ANOMALOUS MAGNETORESISTANCE IN ORGANIC CONDUCTORS
AND THE DIMENSIONALITY OF THE ELECTRON SYSTEM

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ABSTRACT

Anomalous magnetotransport phenomena observed in \( \theta \)-(BEDT-TTF)\(_2\)I\(_3\) and \( \alpha \)-(BEDT-TTF)\(_2\)I\(_3\) are discussed in connection with the dimensionality of the electron systems. An oscillatory magnetoresistance in \( \theta \)-(BEDT-TTF)\(_2\)I\(_3\), which oscillate as a function of the magnetic field direction is understood in terms of the effect arising from the quasi-two-dimensional nature of the electron system. On the other hand, the magnetic field on an \( \alpha \)-(BEDT-TTF)\(_2\)I\(_3\) crystal, of which the Peierls transition is suppressed by applying the pressure, seems to make the one dimensional nature of the system stronger.

1 INTRODUCTION

\( \text{(BEDT-TTF)}_2\text{I}_3 \) (hereafter, we abbreviate BEDT-TTF molecule as ET) is known to crystallize into several different crystals. This family of materials has been attracting attentions because some of them are superconductors.

In this paper, the magnetotransport phenomena in \( \theta \), and \( \alpha \)-type crystals of ET\(_2\)I\(_3\) are discussed. We would like to show that the dimensionality of the electron system plays an essential role in determining the transport phenomena in the magnetic field. Such a characteristic feature of the material in the normal state is not only intersting from the physical point of view, but also gives important informations to understand the superconducting state.

These two crystals have layered structures, layers of ET molecules and of I\(_3\) molecules piled up alternatively. The electric transport is supported by holes on ET molecules. Although the arrangement of ET molecules in these materials is similar to each other, the small difference in the arrangement of molecules gives rise to the qualitative difference in the energy spectrum. A \( \theta \)-type crystal has a two dimensional energy spectrum, while an \( \alpha \)-type crystal is one dimensional.
Recently, we have found a new type of oscillatory magnetoresistance phenomena in $\theta$-ET$_2$I$_3$[1], as will be discussed in section 2. A current interpretation of the phenomena is based on the two dimensional electron energy spectrum with slight three dimensionality[2,3]. This is the first theme of this paper.

The $\alpha$-type crystals are of one dimension and show the Peierls instability becoming insulating in the low temperature phase. Suppressing the instability by applying hydrostatic pressure, we have performed the magnetoresistance measurements. Fairly a large magnetoresistance is observed in this crystal, which was found to be qualitatively different from that in $\theta$-type crystals. However, it seems that the dimensionality of the system is also important here. The phenomena is discussed in section 3.

2 $\theta$-ET$_2$I$_3$

A $\theta$-type crystal of ET$_2$I$_3$ has two dimensional electron energy spectrum. It exhibits metallic conduction down to low temperatures and undergoes the superconducting transition at about 3.6K[4].

Recently, we have found a new type of oscillatory magnetoresistance phenomena in this crystal[1]. As is shown in Fig.1, the magnetoresistance oscillates as a function of the magnetic field direction (not as a function of the magnetic field strength). For the magnetic field in the $c^*$-crystal plane, the angles corresponding to the maximums and the minimums of the oscillation are given as

$$\tan(\Theta_{\text{max}})=0.37N-0.15$$
$$\tan(\Theta_{\text{min}})=0.36N+0.053 \ (N=1,2,3,\ldots) \ (1).$$

The amplitude of the oscillation, on the other hand, is found to depend on the temperature through the zero field resistivity $\rho_0$ as $A=A(\rho_0(T))$. This implies that the origin of the oscillation should be hunt up in the orbital nature of the carriers.

