反強磁性金属体β"-(EDO-TTFVODS)$_2$FeBr$_4$(DCE)$_{0.5}$における磁気抵抗
Magnetoresistance in an antiferromagnetic metal β"-(EDO-TTFVODS)$_2$FeBr$_4$(DCE)$_{0.5}$

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1. Introduction

Recently organic conductors including magnetic moments in the counter ions have attracted a great deal of attention, because they show intriguing phenomena originating from the π-d interactions between the local magnetic moments and the itinerant conduction electrons [1,2]. In particular, large negative magnetoresistance (MR), field-induced metal-insulator (M-I) transition and field-induced superconductivity. All of these properties are also attractive for the wide scope of application for molecule-based electronic device where the conductivity can be controlled by the magnetic field. However, the π-d interactions are not so strong in the present organic conductors, in comparison to the s-d interaction in traditional metal-based magnets. The reason of this weakness is due to the localized magnetic electrons and the conducting electrons are separately placed on different molecules, in other words, the former are placed on donor layers and the latter are placed on anion layers having d electrons. Therefore, to apply the π-d systems to molecule-based devices, it is indispensable to enhance the intermolecular interaction.

The titled material possesses a bent donor structure. This bent structure is expected to play a role to produce the stronger π-d interaction through the Se---halogen contacts between 1,3-diselenole ring and magnetic anions [3]. According to the molecular orbital calculation, β"-(EDO-TTFVODS)$_2$FeBr$_4$(DCE)$_{0.5}$ (abbreviated as β”-FeBr$_4$ hereafter) gives a relatively large indirect interaction $J_{\pi d}$ (14.5 K) which is comparable to that of λ-(BETS)$_2$FeCl$_4$ (17.7 K), where BETS is bis(ethylenedithio)tetraselenafulvalene [4]. Therefore a strong π-d interaction is expected in β”-FeBr$_4$. In this report, we will present the results acquired at High Field Laboratory for Superconducting Materials (HFLSM), Institute for Materials Research, Tohoku University.

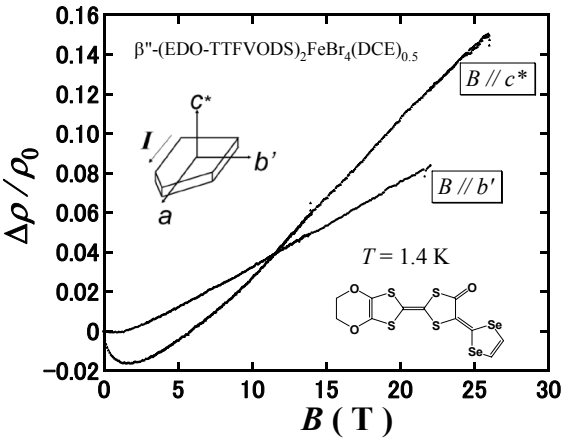
2. Experimental

Electrical resistivity was measured by conventional four-probe method along the directions parallel and perpendicular to the most conducting *ab* plane. Four annealed Au wires (10 μmϕ) were attached with carbon paste to the crystal surface that had been Au deposited in advance. To fix the sample in high magnetic field, we coated the sample with silicon vacuum grease on the holder. The magnetoresistance measurement was carried out by using a 28 T hybrid magnet at the Institute for Materials Research (IMR) at Tohoku University.

3. Results and discussion

This sample shows a metallic temperature dependence of the electrical resistivity down to about 4 K and a slight upturn proportional to log$T$ below 4 K [5]. Heat capacity and static magnetic susceptibility measurements show that β"-FeBr$_4$ undergoes an antiferromagnetic transition at 4.5 K with the easy axis along the c*-direction [3].

Fig.1. Transverse magnetoresistance of



(EDO-TTFVODS)$_2$FeBr$_4$(DCE)$_{0.5}$ vs. B for B//c* and B//b'. T = 1.4 K.

Magnetoresistances were measured with the field directions parallel to b'- and c*-axes at 1.4 K as shown in Fig. 1. Namely, the electrical current is perpendicular to B in both the results in Fig. 1. Only

positive magnetoresistance was observed for $B//b'$ at the temperature. The negative magnetoresistance was observed below 7 T for $B//c^*$. Above 7 T, the magnetoresistance becomes positive and increases more rapidly than that for $B//b'$. No Shubnikov-de Haas oscillations were observed for $B//c^*$ even at 0.5 K and up to 26 T, although at least one Q2D Fermi surface is considered to be exist as shown in angular dependence of magnetoresistance which will be mentioned later. It is interesting that, even for $B//b'$, the magnetoresistance does not show the B^2-dependence as is usually expected to in metals.

