Magneto-transport properties in the density wave phase of $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$

R Kondo,* J Otsuji and S Kagoshima
Department of Basic Science, University of Tokyo, Komaba, Meguro-ku, Tokyo, Japan

e-mail: crkondo@mail.ecc.u-tokyo.ac.jp

Abstract. Magneto-transport properties of the newly prepared layer-structured organic conductor $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ are investigated. Temperature dependence of the interlayer resistance without magnetic fields shows a hump-like anomaly between 32 K and 5 K at ambient pressure. Measurements of the Shubnikov-de Haas oscillations indicate that the electronic structure at low temperature is different from that calculated from the crystal structure at room temperature. Under magnetic fields parallel to the layer, magnetic field dependence of the interlayer resistance also shows a large dip-like anomaly below 32 K, where the hump-like anomaly starts to appear. We suggest that the hump-like anomaly of $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ is a density wave transition accompanied by nesting of its one dimensional Fermi surfaces, and that the dip-like anomaly in the magneto-resistance is ascribed to magnetic field dependence of the interlayer hopping of electrons.

1. Introduction
Low dimensional organic conductors belong to one of the most interesting material groups in solid-state physics and chemistry [1]. Among them, quasi one-dimensional ones have provided rich variety of phenomena such as not only superconductivity and density waves but also oscillatory resistance phenomena and field-induced density waves under magnetic fields [2, 3].

Recently, we synthesized the organic conductor $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ (abbreviated as $\beta''$-CsCd), where BEDT-TTF denotes bis(ethylene)dithio-tetrathiafulvalene. This material also has a pair of large one-dimensional Fermi surfaces and a pocket, as shown in figure 1. Temperature dependence of the resistance of this salt showed a hump-like anomaly between 32 K and 5 K, which is reminiscent of a charge density wave transition in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ around 8 K [4]. Measurements of the Shubnikov-de Haas oscillations indicate that the electronic structure of $\beta''$-CsCd at low temperature is different from that calculated from the crystal structure at room temperature. Moreover, we found magnetic field dependence of the interlayer resistance under the magnetic field parallel to the layer also

Figure 1. Calculated Fermi surface of $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ based on the crystal structure at room temperature.
showed a large dip-like anomaly below 32 K where the hump-like anomaly started to appear. These results suggest that the hump-like anomaly is a density wave transition accompanied by nesting the one-dimensional Fermi surfaces, and that the dip-like anomaly in the magneto-resistance is ascribed to magnetic field dependence of the interlayer hopping of electrons.

In this report, we present the interlayer magneto-resistance under magnetic field perpendicular and parallel to the layer at low temperature, and discuss the origin of the hump-like anomaly and the dip-like one of $\beta''$-CsCd.

2. Experimental procedure
The samples were prepared by electro-oxidization of BEDT-TTF in the presence of suitable electrolytes.

Measurements of the interlayer resistance were performed by conventional four-probe method with gold wires (25 µm φ) attached to a crystal using carbon paste in the temperature range of 0.6 - 300 K.

The magnetic fields were applied up to 13.5 T with a solenoid type magnet. The sample orientation was set by hand and errors in the orientation were estimated as less than 3 deg.

3. Results and discussion

3.1. Under magnetic fields perpendicular to the layer
Figure 2 shows the magnetic field dependence of the interlayer resistance of $\beta''$-CsCd. The magneto-resistance shows clear Shubnikov-de Haas (SdH) oscillations from very low magnetic fields below 1 T, as shown in the inset. The oscillations are composed of large-amplitude-slow-frequency ones in the low magnetic field region and small-amplitude-high-frequency ones in the high magnetic field.

The analysis of FFT spectrum of the oscillations indicates the existence of at least four kinds of frequencies. They are relevant to the areas of the first Brillouin zone, 0.02 %, 0.09 %, 1.01 %, and 3.39 %, respectively. The calculated Fermi surface in figure 1 gives two possible orbits for the SdH oscillations, the small pocket whose center is at X (11.2 % of the first Brillouin zone) and the large orbit whose center is at Γ (50 %). This discrepancy in the observed and calculated results indicates that

![Figure 2](image)

Figure 2. The interlayer resistance of $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ under the magnetic field perpendicular to the layer. The inset shows the enlarged view below 1.25 T.
the Fermi surface at low temperature is quite different from that at room temperature even when we take account of the misalignment of the sample against the magnetic field.

3.2. Under magnetic fields parallel to the layer
Figure 3 shows the magnetic field dependence of the interlayer resistance under the field parallel to the layer (ac-plane) up to 12 T. The inset shows the sample configuration against the magnetic field. With increasing the field strength from zero, the resistance starts to decrease rapidly and has the minimum around 8 T, and then increases rapidly after the minimum point.

The position of the resistance minimum (dip) has angular dependence against the direction of the magnetic field in the layer; under the field along c-axis the minimum was around 6 T while under the field along a-axis that was above 12 T and could not be determined precisely.