Yamaji[2] has pointed out the importance of the dimensionality of the electron system. He has shown that for a two-dimensional electron system with a slight three dimensionality, of which the energy spectrum is given as

$$\varepsilon=\hbar^2/2m(k_x^2 + k_y^2)-2t\cos(ck_z) \ (2),$$

Fig.1 Magnetoresistance $M$ is plotted against the angle between the magnetic field direction and $c^*$-crystal axis. Magnetic field strength are 6.5T, 6T, 5.5T, 5T, 4.5T, 4T, 3.5T, 2.5T, 2T, 1T.
a magnetic field in the direction which satisfies
\[ \tan(\Theta) = \pi/c_{kF}(N-0.25) \quad (N=1,2,3,\ldots) \quad (3), \]
enhances the two dimensionality of the system.

Yagi[3] has extended the idea of Yamaji and has calculated the resistivity tensor by the Boltzmann equation treatment. He has shown that the magnetic field in the direction which satisfies eq.(3), gives the peak to the resistivity \( \rho_{zz} \).

These two theories seem to give good understanding of the phenomena.

3 \( \alpha \)-ET\(_2\)I\(_3\)

A crystal of \( \alpha \)-type ET\(_2\)I\(_3\) has one dimensional electron energy spectrum. It undergoes the Peierls transition at about 135K, becoming insulating below this temperature.

![Fig.2](image1.png)

**Fig.2** Temperature dependences of the resistivity \( \rho \) under several hydrostatic pressures.

![Fig.3](image2.png)

**Fig.3** Magnetic field dependences of the resistivity, for \( H \) normal to the conducting plane and parallel to it.

In order to perform the magnetoresistance measurement at low temperatures, we have to suppress the metal-insulator transition. It can be achieved by applying the hydrostatic pressure on the crystal. Similarly to other organic conductors, the pressure on the crystal enhances the electron transfer between neighboring ET molecules. The direction of the effect is that the pressure brings the dimensionality of the electron system from one dimensional side to two dimensional side. As the results, the Peierls transition tends to be suppressed by the pressure.

Figure 2 shows how the pressure affects the transition. The pressure moves the transition towards low temperatures, and on the same time, makes the low temperature phase "semimetallic"; Some part of the Fermi surface survives after the transition. Under the pressure above about 12kbar, the insulating phase disappears and the crystal is metallic over the whole temperature region. We have performed the magnetoresistance measurement on such a pressurized crystals.

As is shown in Fig.3, the magnetic field parallel to the conducting plane affects the resistivity only very weakly. While, in the normal magnetic field, the resistivity increases linearly with increasing magnetic field up to about 0.3T, above which the resistivity tends to saturate. The effect of the magnetic field is fairly large [5]. At 1.4T, the magnetoresistance is about 2.
Fig. 4 Temperature dependences of the resistivity for $H=0$ and $H=1.2T$.

It can be shown that the present phenomena are determined by the magnetic field normal to the conducting plane[5]. The magnetic field parallel to the plane affects the resistivity only very weakly, as is seen in Fig.3.

The most astonishing feature of the phenomena is shown in Fig.4. In this figure, the temperature dependence of the resistivity, one in the absence of the magnetic field, and one in $H(=1.2T)$ normal to the conducting plane are presented. One can see that the influence of the magnetic field remains in the temperature up to 50K. This is important because the magnetic field energy is only about 1K (for $H=1.2T$) which is very low compared with 50K. What is the origin of the effect.

Inspecting the temperature dependence of the zero field resistivity, carefully, we find that $\rho-T$ curve has a singularity at 50K. Above 50K, the resistivity is a decreasing function of the temperature; i.e., the resistivity increases with decreasing temperature. The $\rho-T$ curve changes the derivative at 50K. Below this temperature, the resistivity decreases with decreasing temperature. It seems that some mechanism comes up at 50K which suppresses the increase of the resistivity.

In the magnetic field, on the other hand, the singularity at 50K disappears. The resistivity increases continuously with decreasing temperature in this region.

From these observation, we interpret the phenomena as follows. Under the hydrostatic pressure, the metallic state becomes stable. However, it is in the critical region between one dimension and two dimension, and a slight perturbation can cause the Peierls instability to appear. The magnetic field works to make the electron system more one dimensional, which gives rise to the increase of the resistivity due to the Peierls transition.

REFERENCES
3. Yagi et al., in this proceedings.
5 K.Kajita et al., to be published.