existence of true phase transitions at these MR anomalies.

The resulting phase diagram is now well understood, at least below 10 T, within the framework of a nesting model describing the instability of an anisotropic electron gas with an open Fermi surface.[10-13] The magnetic field makes the electron motion one dimensional and, as a result, favors spin- (or charge-) density-wave ordering with a small density of carriers. In the magnetic field these carriers are in quantized Landau levels. Each FISDW phase results from a competition between the nesting energy and the diamagnetic energy. The latter is minimized if the Landau levels are completely filled, which can be achieved in each phase with a variation of the nesting vector.[10,11] This mechanism leads to a quantization of the Hall effect in each FISDW phase,[11,14] as observed experimentally, and reproduces fairly well the magnetic-field dependence of all the thermodynamic quantities.[15]

Although transport experiments are the simplest to perform, they are far from being understood. In the metallic phase, the MR is unexpectedly large for current along the conducting chains (a axis).[16] Also, the MR violates Kohler's rule, an indication that the metallic-phase transport cannot be understood in terms of the Boltzmann equation.[17,18] The angular dependence of the MR has been studied at fixed magnetic field.[19,20] The resulting plots vary dramatically for different values of the field with no precise description given for the field variation. For rotation of the magnetic field in the b^*-

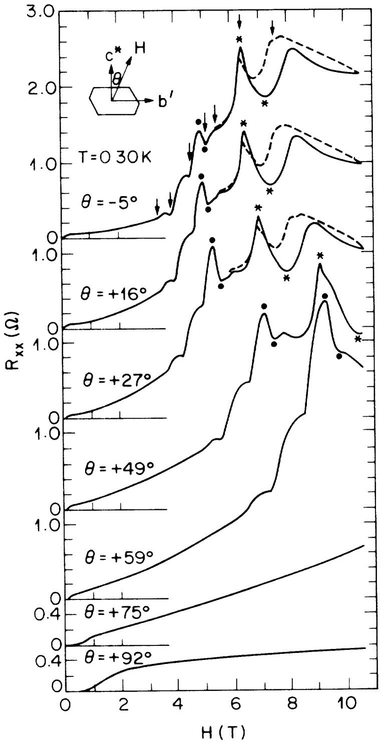

FIG. 1. Angular-dependent magnetoresistance of (TMTSF)$_2$ClO$_4$. Inset: Sample cross section; current is along the a axis (normal to the page); θ is measured from the c^* axis. Arrows mark FISDW transitions from thermodynamic measurements (Refs. 2, 5, and 6). Stars and dots mark oscillations discussed in the text. Dashed line indicates decreasing magnetic field.

c^* plane, there are minima in MR for fields along the b' and c' axes. (In this triclinic system, magnetotransport is sensitive to the b' and c' axes, which are the projections onto the b^*-c^* plane of the b and c axes which, in turn, correspond to electron motion in the a^*-c^* and a^*-b^* planes.) There is evidence of MR saturation for magnetic fields along b'.[19,20] In the FISDW phases the transport is even less understood. Although the observed Hall plateaus are consistent with theory, there are no coexisting zero-resistance states.[4,9,21] It has been suggested that the nonvanishing resistance is due to disorder giving rise to extended states in the gap.[22] Investigations with tilted magnetic fields show that the FISDW transitions occur at fixed $H_z = H\cos\theta$ out to 45°, suggesting that they are 2D in nature.[23,24]

From angular-dependent transport measurements, we find anomalous MR anisotropy in both the metallic and FISDW phases. Our samples were electrochemically grown at constant voltage ($\sim$2.5 V) in an electrolyte of $\sim 2\times10^{-2}M$ tetrabutyl ammonium perchlorate in 1,1,2-trichloroethane. A $\sim 3\times10^{-3}M$ TMTSF solution is in one leg of the cell. The crystals on the Pt electrode were up to 15 mm long and 0.5 mm wide with parallelogram or quasihexagonal cross sections. All four samples studied, each from a different growth, exhibit the behavior described below. The data presented herein are from a sample (3 mm between voltage contacts, quasihexagonal cross section with sides from 0.08 to 0.20 mm) studied in detail on several different cooling runs (same cooling rate). X-ray diffraction revealed twinned phases (180° twinning along the a axis) of roughly equal volume. Note that these twinned crystal phases share a common b^*-c^* plane, including common b' and c' axes.

