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Takehiko Ishiguro [a], Koji Kajimura [a], Hiroshi Bando [a], Keizo Murata [a] & Hiroyuki Anzai [a]
[a] Electrotechnical Laboratory, Sakuramura, Ibaraki, 305, Japan
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ELECTRONIC STATE OF $(TMTSF)_2ClO_4$ IN METALLIC REGION

TAKEHIKO ISHIGURO, KOJI KAJIMURA, HIROSHI BANDO,
KEIZO MURATA and HIROYUKI ANZAI
Electrotechnical Laboratory, Sakuramura, Ibaraki 305, Japan

Abstract The transverse magnetoresistances along both the a (ρ_{aa}) and the b (ρ_{bb}) directions are compared with the calculated based on the tight binding band model. The magnetic field dependence of ρ_{bb} can be well explained with the model, whereas ρ_{aa} cannot be. This suggests that some mechanism other than the single particle transport is dominating. Possibilities of the contribution from the fluctuating superconductivity, the magnetic breakthrough, and the SDW are discussed.

INTRODUCTION

The organic synthetic metal $(TMTSF)_2ClO_4$ has attracted great attention due to unusual characteristics ascribable to novel phases such as the superconductivity and the SDW phases.[1] The efforts to understand the material have succeeded in clarifying the principal features on the whole, and the comprehensive picture drawn for this material will be a useful guide to investigate the other organic synthetic metals, which may bring about unknown nature hereafter.

However, it is true also that we still meet not a few unsolved problems with this material, if we look at it in detail. We throw here a light on the metallic behavior of $(TMTSF)_2ClO_4$ at low temperatures. Jerome has pointed out the unusual features, such as the continual decrease in resistivity down to 1 K without satulation and the large magnetoresistance in contrast to the ordinary metals, and has insisted on the presence of the fluctuating superconductivity specific to the low dimensionality even in

the metallic regime.[2]

In this paper, we present and discuss our recent results on the metallic region, with respect to two experiments: (i) The transverse magnetoresistance at low temperatures in the intermediate field region and (ii) the tunnel junction spectroscopy of the density of states near the Fermi surface. We investigate the magnetoresistance in the intermediate field, since it is known that the field higher than 5 T may induce the SDW phase[3,4] and hinder the ground electronic state dominating in the low field region.

TRANSVERSE MAGNETORESISTANCE

A possible Fermi surface of $(TMTSF)_2ClO_4$ is derived by the two dimensional tight binding band model,[5,6] as follows,

$$\varepsilon(\underline{k}) = -2t_a \cos \underline{a}\,\underline{k} - 2t_b \cos(\underline{b}\,\underline{k} - \theta \text{ sign } k_a) \qquad (1)$$

where $\varepsilon(\underline{k})$ is the energy of the electron, $\underline{k}$ is the pseudomomentum of the electron, t_i is the electron transfer energy along the i-th direction (i = a,b), θ is a phase factor.[5]

The effect of a magnetic field on an electron state is to give the change in the electron wavevector normal to both the direction of the magnetic field $\underline{H}$ and the electron velocity $\underline{v}$ which is itself normal to the energy surface. Therefore, the electron motion is confined to the orbit defined by the intersection of the Fermi surface with a normal plane to $\underline{H}$. For the Fermi surface described by eq. (1), the induced electron movement by $\underline{H}$ is of open orbit, in any case. However, different situations are realized with respect to the current direction; whether it is in the direction along the open orbit or not.

We have carried out the numerical calculation of the conductivity tensor component $\sigma_{\xi\eta}$ in the presence of $\underline{H}$ by using the following formula, which is derived by applying the time

relaxation approximation for the Boltzmann equation,

$$\sigma_{\xi\eta} = -\frac{e^2}{4\pi^3}\,\tau \int d^3k\,\frac{\partial f_0}{\partial \varepsilon}\,v_\eta(0)\int_0^\infty dt\; v_\xi(t)e^{-t/\tau}/\tau \qquad (2)$$

with $\xi, \eta = a, b$ and $v_\xi(t)$ is the electron velocity in the presence of $\underline{H}$,

$$\frac{dk_\xi}{dt} = -\frac{e}{\hbar c}\,(\underline{v}\times\underline{H})_\xi \qquad (3)$$

$$v_\xi(\underline{k}(t)) = \frac{1}{\hbar}\,\frac{\partial\varepsilon(\underline{k}(t))}{\partial k_\xi} \qquad (4)$$

where τ is the minimum relaxation time of the system, f_0 is the Fermi-Dirac function.

