# Commensurability Effect of Magnetoresistance Anisotropy in the Quasi-One-Dimensional Conductor Tetramethyltetraselenafulvalenium Perchlorate, $(TMTSF)_2ClO_4$

T. Osada, A. Kawasumi, and S. Kagoshima
*Department of Pure and Applied Sciences, University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153, Japan*

N. Miura
*Institute for Solid State Physics, University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106, Japan*

G. Saito
*Department of Chemistry, Kyoto University, Oiwake-cho, Kitashirakawa, Sakyo-ku, Kyoto 606, Japan*
(Received 13 November 1990)

We have observed a clear series of fine structures superposed on the angular dependence of magnetoresistance in the metallic and magnetic-field-induced spin-density-wave phases in the low-dimensional organic superconductor $(TMTSF)_2ClO_4$. They appear at the field angles whose tangent satisfies a fractional condition reflecting the crystal geometry. This new oscillatory phenomenon is considered to result from a resonance effect, i.e., commensurate versus incommensurate, between two periodicities in the electron motion along an open orbit under tilted magnetic fields.

PACS numbers: 72.15.Gd, 71.25.Hc, 74.70.Kn, 75.30.Fv

The $(TMTSF)_2X$ family of organic conductors, where TMTSF denotes tetramethyltetraselenafulvalene and $X=ClO_4,PF_6,ReO_4$, have attracted a great deal of attention because of the gapless superconductivity and the magnetic-field-induced spin-density-wave (FISDW) transitions.[1] These compounds have very anisotropic electronic structures. The electronic spectrum is well described by a simple tight-binding model with the nearest-neighbor transfer integrals along the **a**, **b**, and **c** axes of $t_a:t_b:t_c=300:30:1$. They have a pair of sheetlike open Fermi surfaces (FS's) parallel to the $\mathbf{b}^*$-$\mathbf{c}^*$ plane (normal to the **a** axis) with a weak periodic warping due to $t_b$ and $t_c$. Since $t_b$ is too large to be ignored, the electronic system is usually regarded as an anisotropic two-dimensional (2D) one. The standard model for the FISDW is built on this picture which ignores the effect of finite $t_c$. In fact, the angular dependence of the threshold field for the FISDW phase obeys a cosine law expected from the 2D model.[2] On the other hand, the unusual anisotropy of the metallic-phase magnetoresistance (MR) suggests the three-dimensional (3D) nature of the system.[3,4]

When the effect of $t_c$, i.e., the 3D nature, is taken into account, a kind of resonance effect is expected for the variation of the field direction. It is a commensurability effect (commensurate versus incommensurate) between two periodicities in the electron motion along an open orbit on the FS warped by $t_b$ and $t_c$. The angular dependence of the MR or the FISDW instability could be affected by this effect.[5-7] Boebinger *et al.* reported the enhancement of the FISDW structures in the MR of $(TMTSF)_2ClO_4$ at the special field angles.[8] Naughton *et al.* observed several structures having no dependence on the field strength in the MR anisotropy in the metallic and FISDW phases.[9]

In order to find clear evidence for this commensurability effect, we investigated the details of the MR anisotropy of $(TMTSF)_2ClO_4$. In this paper, we report the first observation of a fractional series, expected from the commensurability effect, of fine structures in the MR anisotropy in the metallic and FISDW phases.

The MR measurements were carried out with a current of, typically, 100 $\mu$A along the **a** axis (1D conducting axis) in static magnetic fields up to 12 T. The typical size of the sample crystals was $4\times0.5\times0.3$ mm$^3$. The samples were sustained by four leads of gold (25 $\mu$m in diameter) bonded by gold paint, and mounted on a rotating stage. To achieve the "relaxed state" with sufficient ordering of the orientation of $ClO_4^-$ anions, the samples were cooled with speeds less than 50 mK/min near the anion ordering transition temperature (24 K).

For the magnetic fields rotated in the $\mathbf{a}$-$\mathbf{c}^*$ plane, the MR anisotropy in the metallic phase almost obeyed a cosine law, that is, the $\mathbf{c}^*$ component of the magnetic field is dominant for the MR. It takes a local minimum for the field along the **a** axis and a local maximum around the $\mathbf{c}^*$ direction.