Four-point resistance measurements were performed in a ^{3}He cryostat and a 10.8-T superconducting magnet. Each silver-paint Ohmic contact encircled the needlelike sample to minimize any admixture of conductivity along the b and c axes. The current was along the conducting chains (a axis), with magnetic field in the b^*-c^* plane, its direction given by the angle, θ, from the c^* axis. The samples were mounted on a rotating stage for *in situ* rotation about the a axis over a range of 130°. They were cooled in zero field and the cooling rate between 30 and 10 K was controlled at 0.15–0.20 K/min to give a relaxed ground state with sufficient anion ordering to observe clear FISDW transitions. Typical excitation currents (20 μA) and magnetic-field sweep rates (2 T/min) avoid any evidence of electron heating at T=0.3 K. Reversal of the magnetic field yielded identical MR traces, indicating no appreciable mixing in of the Hall resistance.

Figure 1 shows the resistance, $R_{xx}(H)$, versus field for various values of the angle θ. The inset shows schematically the crystallographic axes of the sample, where the a axis is perpendicular to the page and corresponds to the long axis of the needlelike crystal. Below H_{c2}, the sample is superconducting, the b' axis experimentally determined by the maximum H_{c2}. Above H_{c2}, the metallic phase exhibits a smooth, increasing MR. The anomalies at higher fields marked with arrows are the FISDW transitions, including one which exhibits hysteresis (dashed line is for downward magnetic-field sweep). The c^* axis is experimentally determined by the FISDW transitions, which are found at fixed $H_z = H\cos\theta$ out to $\theta \sim 60°$, corresponding to the limit of our magnetic field.

For the metallic phase, a non-cosθ angular dependence is not surprising, since the metallic phase is not 2D in nature. We find that the magnetoresistance obeys a power law, $\Delta R_{xx} = C_0 H^\alpha$, over a wide range of angles. This power law extends over the entire range of metallic-phase data between the superconducting and first FISDW phases. For $\theta < 80°$, the dynamic range of the data is more than an order of magnitude. At larger angles (i.e., H nearly along b'), the dynamic range of the data is greatly reduced and saturation of ΔR_{xx} is observed. Figure 2 contains the observed angular variation of the power-law prefactor and exponent. (The prefactor can be interpreted as ΔR_{xx} at H=1 T in the absence of a superconducting phase.) The open circles denote angles for which the power-law behavior is unconfirmed because the dynamic range of the data is less than 1 order of magnitude. Note that both prefactor and exponent appear symmetric about $\theta \sim -5°$. This angle corresponds well to the c' axis, which is 95.1° from the b' axis, as calculated from the unit cell at T=7 K.[25] ΔR_{xx} is nearly quadratic for the field along c' and becomes sublinear for the field close to b' (±10°). We observe that α can vary by $\sim$20% on a given sample from one cooling

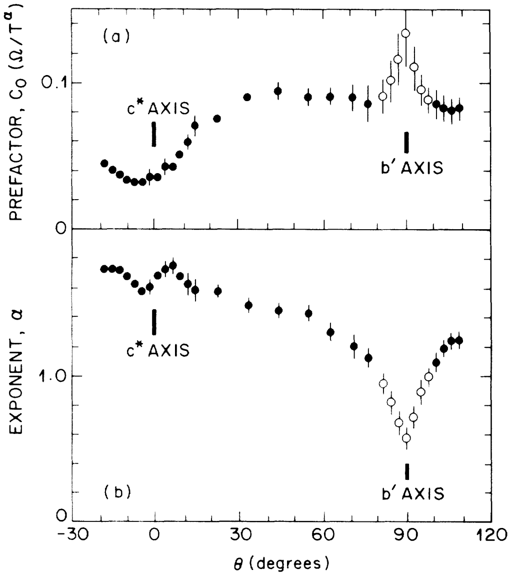

FIG. 2. Power-law magnetoresistance (a) prefactor, C_0, and (b) exponent, α. Open circles near the b' axis denote less than 1 order-of-magnitude dynamic range.

