Transport properties of organic conductor $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\theta$-(BEDT-TTF)$_2$I$_3$ under hydrostatic pressure or uniaxial strain

N. Tajima$^a$, A. Tajima$^b$, M. Tamura$^a$, Y. Nishio$^b$, K. Kajita$^b$

$^a$Condensed Molecular Materials Laboratory, RIKEN, Wako, Saitama 351-0198, Japan
$^b$Department of Physics, Toho University, Miyama, Funabashi, Chiba 274-8510, Japan

Abstract

Pressure controlled switching between a metallic state and a semiconducting state was successfully realised on organic conductors $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\theta$-(BEDT-TTF)$_2$I$_3$. Under high hydrostatic pressures, $\alpha$-(BEDT-TTF)$_2$I$_3$ behaves as an ultra-narrow gap (about 1 meV at $p = 18$ kbar) semiconductor. It is characterised by strongly temperature($T$)-dependent carrier density. It decreases by about $10^6$ times between 300 and 1 K. When strained in $b$-axis, on the other hand, this material was found to behave as a quasi-two-dimensional (Q2D) metal with a large Fermi surface. The carrier density is independent of $T$ while conductivity increases by about $10^2$ times between 300 and 4 K. $\theta$-(BEDT-TTF)$_2$I$_3$ exhibits a similar change. Under the ambient pressure, it is a typical Q2D metal and changes to a narrow gap semiconductor under pressures above about 5 kbar.

Keywords: Narrow gap semiconductor; Mobility; Carrier density; Pressure; Uniaxial strain

1. Introduction

Pressure has been one of the important parameters in the investigation of organic conductors, since organic crystals are soft and sensitive to pressure. A weak pressure can, sometimes, cause a drastic change in the properties of organic conductors.

In this paper, we report the effect of pressure on the transport property of two organic conductors, $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\theta$-(BEDT-TTF)$_2$I$_3$. These two crystals have layered structure in which conductive layers of BEDT-TTF molecules and insulating layers of I$_3$ molecules pile up alternately. Although the arrangement of BEDT-TTF molecules in these materials is similar to each other, small difference in the arrangement of molecules gives rise to the qualitative difference in the transport property [1,2]. Under the ambient pressure, $\theta$-(BEDT-TTF)$_2$I$_3$ is a typical two-dimensional metal. On the other hand, $\alpha$-(BEDT-TTF)$_2$I$_3$ is also metallic at high temperatures above 135 K where it undergoes a transition to an insulator.

We found that the property of these materials can be controlled by pressure or strain, i.e., we can change them from metals to semiconductors or from semiconductors to metals by applying pressure.

In the experiment, we measured resistance and Hall voltage as functions of temperature and magnetic field. A conventional d.c. method with six probes was adopted. Either hydrostatic pressure or uniaxial strain was applied on samples.

2. Experimental results and discussion

2.1. $\alpha$-(BEDT-TTF)$_2$I$_3$

$\alpha$-(BEDT-TTF)$_2$I$_3$ behaves as a metal above 135 K where it undergoes a transition to an insulator. We can suppress this transition by applying pressures [1]. The hydrostatic pressure acts to shift the critical point of metal–insulator transition towards low temperatures, and finally, under the pressures above about 15 kbar, the transition disappears [3] (see the inset of Fig. 2). Under a pressure between 15
and 20 kbar, we found that the temperature dependence of the resistance remains qualitatively the same. It is nearly constant between 300 and 1.5 K.

The crystal in the high-pressure state looks like a metal, since the conductivity at the lowest temperature (1.5 K) is finite. It, however, is not a metal but a semiconductor. Experimental results of Hall effect indicate that the carrier density is not constant but decreases by about six orders of magnitude from 300 and 1.5 K (Fig. 1). From the carrier density at the lowest temperature region below 4 K, the energy gap was estimated to be about 1 meV. As a conclusion, \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) under high hydrostatic pressures is a semiconductor with an extremely narrow energy gap [4].

Recently, we adopted a new experimental technique developed by Maesato et al. [5] and examined the property of this material under uniaxial strains. We found a drastic and interesting change occurs in the transport phenomena when the crystal is uniaxially compressed in the \(b\)-axis of the crystal.

As is mentioned in the previous section, temperature independent resistance is the characteristic feature of this sample under high hydrostatic pressures (20 kbar, for example). When the sample is compressed in the \(b\)-axis (refer to the data for \(p_{/b\text{-axis}} = 5\) kbar), on the other hand, we find the resistance decreases by about two orders of magnitude from 300 to 4 K. This change in the behaviour of resistance suggests the change in the character of the electron system. Actually, the effective carrier density (\(n_{\text{eff}}\)) estimated from Hall coefficient is almost constant all over the temperature region. It is astonishingly different from the one shown in Fig. 1.

More detailed information is given by examining the data in Fig. 2. We found the curve of \(n_{\text{eff}}\) resembles to that of \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) under the ambient pressure. The coincidence is not only qualitative but also quantitative including a drop that occurs between 30 and 10 K. As \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) under the ambient pressure is a two-dimensional metal, we can conclude that under a strain along \(b\)-axis, this material behaves as a Q2D metal with a large Fermi surface. Moreover, we expect that the energy band structure near the Fermi level is similar to that of \(\theta\)-(BEDT-TTF)\(_2\)I\(_3\).

Concluding this section, we found that \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) behaves as a narrow gap semiconductor when placed under hydrostatic pressures above 15 kbar, while if compressed in the \(b\)-axis, it is a metal with a large Fermi surface.

2.2. \(\theta\)-(BEDT-TTF)\(_2\)I\(_3\)

Under the ambient pressure, this material is a typical Q2D metal with the carrier density of about \(10^{21}\) cm\(^{-3}\). As the carrier mobility increases with decreasing temperature while the density is constant, a reduction of resistance occurs with decreasing temperature. Under hydrostatic pressures above about 5 kbar, on the other hand, it changes to a narrow gap semiconductor. In the high-pressure state, both the carrier density and the mobility depend strongly on the temperature. Each of them changes by about six orders of magnitude from 300 to 1.5 K. These behaviours are similar to what we observe in \(\alpha\)-(BEDT-TTF)\(_2\)I\(_3\) under high hydrostatic pressures. Here again, we find an organic conductor that pressure can change from a metal to a semiconductor.

3. Conclusion

In conclusion, pressure controlled switching between a metallic state and a semiconducting state was successfully
realised on organic conductors $\alpha$-(BEDT-TTF)$_2$I$_3$ and $\theta$-(BEDT-TTF)$_2$I$_3$. Characteristic feature of the transport phenomena of two crystals in the metallic state resembles each other as well as those in the semiconducting state.

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References