On the other hand, when the magnetic fields were rotated in the $\mathbf{b}^*$-$\mathbf{c}^*$ plane, which is perpendicular to the conducting **a** axis, the MR showed a large deviation from the cosine law, as reported before.[3,4] Figure 1 shows the plots of the angular dependence of the **a**-axis resistance at 1.5 K for several fixed magnetic fields. The field direction in the $\mathbf{b}^*$-$\mathbf{c}^*$ plane is measured by the angle $\theta$ between the $\mathbf{c}^*$ axis and the field direction. At this temperature, the system undergoes a transition from the metallic phase to the FISDW phase with the steep increase of MR at the threshold field $B_{th}=6.1$ T under fields along the $\mathbf{c}^*$ axis ($\theta=0°$). In Fig. 1, the data in the metallic phase are plotted by the solid circles and

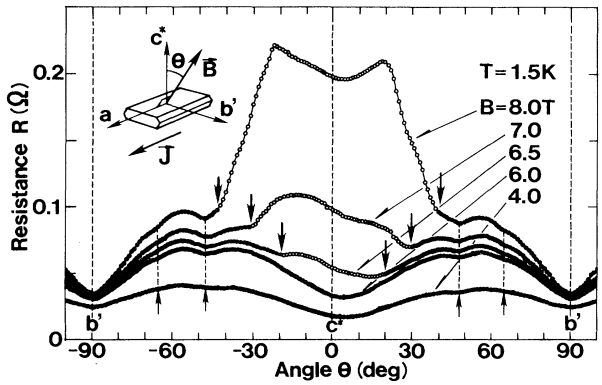


FIG. 1. Magnetoresistance anisotropy of $(TMTSF)_2ClO_4$ at several field values. The solid and open circles stand for the data in the metallic and FISDW phases, respectively. The downward and upward arrows indicate the FISDW threshold and the fine structures, respectively.

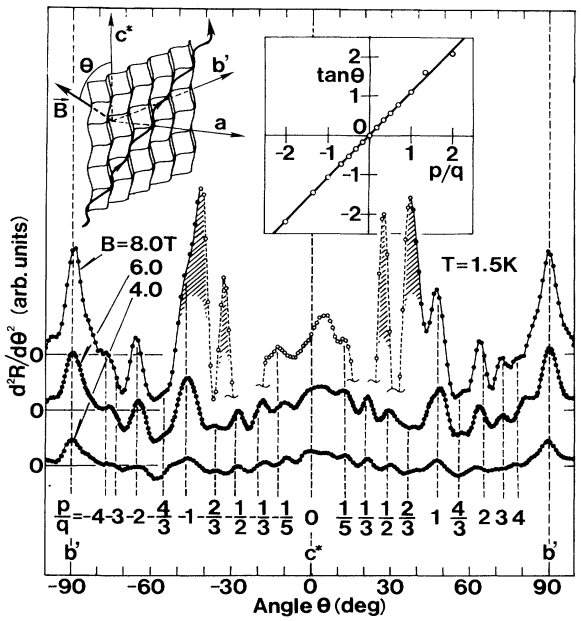


FIG. 2. Second-derivative traces of the anisotropy patterns at different field values. The data in the FISDW phase are plotted by the open circles. The hatched peaks correspond to the FISDW transitions. The angles which satisfy $\tan\theta = 1.1 \times p/q$ are shown. Right inset: Plot of the tangent of peak position vs the fraction $p/q$. Left inset: Semiclassical electron motion on the FS.

those in the FISDW phase by the open circles. The threshold angles which separate the metallic phase and the FISDW phase are indicated by large downward arrows in Fig. 1. Their positions satisfy the cosine law $B \times \cos\theta = B_{\rm th}(\theta = 0°) = 6.1$ T as expected from the 2D model.[2] As seen in the 4- and 6-T curves in Fig. 1, the resistance in the metallic phase shows an unusual angular dependence deviating from the cosine law: It takes local minima for the fields along the **b**′ ($\theta = \pm 90°$) and **c**′ ($\theta = 5°$) axes, where **b**′ and **c**′ are the projections of the **b** and **c** axes onto the **b***-**c*** plane. In addition, we find weak dip structures indicated by the small upward arrows around $\pm 48°$ and $\pm 65°$ in Fig. 1. The positions of these fine structures are independent of the strength of the magnetic field, in contrast to the threshold structure of the FISDW transition. The dips at $\pm 48°$ are considered to be the same ones observed by Boebinger *et al.* and Naughton *et al.*[8,9]