Temperature dependence of the behavior of the dip is as follows; with decreasing temperature from 32 K to 5 K the position of the minimum appeared and shifted from zero to higher magnetic field, and the depth gradually increased. Below 5 K the position and the depth did not vary any more. The temperature range where the minimum appeared and grew was nearly same as that where the hump-like anomaly appeared, indicating the origin of the resistance minimum has close relationships with the hump-like anomaly.

One of promising explanations for this dip-like anomaly is given by the incoherent interlayer transport model [5, 6, 7], here “incoherent” means that the intra-layer scattering rate is much larger than the interlayer tunneling rate. In this situation the interlayer resistance is proportional to the tunneling rate between two neighboring layers. According to the model, this rate is proportional to an overlap between wave functions on the two neighboring layers. Since the wave functions contain a vector potential, the overlap of wave functions and the tunneling probability generally varies with changing the magnetic field [6, 7].

When the magnetic field parallel to the layer arrives at a threshold value where the overlap vanishes, the interlayer resistance increases rapidly. The threshold field depends on the carrier density.
on each layer; for example, a large density needs a high threshold field. This has been observed experimentally in the interlayer resistance of the semiconductor under magnetic fields [8]. It is to be noted that this model does not need an electronic-phase transition for the dip-like anomaly of resistance.

According to the above model, the rapid increase in the interlayer resistance above the resistance minimum, 8 T, is considered to correspond to the decrease in the tunneling rate for the electronic system having a much smaller carrier density than that of the Fermi surfaces of figure 1. It is because a few thousand Tesla of magnetic field needs for the size of the pockets in figure 1 if the above phenomena would be observed. The appearance of the dip-like anomaly under such “low “ magnetic field might indicate I) the electronic structure of $\beta''$-CsCd changes from that shown in figure 1 to another having small pockets whose SdH oscillations are considered to be observed in figure 2, and II) the process of the interlayer transport in $\beta''$-CsCd is incoherent, which is consistent with the observation of an anomalous background magnetoresistance in the measurement of angular dependent magneto resistance oscillation (ADMRO) [3].

Based on the above results, the hump-like anomalies below 32 K in the temperature dependence of the interlayer resistance should be ascribed to a density wave transition accompanied by the variation of the electronic structure, since I) $\beta''$-CsCd has a pair of large one-dimensional Fermi surface, which may be nested and II) Actually, $\beta''$-(BEDT-TTF)$_2$AuBr$_3$ [9, 10], which has the same molecular arrangement as $\beta''$-CsCd, also has a density wave transition although they showed no remarkable anomaly in their resistance measurement [10, 11, 12].

4. Summary
We investigated the magneto-transport properties of newly prepared organic conductor $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ having the hump-like anomaly below 32 K in the temperature dependence of the interlayer resistance. Based on the measurements of the interlayer resistance under the magnetic field perpendicular and parallel to the layer, we suggested that the hump-like anomaly of $\beta''$-(BEDT-TTF)$_2$CsCd(SCN)$_4$ is a density wave transition accompanied by nesting of its one dimensional Fermi surfaces, and that the dip-like anomaly in the magneto-resistance is ascribed to magnetic field dependence of the interlayer hopping of electrons.

We thank Professor T. Osaka (ISSP) for his valuable discussion about interlayer transports of layer-structured materials under magnetic field.

References
[1] Ishiguro T, Yamaji K, and Saito G 1997 Organic Superconductors 2nd ed (Berlin:Springer)
[2] Karsovinik M 2004 Chem. Rev. 104 5737
[3] Osada T and Ohnishi E 2006 J. Phys. Soc. Jpn. 75 051006
[4] Sasaki T, Toyota N, Tokumoto M, Kinoshita N and Anzai H 1990 Sol. Stat. Commun. 75 93
[5] Yoshioka D 1995 J. Phys. Soc. Jpn. 64 975
[6] McKenzie R H and Moses P 1998 Phys. Rev. Lett. 81 4492
[7] Osada T 2002 Physica E 12 272
[8] Kobayakawa M 2006 Doctor thesis, University of Tokyo
[9] Mori T, Sakai F, Saito G and Inokuchi H 1986 Chem. Lett. 1037
[10] Kurmoo M, Talham D R, Day P, Parker I D, Friend R H, Stringer A M, and Howard J A K 1987 Solid State Commun. 61 459
[11] Doporto M, Singleton J, Pratt F L, Caulfield J, Hayes W, Perenboom J A A J, Deckers I, Pitsi G, Kurmoo M, and Day P 1994 Phys. Rev. B 49 3934
[12] House A A, Harrison N, Blundell S J, Deckers I, Singleton J, Herlach F, Hayes W, Perenboom J A A J, Kurmoo M, and Day P 1996 Phys. Rev. B 53 9127