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Transport property of the organic conductor α-(BEDT-STF)$_2$I$_3$ in the magnetic field

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Abstract. One of the recent progresses in the field of investigation of organic conductors, is the discovery of a zero-gap conductor α-(BEDT-TTF)$_2$I$_3$. Under high pressure, this material exhibits mysterious transport behavior. For example, the resistance in the absence of magnetic field is nearly constant in a wide region of temperature. Now, we are in a stage where we can understand the behavior of α-(BEDT-TTF)$_2$I$_3$ based on the knowledge of zero-gap conductors. Quite recently, we found that, as out-of-plane magnetic field is applied, the Hall resistance, $\rho_{xy}$ increases linearly with $B$ and is saturated at a value $\hbar/e^2$/number of layers. The saturation occurs in the field where zero-mode Landau carriers dominate the transport. We believe this is one of the key phenomena to characterize the transport of zero-gap conductors. In this work, we intend to make it clear if this characteristic behavior of $\rho_{xy}$ is universal to zero-gap conductors. α-(BEDT-STF)$_2$I$_3$ has a crystal structure similar to that of α-(BEDT-TTF)$_2$I$_3$. The electron energy structures resemble each other. A serious difference, however, exists between these two materials. α-(BEDT-STF)$_2$I$_3$ contains "intrinsic" lattice imperfections due to unsymmetrical molecular structure of BEDT-STF so that carriers receive strong scattering. The purpose of this work is to answer the question whether the carriers in dirty zero-gap conductors shows transport phenomena similar to α-(BEDT-STF)$_2$I$_3$. The answer is yes.

1. Introduction

α-(BEDT-STF)$_2$I$_3$ is a two dimensional molecular conductor. A crystal consists of conductive layers and insulating layers piled up alternatingly. This material essentially is a semiconductor. A unit cell contains four BEDT-STF molecules and a molecule has electrical charge of +0.5e. Therefore, the electrons have four energy bands, of which the lower three bands are occupied and the top band is empty.

Since a conductive layer is sandwiched between two insulating layers, the electron transport occurs almost independently in each conductive layer.

This material exhibits a metallic conduction at high temperatures above 100K. In this region, however, the conductivity is nearly constant indicating that this metal is not a "normal" metal. Below 100K, the system changes to an insulator.

There are two other materials that have the transport properties similar to this material. They are α-(BEDT-TTF)$_2$I$_3$ [1], and α-(BEDT-TSF)$_2$I$_3$. These three materials have similar crystal
Figure 1. Crystal structure of $\alpha$-$\text{I}_3$ salts.

structures and hence, the electron energy structures also resemble each other. The characteristic features of transport phenomena are common to them. They have metallic conductivities at high temperatures and change to insulators at low temperatures [2].

As is shown in Figure 3, this metal-nonmetal transition is suppressed by applying hydrostatic pressure. At high pressures, the high temperature metallic phase expands to low temperatures.

Recently, $\alpha$-(BEDT-TTF)$_2$I$_3$ was found to be a zero-gap conductor with the Fermi energy located just on the contact points of two energy bands [3][4]. In the vicinity of the contact point, the energy band has linear dispersion. (A schematic figure of the energy band is given in Figure 4.) Experimental results [5] accumulated hitherto were reanalyzed based on the new scheme. For example, the conductivity independent of temperature is a characteristic of such a zero-gap conductor. The absolute value of the conductivity is predicted by theories to be $\frac{e^2}{h}$ per conductive layer. It does not depend on temperature nor material. This is just what we observed in the experiments. Therefore, we may expect that those three materials under high pressures are zero-gap conductors. In this work, we examine the Hall effect. We observed a peculiar Hall effect in $\alpha$-(BEDT-TTF)$_2$I$_3$ that, we believe, is ascribed to the zero-gap electron energy structure. The purpose of this work is to find whether the same phenomena occur in $\alpha$-(BEDT-STF)$_2$I$_3$. This material is interesting as it is expected to be intrinsically dirty because of unsymmetrical molecular structure of BEDT-STF. It is an important difference of $\alpha$-(BEDT-STF)$_2$I$_3$ from other two materials. We are interested in how the transport phenomena in a zero-gap conductor are affected by the imperfections of the crystal.
2. Experimental results and discussions

