Non-Ohmic Transport in the Field-Induced Spin-Density–Wave State in Tetramethyltetraselenafulvalinium Chlorate, $(TMTSF)_2ClO_4$

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A nonlinear transport behavior was observed in the magnetic-field–induced spin-density–wave state above $B=7.5$ T at $T=1.5$ K in $(TMTSF)_2ClO_4$. The nonlinearity appeared in the transverse conductivity, but not in the Hall conductivity. The threshold electric field was very small and undetectable in some samples. The sliding spin-density wave is one of the plausible mechanisms of this nonlinear transport. Small-period oscillations with the period $\Delta(1/B)=0.004$ T^{-1}, similar to the Shubnikov–de Haas effect, showed a decrease in amplitude with increasing electric field.

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Many novel electronic properties have been found in the tetramethyltetraselenafulvalinium family of organic conductors, $(TMTSF)_2X$ ($X=PF_6$, ClO_4, ReO_4, etc.). Among them, the magnetic-field–induced spin-density-wave (MFISDW) phase transition is one of the most remarkable phenomena found in this system.[1-9] The spin-density–wave (SDW) state is stabilized by the orbital effect of magnetic fields, and the SDW state consists of many subsidiary phases (subphases). When a magnetic field is applied parallel to the c^* axis of a slowly cooled $(TMTSF)_2ClO_4$ sample[3-8] [or $(TMTSF)_2PF_6$,[1,2] $(TMTSF)_2ReO_4$[9] under high pressure] at sufficiently low temperatures, the normal metallic phase undergoes a second-order phase transition into one of the SDW subphases at a certain threshold B_{th}, and several first-order phase transitions between different subphases occur successively above B_{th}.

The mechanism of this phenomenon has been basically explained by various authors.[10-14] The electron system of $(TMTSF)_2X$ can be regarded as an anisotropic two-dimensional (2D) electron gas with open Fermi surfaces. Under a magnetic field perpendicular to the 2D plane, electrons near the Fermi level carry out a periodic motion (wave vector $G=eBb/\hbar c$) along the open orbit (call it the x direction), and lose the degree of freedom of the motion perpendicular to this periodic motion (y direction). Consequently, the electron energy dispersion becomes one dimensional (1D) along the x direction. Because of the periodicity of the electron motion, the wave vector G plays a role of reciprocal-lattice vector in this 1D dispersion. At sufficiently low temperatures, such a system is unstable against an infinitesimal periodic potential with the wave vector which connects two "Fermi points" in this 1D dispersion. The x component Q_x of the allowed nesting vectors is not only $2k_F$, but also $2k_F+nG$ (n an integer), and the corresponding y component Q_y is determined by the condition that the Fermi surface is tangent to itself when translated by $\mathbf{Q}$.[10] This nesting vector $\mathbf{Q}$ is generally incommensurate with the lattice potential. Each nesting vector with different n corresponds to a different SDW subphase. In the nth subphase, the nesting vector $\mathbf{Q}$ opens an SDW gap at $|k_x|=k_F+nG/2$ in the 1D dispersion, and further subsidiary gaps at $|k_x|=k_F+mG/2$ [m an integer ($\neq n$)] by the periodicity of the electron motion. These subsidiary gaps correspond to gaps between neighboring Landau levels of the valleys below and above the gap produced by the nesting. The Fermi level is always located at the center of one of these gaps. In other words, n Landau levels of the carrier pocket are always fully occupied in the nth MFISDW subphase. This situation is very similar to the integral quantum Hall state in the 2D electron gas formed in metal-oxide-semiconductor or semiconductor heterostructures.

Despite the basic understanding of the MFISDW in $(TMTSF)_2ClO_4$ as mentioned above, there remain several unsolved problems concerning the details of the phenomena, such as the new phase transitions at higher fields,[15] the origin of the small-period oscillations,[15] etc.