By assuming that the unit cell is approximated by an orthorhombic lattice by putting $\alpha = \beta = \gamma = 90°$ with $a=3.63$ Å (we assume that the dimerization is negligible), $b = 7.68$ Å and by using the following values,[8] $t_a = 0.29$ eV, $t_b = 0.024$ eV and $\tau = 7.3\times10^{-12}$ sec, we have calculated the magnetic field dependences of the conductivity and resistivity tensor components. The results are shown in Fig. 1a. The typical features of the calculated are summerized as follows: ρ_{aa} is independent of the magentic field. On the other hand $\rho_{bb}(H)-\rho_{bb}(0)$ is almost proportional to H^2.

In Fig. 2, we show the observed magnetic field dependences of ρ_{aa} and ρ_{bb} at 0.5 K under the magnetic field $\underline{H}//c^*$ for slowly cooled samples to realize the relaxed state in which the anions are ordered. It is evident that $\rho_{aa}(H)-\rho_{aa}(0)$ varies almost with H^2 as well as $\rho_{bb}(H)-\rho_{bb}(0)$, although the magnitude is smaller than for the latter. Note that the undulation in ρ_{bb} appearing above 4.2 tesla is ascribed to the onset of the Shubnikov-de Haas like oscillation and hence an indication of the appearance of the field induced state. In Fig. 2, we fit the calculated ρ_{bb} to the observed with $\tau = 2.3\times10^{-12}$ sec. The calculated ρ_{aa} for the same τ

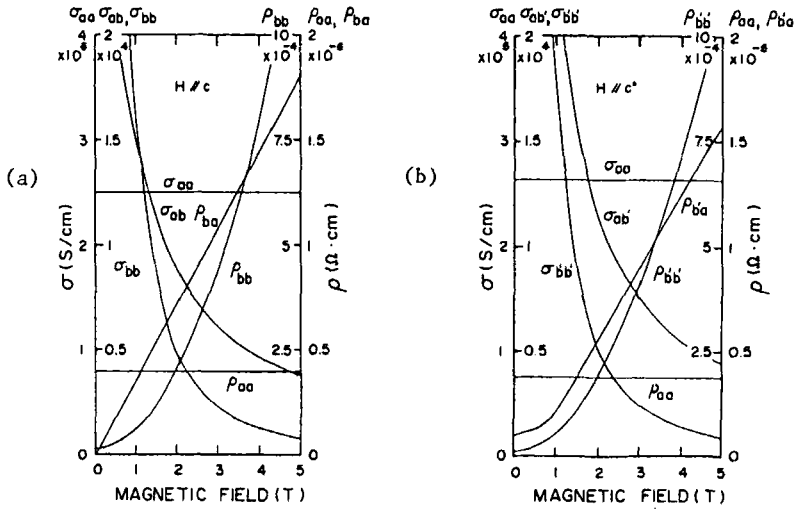

FIGURE 1 Calculated magnetic field dependence of the conductivity and resistivity tensor components in an orthorhombic structure (a) and a triclinic structure (b).

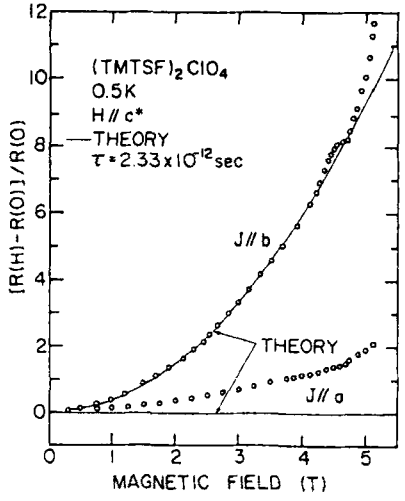

FIGURE 2 Comparison of the observed magnetic field dependence of ρ_{aa} (J//a J: current) and ρ_{bb} (J//b) with the calculated.

is exhibited in the same figure, which shows that the effect of the magnetic field up to 5 T is less than 0.1% of $\rho_{aa}(0)$.

From the test whether the magnetoresistance obeys the Koehler's rule, Cooper et al. have claimed that the a axis is not exactly a principal axis of the conductivity tensor.[9] In fact the TMTSF molecules are not on the (100) plane but almost on the (210) plane and the direction of the strongest interaction may deviate from the a axis. However, by taking account of the fact that the overall stacking direction is in the a axis, it is reasonable to consider that electron motion is coherent along the a axis and hence the a axis is the principal direction.

It is worth-while to inspect the effect of the tricinic nature of the lattice. To do so, we have carried out the numerical calculation by taking account of the inclination angles[10] $\alpha = 84.6°$, $\beta = 86.7°$, $\gamma = 70.4°$ and with $\theta = -33°$.[5] The calculated results are shown in Fig.1b, which have quite similar features as Fig. 1a. Note that although the off-diagonal component $\rho_{b'a}$ is not zero at H=0 due to the triclinic structure, we find that ρ_{aa} is independent of H.