run to the next, due presumably to disorder dependence. Although angular-dependent power-law MR occurs in metals with open orbits, it is unexpected in this geometry since the open orbits on the corrugated planar Fermi surface carry current along the direction of the applied current.[26] This MR also contrasts with observations at higher magnetic fields ($8 < H < 30$ T) and temperatures ($T > 5.5$ K, above the highest critical temperature for FISDW formation), where the MR of the metallic state is essentially exponentially increasing with magnetic field.[7]

Turning our attention to the FISDW phases in Fig. 1, as the angle is increased, there is a smooth evolution of the MR traces. Specifically, there are no existing FISDW states which disappear and no new states which appear. Nevertheless, the data show a surprisingly strong angular dependence of MR for these phases. If the FISDW states were in fact 2D, the magnitude of the magnetoresistance would be unchanged at a given FISDW transition. The observation to the contrary initially suggests that there is an admixture of contributions to the MR: one from regions with FISDW ordering, which exhibit anomalies at the FISDW transitions and are thought to be 2D (depending only on H_z); the other from metallic or disordered regions without FISDW ordering (perhaps near sample edges, impurities, or microcracks) which would exhibit 3D behavior. However, from Fig. 1 it is clear that this is not the complete story: At the FISDW transitions the MR anomalies *themselves* display an angular dependence.

In Fig. 1 we identify the two best developed oscillations with pairs of stars and dots ("star" and "dot" oscillations, respectively). As an experimental measure of the "strength" or degree of development of the FISDW phase, we define $\delta R_{\mathrm{FISDW}}$ as the difference in MR between the local maximum and local minimum indicated by the stars or dots in Fig. 1. Figure 3 shows the angular dependence of the MR oscillations, $\delta R_{\mathrm{FISDW}}$, at the star and dot transitions. Despite the large variations in magnitude for different temperatures or different cooling runs (i.e., different disorder), each oscillation shows a striking reproducible angular dependence. Most dramatically, there is a remarkable strengthening of the FISDW transitions in tilted fields, yielding up to an order-of-magnitude increase in $\delta R_{\mathrm{FISDW}}$. (In one lower-quality sample, only for $\theta \gtrsim 20°$ did the dot transition appear as an oscillation in R_{xx}. For $\theta \lesssim 20°$, it was visible only as a kink in R_{xx}, as with the lower-field FISDW transitions in Fig. 1.) The strengthening of the oscillations does not saturate even at the largest angles corresponding to our maximum magnetic field. This general enhancement with increasing angle is completely unexpected and would be interesting to explore at higher angles in a larger magnetic field. In addition to the general enhancement of the FISDW transitions in tilted fields, the dot oscillation exhibits a local maximum and minimum in each of the three curves of Fig. 3(b) at $\theta = 27° \pm 45°$ and $\theta = 45° \pm 5°$. Since the magnetotransport in the metallic phase shows no features at these angles, we ascribe the observed maximum in $\delta R_{\mathrm{FISDW}}$ to either an increase in critical temperature, T_c, or a decrease in the threshold field, H_t, for FISDW formation. The observed minimum in $\delta R_{\mathrm{FISDW}}$ would then be due to opposite shifts in T_c or H_t.