We measured resistance ($\rho_{xx}$: resistivity parallel to the current, $\rho_{xy}$: Hall resistivity) of $\alpha$-(BEDT-STF)$_2$I$_3$. A conventional dc method with the current in the 2D plane was used. The sample was placed in a pressure bomb made of BeCu. Figure 3 is the resistivity in the absence of magnetic field. Above 100K, the sample exhibits metallic transport with the resistivity that is almost independent of temperature. In this region, three curves for different pressures coincide. Below 100K, on the other hand, the resistivity depends on pressure. Under the pressure of 1.8GPa, the growth of resistance at low temperatures is almost suppressed. It is noted, however, that a remnant of the resistivity rise is not small enough, so that it can not be neglected in the analysis.

In Figure 5, magnetic field dependences of $\rho_{xx}$ and $\rho_{xy}$ are plotted. The field dependence of $\rho_{xy}$ consists of two field regions. At low fields below 2T, $\rho_{xy}$ grows linearly with $B$. Above 2T, it is saturated at a value that is independent of magnetic fields. The saturated value of $\rho_{xy}$ is about $1.3 \times 10^{-2} \Omega\text{cm}$. This is about the twice of $\frac{\hbar}{e^2} c_z$ ($c_z$ is the lattice constant along the z-direction). These features of $\rho_{xy}$ are the same as those observed in $\alpha$-(BEDT-TTF)$_2$I$_3$. The difference exists only in the critical magnetic field where the saturation occurs. In the present system, saturation occurs in the field above 2T. On the other hand, the critical field is about 0.3T in $\alpha$-(BEDT-TTF)$_2$I$_3$. We claim that the same mechanism works on the transport phenomena of $\alpha$-(BEDT-TSF)$_2$I$_3$ and $\alpha$-(BEDT-TTF)$_2$I$_3$. The effect of strong scattering potential that the carriers in $\alpha$-(BEDT-STF)$_2$I$_3$ must feel, could not be detected in the experiments.

In the following, we mention about the picture we have on this phenomenon.

It is based on the zero mode Landau level that is located at the contact points. Since the Fermi energy of this system is also on the contact points, the value of Fermi distribution function of zero mode Landau electrons is 1/2 irrespective of temperature and magnetic field strength. Therefore, the carrier system is never degenerated, and we can treat it as a classical system. In order to interpret the experimental result, we make some assumptions. We consider a carrier in the zero mode Landau level as a combined particle of a hole and an electron. This combined
particle is assumed to be fragile, however, and the electron and the hole move independently. We should remind that if the hole band and the electron band is strictly symmetrical, we cannot observe the Hall effect at all. Actually, a large Hall effect with hole polarity is observed as shown in Figure 5. Therefore, we expect that some break of the symmetry exists in the system that gives rise to the unbalance between electrons and holes. If as a result, the hole mobility becomes a few times as large as the electron mobility, we can treat the system as a single carrier system.

Revisiting Figure 5, we discuss how to explain it. In the low field region, \( \rho_{xy} \) is proportional to \( B \). This is a typical behavior of \( \rho_{xy} \) in normal metals. Many conductors show the Hall effect of this type. Usually, the slope of the curve of \( \rho_{xy} \) is called as Hall coefficient and we calculate the carrier density from it. For a region of low magnetic field in the present system, we can adopt this picture. If we apply this simple picture to the higher field region where the saturation of \( \rho_{xy} \) occurs, we can calculate the magnetic field dependence of the carrier density. In this model, the magnetic field dependence of \( \rho_{xy} \) is ascribed to the change in the carrier density. Standing on a single carrier picture, we can estimate the carrier density as a function of magnetic field. The results are given in Figure 6. For \( B=0 \), the density is about \( 3.8 \times 10^{16} / \text{cm}^3 \). It increases with increasing \( B \) to a value, \( 4.2 \times 10^{17} / \text{cm}^3 \), at \( B=7 \text{T} \). The density at \( 7 \text{T} \) is close to the calculated density of zero mode Landau electrons.