We should remind the anomalous angular dependence of ρ_{aa} for the transverse magnetic field.[11] The angular dependence is anomalous with the following respects; (1) the angular dependence exhibits a local minimum for H//c' (Fig. 2 of ref. 11) and (2) the magnetic field dependence is very weak for H//b' but ρ_{aa} at the superconducting critical field H_{c2} is already larger than that for the other direction (Fig. 1 of ref. 11). The temperature and the magnetic field dependences of the local minimum for H//c' suggest contribution of some field-induced state.[3,4]

Then, a question should arize; what mechanism is dominating the ρ_{aa}? For this we can remind of three factors: The first one is the presence of the fluctuating low dimensional superconductivity as insisted by Jerome et al.[2] The second is the effect of the magnetic breakthrough as proposed by Cooper et al.[9]: The

breakthrough may occur accross the superlattice gap which is caused by the anion ordering; the gap is expected to be small due to weak potential modulation associated with the anion ordering. The third is the effect of the precursor of the SDW phase condensation.

In the following we discuss the possibility of the fluctuating superconductivity just above the superconductivity transition temperature T_c. This has been the controversial point since the discovery of the large magnetoresistance in the TMTSF salt.[1]

TUNNEL JUNCTION SPECTROSCOPY

The most striking key to raise the fluctuating superconductivity is, after all, the tunnel junction experiment performed by Fournel et al. with GaSb Schottky diode structure.[12] What are noted are: (1) They did not observe the three dimensional superconducting gap corresponding to T_c of 1.2 K and, instead, (2) they observed large energy gap 2Δ of 8 meV corresponding to T_c of 26 K.

In the light of the importance of these experimental results, we have investigated the tunneling junction characteristics using a different structure with $(TMTSF)_2ClO_4$/a-Si/Pb.[13] (Fig. 3) This can give a superconductor/insulator/superconductor (S/I/S) configuration, which can give the density of states for the superconducting state most clearly. Further we can inspect the density of states of $(TMTSF)_2ClO_4$ above T_c, which may be directly concerned with the electronic state above T_c.

From our experimental results we have reduced that (1) The 2Δ of 0.34 meV corresponding to T_c of 1.2 K is observed under (S/I/S) configuration, (2) whereas the 2Δ corresponding to that reported by Fournel et al. are not observed. Under the (N/I/N) (N; normal state) configuration which is realized above 7.2 K, we can see a slight decrease in dI/dV centering at zero bias region, but this cannot be directly related to the presence of the pseudo gap of

the fluctuating superconductivity.[13] Thus we cannot recognize the fluctuating superconductivity above T_c and hence in the metallic region.

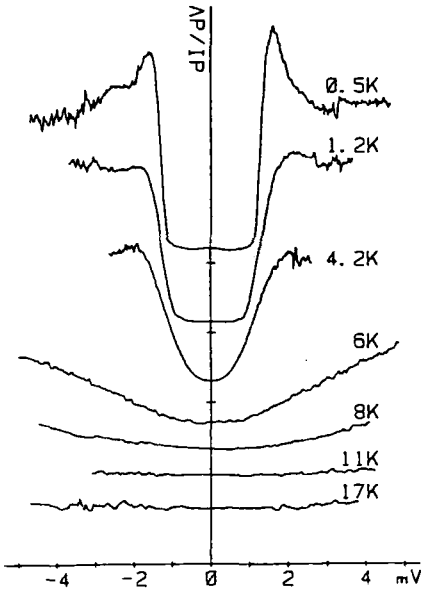

FIGURE 3 Differential conductance of $(TMTSF)_2ClO_4$/a-Si/Pb tunnel junction at various temperatures.

DISCUSSION

The reason for the absence of the magnetic field effect in the calculated ρ_{aa} is that ρ_{aa} is determined principally by the velocity normal to the quasi one-dimensional Fermi surface which is dominated by the dispersion of the band structure.

The magnetic breakthrough across the gap formed by the anion ordering with (0,1/2,0) does not bring about a change in direction of the velocity to form closed orbits and hence $\rho_{aa}(H)$ is little affected, even if it occurs. Furthermore, if the magnetic breakthrough works, some threshold is expected in $\rho_{aa}(H)$ contrary to the observed which shows smooth H-dependence down to low field.

The only way to explain the $\rho_{aa}(H)$ within the present context is to introduce the effect of nesting with which the direction of the electron motion induced by the magnetic field is changed to

form closed orbits. The nesting is permitted at limited parts[5] of the Fermi surface with a limited probability since it is not condensed. This is the effect of the fluctuating SDW. However, the associated anomaly in the angular dependence of $\rho_{aa}(H)$ seems not to be explained satisfactorily by the plain contibution of the SDW.[13]

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