In this paper, we report the first observation of a non-Ohmic transport behavior in the MFISDW state of $(TMTSF)_2ClO_4$. The magnetotransport measurements were carried out with a current along the a axis (x direction) in static magnetic fields up to 15 T applied to the c^* axis (z direction). The typical size of the sample was $4\times0.3\times0.2$ mm^3. Six gold lead wires (25 μm in diameter) were bonded on the samples by gold paint. Gold-paint current contacts covered the two end surfaces [(100) surfaces] and two pairs of voltage contacts were put on the (010) surface for both magnetoresistance and Hall-resistance measurements. The size of the painted voltage contacts was about 0.1 mm in diameter. To achieve the relaxed state, the samples were cooled at speeds of less than 0.2 K/min around 24 K, and annealed at about 20 K for ordering of the ClO_4^- anions.

Typical magnetoresistance traces for different current values are shown for sample 1 in Fig. 1. A sharp increase of resistance at $B=6$ T is the manifestation of the second-order transition from the normal metallic phase to one of the SDW subphases. The small kinks at

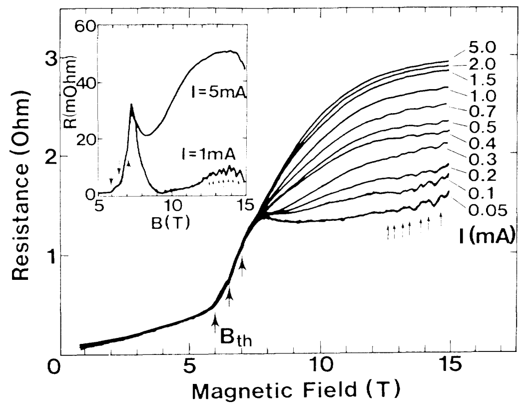

FIG. 1. Transverse magnetoresistance of $(TMTSF)_2ClO_4$ at $T=1.5$ K for different current values in sample 1. The field positions of phase transitions are indicated by large arrows. Small arrows show small-period oscillations. Inset: The magnetoresistance in another sample, 2.

$B=6.6$ T and 7 T correspond to the first-order transitions between different subphases. The small-period oscillatory structures appeared superposed on the magnetoresistance above 13 T as shown by small arrows. Their fundamental period is $\Delta(1/B)=0.004\ T^{-1}$. It is seen that above 7.5 T the magnetoresistance shows a remarkable dependence on current value. As the current was increased, the resistance increased gradually and saturated for sufficiently large current values. The field positions of the phase transitions, indicated by large arrows, showed no change for the different current values.

The inset of Fig. 1 shows magnetoresistance traces for another sample (2) which has a larger residual resistivity ratio (RRR > 200). In the limit of zero electric field, the magnetoresistivity ρ_{xx} around 10 T becomes very small and appears to tend to almost zero. This is reasonable if we consider the theoretical prediction that the density of states at the Fermi level is always zero at the MFISDW state. The nonlinearity appeared above the same field as in sample 1 (7.5 T), but more remarkably in this sample than in sample 1. A similar nonlinearity was observed in all of the five samples investigated in the present study.

Figure 2 shows the Hall resistance for sample 1. It has been reported that the Hall resistance has a steplike magnetic field dependence similar to the quantum Hall effect at sufficiently low temperatures.[5] In these traces, the steplike structures are smeared out because of the relatively high temperature. It is seen that the Hall resistance has no explicit dependence on the current value. The inset of Fig. 2 is the curve of current (I) versus Hall voltage (V_H) at 14.9 T. It should be noted that this I-V_H characteristic is nearly Ohmic.

The magnetoresistivity ρ_{xx} and the Hall resistivity ρ_{xy} are represented by conductivity tensor components σ_{ij}: $\rho_{xx}=\sigma_{yy}/(\sigma_{xx}\sigma_{yy}+\sigma_{xy}^2)$, $\rho_{xy}=-\sigma_{xy}/(\sigma_{xx}\sigma_{yy}+\sigma_{xy}^2)$.