We concentrate our attention on these fine structures of the MR anisotropy. In addition to the structures marked by arrows, much weaker undulating structures are superposed on the curves in Fig. 1. By taking the second derivative of these curves, we can see these fine structures more clearly as a series of peaks. Figure 2 shows the second-derivative patterns for different fields. We checked that the smoothing procedure in the numerical differentiation did not affect the positions of these peaks. The hatched peaks correspond to the FISDW threshold and the transition between FISDW subphases. It is evident that the positions of the fine structures are independent of the field strength and that even in the FISDW phase the fine structures also appear at the same positions as in the metallic phase. These facts suggest that the fine structures originate neither from the FISDW transitions nor from the "fast oscillations" peculiar to $(TMTSF)_2X$,[10] but from some kind of topological effect related to the electronic structure. Experimentally, the fine structures appear when the fractional condition $\tan\theta = 1.1p/q$ is satisfied, where $p$ and $q$ are integers. Here, we set $p = q = 1$ for the largest peak at 48°. In Fig. 2, each peak is labeled by the fraction $p/q$. The right inset of Fig. 2 shows the tangent of the peak angles plotted against the assigned fraction $p/q$.

In order to examine the possible topological effect, we employ the following quasi-1D energy dispersion as a simplified band model for $(TMTSF)_2X$ in the metallic phase:

$$E(\mathbf{k}) = \hbar^2 k_x^2/2m^* - 2t_b \cos(bk_y) - 2t_c \cos(ck_z)\,, \qquad (1)$$

where we assume the system to be orthorhombic and choose the Cartesian coordinates $x$, $y$, and $z$ along the **a**, **b**, and **c** axes. The Fermi level $E_F$ is larger than $2(t_b + t_c)$, so that the system has a pair of open sheetlike FS's. They extend parallel to the **b***-**c*** plane with weak periodic warping due to $t_b$ and $t_c$ as shown in the left inset of Fig. 2. Under the magnetic field $\mathbf{B} = (0, B\sin\theta, B\cos\theta)$ applied with an angle $\theta$ tilted from the $z$ axis, the effective Hamiltonian is written as follows, using the Landau gauge $\mathbf{A} = (0, Bx\cos\theta, -Bx\sin\theta)$:

$$H = \hbar^2 k_x^2/2m^* - 2t_b \cos(G_b x + bk_y) - 2t_c \cos(G_c x - ck_z)\,. \qquad (2)$$

Here, $[x, k_x] = i$, $G_b = beB\cos\theta/\hbar$, and $G_c = ceB\sin\theta/\hbar$.

This Hamiltonian essentially describes a 1D problem under two periodic potentials whose periods are $2\pi/G_b$ and $2\pi/G_c$. Around $E_F$ the energy spectrum has an almost continuous 1D dispersion with respect to $k_x$. Semiclassically, the electron on the FS moves in **k** space along the edge of the FS cross section perpendicular to the field direction as shown in the inset of Fig. 2. In real space, the electron carries out a double-periodic motion along an open orbit, which is a winding linear trajectory along the 1D axis with modulations of two periods $2\pi/G_b$ and $2\pi/G_c$. Although the two periods are generally incommensurate, they become commensurate; that is, the electron motion becomes periodic at the special field directions where $G_b$ and $G_c$ take a rational ratio $G_b/G_c=q/p$ ($p$ and $q$ are integers). These special angles ("magic angles") satisfy the following fractional condition:

$$\tan\theta=(p/q)b/c\,. \tag{3}$$

This commensurability condition is fulfilled when the field is parallel to one of the lattice planes containing the 1D axis. It is dominated only by the geometry of the crystal structure. Some kind of resonance effect might be expected at these magic angles.[5-7]