Figure 5. Magnetic field dependence of \( \rho_{xx} \) and \( \rho_{xy} \) under hydrostatic pressure. The green line the value of the resistivity predicted by theories to be \( c_2 \frac{h}{e^2} \).

Figure 6. Magnetic field dependence of carrier density, \( n \), under hydrostatic pressure. The red line give the density of the zero-mode Landau carriers that is written as \( \frac{eB}{4\pi \hbar c_2} \).

In zero gap conductors, the carrier density \( n(T, 0) \) at zero field is given as \( n(T, 0) = aT^2 \) [5], where \( a \) is a parameter determined by the energy band structure. At a finite magnetic fields, on the other hand, there appear zero mode Landau carriers. The density of zero mode carriers are given as, \( n(0, B) = N_{LL} bB \), where \( N_{LL} \) is the Landau degeneracy. Since \( N_{LL} \) do not depend on temperature, and \( n(T, 0) = aT^2 \), the density of Landau carriers exceeds that of thermally excited carriers at low temperatures. Therefore, at low temperatures or at high magnetic fields, Landau carriers give the dominant contribution on the transport of this system. Among the Landau levels, zero mode carriers are important at high fields where the energy gap between Landau levels with the index 0 and 1 exceed the thermal energy, because in such a situation, most carriers are in the zero mode Landau level. It is often called as the quantum limit. In the quantum limit, the carrier density is given as the Landau degeneracy. It is depicted in Figure 6.

In the last part of this paper, we discuss the Zeeman effect on the transport in zero gap international symposium on molecular conductors (ismc 2008) iop publishing journal of physics: conference series 132 (2008) 012001 doi:10.1088/1742-6596/132/1/012001
Figure 7. The Hall resistivity is plotted as a function of $B \sin \theta$. Magnetic field will a fixed strength was rotated in the plane normal to the current.

In the magnetic field, an electron energy level is split into two levels due to the Zeeman effect. It works to reduce the density of zero-mode carriers by shifting the level from the Fermi energy. In the magnetic field of 1T, the Zeeman energy is about 1K. So, the Zeeman effect may give some influence on the transport at low temperatures around 1K. To examine the Zeeman effect, we measured the Hall resistivity at several magnetic fields. The field is rotated in the plane normal to the current direction. The results are shown in Figure 7. In this figure, $\rho_{xy}$ is plotted against the magnetic field component normal to the conductive layers. All the data are on a single curve. It tells that the Zeeman effect is not strong. Since the Zeeman effect is expected to be nearly isotropic, it gives rise to the resistance change that is independent of the field direction. If the Zeeman effect is strong, therefore, the Hal resistance cannot be a function only of the field component parallel to the 2D-plane.

Although the reason of the absence of the Zeeman effect is not clear now, we tentatively ascribe it to the broadening of Landau levels due to scattering. If the broadening is comparable to the Zeeman energy, it will smear out the Zeeman effect.

3. Summary

In this work, we investigated the Hall effect of $\alpha$-(BEDT-STF)$_2$I$_3$ under high pressure of 1.8GPa. (1) We observed a Hall resistivity that increases linearly in low $B$ and then saturate to a value of Landau degeneracy which is given as $\frac{e^2}{2h}$. (2) This change in the Hall resistivity reflects the change in the carriers: from thermally excited carriers to carriers on Landau levels and then to the zero-mode Landau carriers. (3) The same mechanism works in the transport phenomena in $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\alpha$-(BEDT-TSF)$_2$I$_3$. (4) We could not detect the Zeeman effect.

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References

[1] Bender K et al. 1984 *Mol. Cryst. Liq. Cryst.* **108** 359
[2] Inokuchi M et al. 1995 *Bull. Chem. Soc. Jpn.* **68** 547-53
[3] Kobayashi A et al. 2005 *J. Phys. Soc. Jpn.* **74** 2897
[4] Katayama S et al. 2006 *cond-mat* 0601068
[5] Tajima N et al. 2006 *J. Phys. Soc. Jpn.* **75** 051010