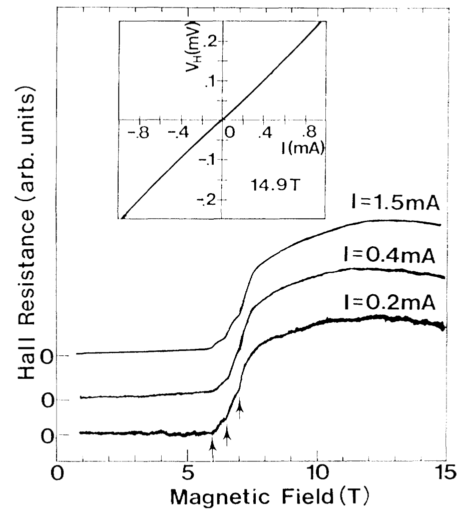

FIG. 2. Hall resistance of $(TMTSF)_2ClO_4$ at $T=1.5$ K for different current values in sample 1. Inset: The current vs Hall-voltage curve at $B=14.9$ T.

According to Oshima *et al.*,[6] $|\sigma_{xy}| \gtrsim \sigma_{xx}$, $\sigma_{xx} \simeq 5\sigma_{yy}$ in the field region $B \geq 8$ T. Therefore, the σ_{xy}^2 term is dominant in the denominator of ρ_{xx} and ρ_{xy}. The nonlinearity appears drastically in $\rho_{xx} \simeq \sigma_{yy}/\sigma_{xy}^2$, but not explicitly in $\rho_{xy} \simeq -1/\sigma_{xy}$. Therefore, the nonlinearity can be ascribed to that of σ_{yy}. As the electric field is increased, the transverse conductivity σ_{yy} increases gradually and saturates for sufficiently high electric fields ($E \simeq 100$ mV/cm). The Hall conductivity σ_{xy} has no nonlinearity versus electric field.

Figure 3 shows the electric field dependence of the resistance for sample 2 at several magnetic fields. The value of the electric field strength may include some uncertainty because of the finite size of the contact area and the inaccuracy of the estimation of the sample cross

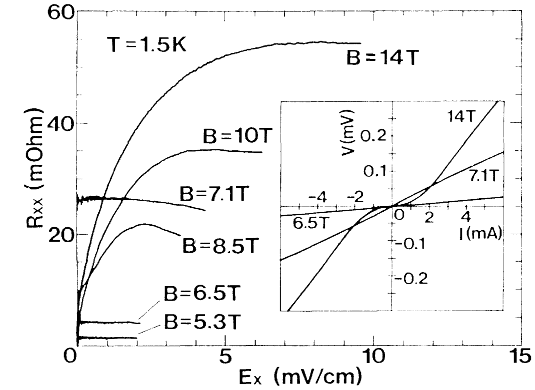

FIG. 3. Electric field dependence of the transverse magnetoresistance R_{xx} ($\sim\sigma_{xx}/\sigma_{xy}^2$) for sample 2 at several magnetic fields. Inset: The raw I-V curves.

section. The raw data of the I-V curve are shown in the inset. In the normal phase and the low-field SDW subphase, the resistance shows an Ohmic behavior. The nonlinearity appears in magnetic fields above 7.5 T at 1.5 K. Above 10 T the resistivity ρ_{xx} (nearly proportional to σ_{yy} because σ_{xy} is constant versus electric field) increases from about zero to a saturated value as the electric field is increased. The threshold electric field for the nonlinear transport properties is not distinctly seen in either sample 1 or 2. In another sample (3), however, it was observed at about 0.5 mV/cm. This fact suggests that there is a sample-dependent threshold electric field and that it is undetectably small in most of the samples.

The amplitude of the small-period oscillations superposed on the magnetoresistance above 13 T has a large electric field dependence. When the oscillatory part $\Delta\rho_{osc}$ of the magnetoresistance was extracted from the background, it was found that $\Delta\rho_{osc}$ decreased as the electric field was increased. Although the oscillation looks similar to the Shubnikov–de Haas effect, it is considered to arise from a different origin.[15] Recently, a theory explaining the small-period oscillations in terms of the oscillation of tunneling probability was presented.[16] The observed electric field dependence of the small-period oscillations suggests the participation of some kind of tunneling mechanism.[17]

As a mechanism of the nonlinearity, an increase of the electron temperature can be excluded because of the following facts: The phase-transition field positions, which have a large temperature dependence,[7,8] showed no change for the different current values. The Hall resistance, whose amplitude depends on temperature,[4,5] was almost Ohmic and independent of the current value. The nonlinearity occurs only in the magnetoresistance above 7.5 T and not below. The electric field dependence of the small-period oscillation amplitude is opposite to its temperature dependence.[15] The Ohmic Hall resistance and abrupt appearance of nonlinearity above 7.5 T discount the possibility of a non-Ohmic contact effect.