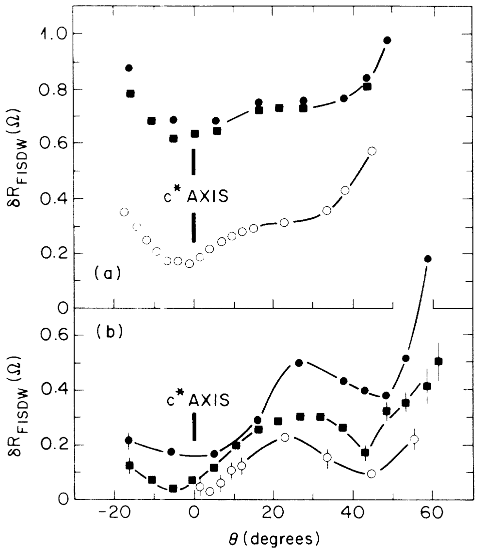

FIG. 3. Angular-dependent MR oscillations, $\delta R_{\mathrm{FISDW}}$, at the FISDW transitions denoted by (a) stars and (b) dots in Fig. 1. Circles (squares) are data taken at $T = 0.30$ (0.36) K, respectively. Open (closed) symbols refer to two separate, nominally identical, cooling runs.

There exists a theoretical model for angular dependence of the FISDW states.[27-29] At particular angles $\tan\theta = mb\sin(\gamma)/nc$ (m,n integers and b,γ,c unit-cell parameters) the electron motion on the Fermi surface in a magnetic field is bounded and periodic. This is because the two periodicities of the semiclassical equations of motion in the b^* and c^* directions are commensurate. Although such commensurate motion has strong effects on the MR of some metals, it is not expected in the metallic phase of Bechgaard salts because of the relatively featureless quasi-one-dimensional Fermi surface.

However, Lebed'[27] notes that at these special angles there is no longer any dispersion along the magnetic field and therefore predicts a decrease in H_t for FISDW formation and a new sequence of the FISDW phases. The first special angle ($m = n = 1$) is $\theta = 28.5°$, at which the plane normal to the magnetic field cuts diagonally through the Brillouin zone. Although it is tempting to identify the observed maximum in $\delta R_{\mathrm{FISDW}}$ with this special angle, other predictions of the Lebed' theory are not consistent with experiment. A study of the angular dependence of H_t has revealed no decrease at the predicted angles.[23,24] On the contrary, a 2D $\cos\theta$ dependence has been confirmed. In Fig. 1, we find no alteration in the FISDW sequence when the field in tilted. Seven transitions can be labeled and followed as the angle varies. Finally, the next special angles are $\theta = 15.2°$ ($m=1$, $n=2$) and $\theta = 47.4°$ ($m=2$, $n=1$). In Fig. 3, $\delta R_{\mathrm{FISDW}}$ shows no particular feature at $\theta = 15°$ and a strong local minimum at $\theta = 47°$.

The unchanging sequence of seven FISDW phases that we observe in tilted fields offers support to an altered version of the Lebed' description. Lebed' considers a nesting vector, $(2k_F, 0, 0)$, for which the relevant energies describing deviations from perfect nesting are $t_b \sim 300$ K and $t_c \sim 10$ K, the transfer energies between chains in the b and c directions. For the field along c^*, this means that T_c should be t_c. However, the experimental $T_c \sim 100$ mK is of order t_c^2/t_a.[10,24] This suggests a transverse nesting vector, $(2k_F, q_b, q_c)$, where q_b and q_c give the best superposition of the two sheets of the Fermi surface. In this case, reorganization of the FISDW phases in tilted magnetic fields is not expected, as it would occur only at very small angles corresponding to extremely high-order special angles.[29]

To summarize, we observe a power-law-dependent magnetoresistance of the metallic state in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ over a wide range of magnetic-field orientations. The power-law exponent decreases from $\alpha \lesssim 2$ as the magnetic field is tilted from the c' axis. In the FISDW states, there is a striking enhancement of the FISDW transitions in tilted fields and evidence of particular angles for stronger and weaker enhancement. These results cannot be understood with the current, essentially 2D, theory of the FISDW phases in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$.

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