We shall apply the above idea to $(TMTSF)_2ClO_4$. The real crystal is not orthorhombic but triclinic. In addition, the unit cell is doubled along the **b** axis and the FS's are folded at low temperatures below 24 K because of the anion ordering transition. Considering these two effects, the commensurability condition for $(TMTSF)_2ClO_4$ is rewritten as

$$\tan\theta=(p/q)2b'/c^*-\cot\alpha^*\,. \tag{4}$$

Here, $b'=b\sin\gamma$ is the interchain distance in the **a**-**b** plane, $c^*=c\sin\beta\sin\alpha^*$ is the interlayer distance between the **a**-**b** planes, and

$$\alpha^*=\cos^{-1}[(\cos\beta\cos\gamma-\cos\alpha)/\sin\beta\sin\gamma]$$

is the angle between the $\mathbf{b}^*$ and $\mathbf{c}^*$ axes. The factor 2 reflects the anion ordering period. Using the lattice parameters at 7 K ($a=7.068$ Å, $b=7.638$ Å, $c=13.123$ Å, $\alpha=84.50°$, $\beta=88.00°$, $\gamma=68.92°$),[11] the commensurability condition turns into $\tan\theta=1.09p/q+0.09$. Within the experimental accuracy, this is in very good agreement with the fractional condition $\tan\theta=1.1p/q$ for the observed fine structures. Therefore, we can conclude that the fine structures appear at the magic angles which satisfy the commensurability condition. The appearance itself of the fine structures gives evidence for the 3D nature of $(TMTSF)_2ClO_4$. The factor 2 in Eq. (4), which is introduced to label the largest dip at 48° as $p/q=1$, is not essential, since this factor should still be 1 when the anion ordering weakly affects the electronic structure. It would be interesting to study the relative amplitude of the fine structures for different cooling rates, that is, different strength of the anion ordering effect.

The next problem is the mechanism of why the MR shows dip structures at the magic angles. The well-known oscillatory phenomenon in MR anisotropy is observed in metals having a multiply connected FS. A similar phenomenon has been found in the quasi-2D organic metals $(BEDT\text{-}TTF)_2X$, where $(BEDT\text{-}TTF)_2$ denotes bis(ethylenedithio)-tetrathiafulvalene, having warped cylindrical FS.[12,13] These two phenomena can be understood in the framework of Boltzmann transport theory with a constant relaxation time.[14] The fine structures found in $(TMTSF)_2ClO_4$ are different from them because $(TMTSF)_2ClO_4$ has only sheetlike FS's. The MR calculation employing the Boltzmann equation in the relaxation-time approximation cannot reproduce the oscillatory MR anisotropy, at least in the weak warping limit ($E_F\gg t_b,t_c$). Essentially, most MR features of $(TMTSF)_2ClO_4$ in the metallic phase, in addition to the present one, cannot be explained by the standard picture: too large MR,[15] violation of Kohler's rule,[16] and anomalous anisotropy.[3,4]

Recently, Lebed and Bak have presented a theory to explain the anomalous MR anisotropy.[7] Beyond the relaxation-time approximation, they estimated the electron relaxation time assuming a 3D band model with finite $t_c$ and electron-electron scattering. They could reproduce the overall anisotropy in the observed MR. They also predicted the appearance of fine structures as a series of peaks at magic angles satisfying the commensurability condition. In spite of the crucial discrepancy that the fine structures are not observed as peaks but as dips in the MR anisotropy, their theory demonstrates the importance of the 3D effect and proposes the change of the relaxation time due to the commensurability effect as a possible mechanism for the fine structures.

It seems apparently strange that the fine structures appear even in the FISDW phase where the sheetlike FS's no longer exist because of FS nesting. The angular dependence of the FISDW instability due to the commensurability effect[5] is unlikely as an origin of the fine structures in the FISDW phase because of the cosine dependence of the threshold field.[2] One of the most plausible explanations is magnetic breakdown. Since the FISDW state is usually in the limit of strong magnetic breakdown (the cyclotron energy $\hbar\omega_c$ is larger than the SDW potential), an excited quasiparticle can move along an open orbit by tunneling across the SDW gap.[1] Therefore, the commensurability effect would work for the quasiparticle motion in the same way as in the metallic phase. Moreover, in the FISDW phase, the finite $t_c$ may open fine gaps within each Landau subband, which has an energy dispersion along the 1D axis due to the magnetic breakdown. The angular dependence of these fine gaps might cause the change of the 1D axis conduction.