The conductivity σ_{yy} ($\sim\rho_{xx}$) saturates in all the samples at $E=100$ mV/cm. This electric field is too small to satisfy the condition $eE\langle r\rangle \simeq \Delta$ for tunneling through the gap at the Fermi level,[18] if we assume $\Delta \simeq 1$ meV and the magnetic length $l=(\hbar c/eB)^{1/2}$ as the spatial extent of the wave function, $\langle r\rangle$. Therefore, the single-particle Zener tunneling of the SDW quasiparticle through the gap at the Fermi level is ruled out as the origin of the observed nonlinear transport. The Ohmic Hall conductivity σ_{xy} rules out the possibility of single-particle processes caused by the effective carrier number change, such as the impact ionization as in semiconductors. The nonlinearity seems to have a different origin from that observed in the Anderson-localized 2D electrons in the quantum Hall regime, because the I-V characteristics have quite a different shape in the quantum Hall case.[19]

An anomalous nonlinear transport has been reported in the normal state just above the superconducting critical current value at $B=0$ in $(TMTSF)_2ClO_4$.[20] In the present case, the resistance was completely Ohmic in the normal state below B_{th}, and the field of the nonlinear region is much higher than the superconducting critical field at $T=0$.

A similar nonlinear conduction, with no clear threshold field, has been reported for $(TMTSF)_2PF_6$,[21,22] and was considered to be due to an extrinsic origin, like microcracks in the sample.[22] In this case, the nonlinear transport was observed both below and above the SDW transition temperature (12 K),[21] in contrast to the case of $(TMTSF)_2ClO_4$. Moreover, in the present experiment, the nonlinearity disappeared at a higher temperature ($T=4.2$ K). Therefore, extrinsic origins are not likely in the present case.

The non-Ohmic transport phenomenon is reminiscent of those observed in quasi-1D compounds such as $NbSe_3$[23] or $K_{0.3}MoO_3$, whose nonlinearity is ascribed to the electric-field depinning of charge-density-wave (CDW) condensates. In such a case, the sliding mode of CDW carries a nonlinear current along the 1D direction, but no Hall current, so that the Hall conductivity does not show a nonlinearity.[24] This characteristic is common with the present observation.

In incommensurate SDW systems, there also exists a sliding mode which contributes to the conductivity in a similar manner as in CDW systems.[25] In contrast to CDW, however, the nonmagnetic impurities do not pin the sliding SDW mode in first order because of the uniform charge density in SDW.[26] Therefore, the pinning force of SDW is much weaker than in the CDW case. This is consistent with the present observation that the threshold field is very small if present. From all these considerations, it is very probable that the observed nonlinearity is caused by the sliding motion of the SDW.

Recently, nonlinear magnetotransport has been reported for other density-wave systems. Nonlinear conduction, with rather gradual onset, was observed in the high-magnetic-field phase of graphite.[18] One of the possible origins of this phase is considered to be a magnetic-field-induced CDW state which has a uniform charge density throughout the crystal as a result of the superposition of two CDW's with opposite phases.[27] In $NbSe_3$, it was reported that the threshold electric field for CDW motion is reduced at high magnetic fields and low temperatures.[28] It is not clear at this moment whether the observed nonlinearity in the present study is related to these phenomena or not.

In conclusion, we observed a non-Ohmic transport in the MFISDW state for the first time. This nonlinearity appeared only in the diagonal conductivity, and not in the Hall conductivity. The threshold electric field is very small. The sliding SDW may be a plausible mechanism of the observed nonlinear characteristics.

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