In conclusion, we have observed fine structures in MR anisotropy of $(TMTSF)_2ClO_4$ as a fractional series of dips in both the metallic and FISDW phases. The field angles of the fine structures obtained experimentally satisfy a fractional condition. They coincide with the magic

angles expected from the commensurability condition that the two periods in an open orbit become commensurate. This phenomenon is a new type of magnetic oscillation reflecting only the crystal geometry. The appearance of this effect in $(TMTSF)_2ClO_4$ suggests the importance of the 3D nature of this compound. The mechanism of the resistance change at the magic angles is still unclear, since the existing theory predicts peaks at the magic angles instead of dips.

The authors are grateful to Dr. M. J. Naughton for kindly sending a preprint. They thank Dr. M. Tokumoto, Dr. N. Kinoshita, and Professor H. Anzai for kindly supplying samples for comparison. This work is partly supported by a Grant-in-Aid for Scientific Research from the Japanese Ministry of Education, Science and Culture.

[1]For a brief review, see, for example, P. M. Chaikin, J. S. Brooks, S. T. Hannahs, W. Kang, G. Montambaux, and L. Y. Chiang, in *The Physics and Chemistry of Organic Superconductors,* edited by G. Saito and S. Kagoshima (Springer-Verlag, Berlin, 1990), p. 81.

[2]X. Yan, M. J. Naughton, O. S. Cheema, R. V. Chamberlin, S. Y. Hsu, L. Y. Chiang, and P. M. Chaikin, Solid State Commun. **66**, 905 (1988).

[3]R. Brusetti, K. Bechgaard, G. G. Lonzarich, and R. H. Friend, J. Phys. (Paris), Colloq. **44**, C3-1055 (1983).

[4]K. Murata, H. Bando, K. Kajimura, T. Ishiguro, H. Anzai, S. Kagoshima, and G. Saito, Mol. Cryst. Liq. Cryst. **119**, 131 (1985).

[5]A. G. Lebed, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 137 (1986) [JETP Lett. **43**, 174 (1986)].

[6]G. Montambaux and P. B. Littlewood, Phys. Rev. Lett. **62**, 953 (1989).

[7]A. G. Lebed and P. Bak, Phys. Rev. Lett. **63**, 1315 (1989).

[8]G. S. Boebinger, G. Montambaux, M. L. Kaplan, R. C. Haddon, S. V. Chichester, and L. Y. Chiang, Phys. Rev. Lett. **64**, 591 (1990).

[9]M. J. Naughton, O. H. Chung, L. Y. Chiang, and J. S. Brooks, Mater. Res. Soc. Symp. Proc. **173**, 257 (1990).

[10]X. Yan, M. J. Naughton, R. V. Chamberlin, S. Y. Hsu, L. Y. Chiang, J. S. Brooks, and P. M. Chaikin, Phys. Rev. B **36**, 1799 (1987).

[11]B. Gallois, D. Chasseau, J. Gaultier, C. Hauw, A. Filhol, and K. Bechgaard, J. Phys. (Paris), Colloq. **44**, C3-1071 (1983).

[12]M. V. Kartsovnik, P. A. Kononovich, V. N. Laukhin, and I. F. Shchegolev, Pis'ma Zh. Eksp. Teor. Fiz. **48**, 498 (1988) [JETP Lett. **48**, 541 (1988)].

[13]K. Kajita, Y. Nishio, T. Takahashi, W. Sasaki, R. Kato, H. Kobayashi, and Y. Iye, Solid State Commun. **70**, 1189 (1989).

[14]R. Yagi, Y. Iye, T. Osada, and S. Kagoshima, J. Phys. Soc. Jpn. **59**, 3069 (1990).

[15]T. Ishiguro, K. Kajimura, H. Bando, K. Murata, and H. Anzai, Mol. Cryst. Liq. Cryst. **119**, 19 (1985).

[16]L. Forro, K. Biljakovic, J. R. Cooper, and K. Bechgaard, Phys. Rev. B **29**, 2839 (1984).