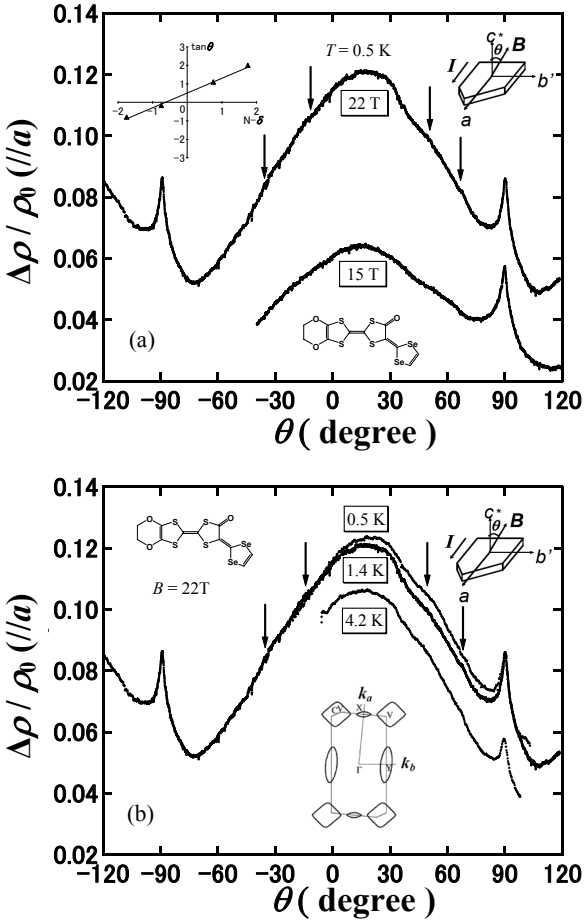


Fig. 2 (a) Angular (θ) dependent magnetoresistance at 15 T and 22 T at 0.5 K. The inset is the index N dependence of $\tan\theta$, where θ is the position of the magnetoresistance maxima. (b) Angular (θ) dependent magnetoresistance at 0.5 K, 1.4 K and 4.2 K at 22 T.

The negative magnetoresistance for $B//c^*$ amounts to only -1.7% at 1.4 K and -2.1% even at 0.5 K (not shown), respectively. These values are much smaller than those observed for other π-d organic conductors such as $(DMET)_2FeBr_4$ (-11%) [6,7], $(EDT\text{-}TTFBr_2)_2FeCl_4$ (-23%) [8] and $(EDT\text{-}DSDTFVO)_2FeCl_4$ (-15%) [9]. These Fe-containing salts show the negative magnetoresistance in their insulating states. Since the electrical conductivity of β''-$FeBr_4$ is, however, kept high even below T_N = 4.5 K, the π-d interaction, that is the most probable origin of the negative magnetoresistance of these insulating systems, is screened by the conducting electrons. Another possible mechanism to explain the negative magnetoresistance is the weak localization [5]. Orbital motions of the conducting electrons within the 2D layer are expected not for $B//b'$ but for $B//c^*$. This is consistent with the weak localization caused by orbital motions of carriers in possible random potentials. On the other hand, as for Kondo effect, the negative MR is also explained by reducing the spin-flip scattering [10]. Therefore, MR associated with Kondo effect can not be discarded.

Figure 2(a) shows the field-orientation dependence at 0.5 K measured at different magnetic field strength (15 and 22 T). The angle θ between B and c^*-axis was varied by rotating B around the a-axis. Sharp cusps are observed when B is parallel to the 2D plane ($B//b'$), which is due to the zero negative component in magnetoresistance as $B//b'$. A broad maximum of the magnetoresistance is at $\theta \sim 15^\circ$. The asymmetric field-orientation dependence is consistent with the triclinic crystal structure. The sample was immersed in liquid ^{3}He during the measurement. The sweep direction of the angle was from right to left for the 22-T data in Fig. 2(a). Slightly higher values of the magnetoresistance at the left side than that at the corresponding angle position at right side is explained by gradual temperature drift (decrease) and the temperature dependence of the resistivity. Four arrows in Fig. 2(a) indicate the peak positions in magnetoresistance probably due to Kajita-Yamaji oscillation [11] because their positions are independent of the magnetic field strength (Fig. 2(a)). This phenomenon is one of angular-dependent magnetoresistance oscillations (AMRO) observed for two-dimensional metals and explained in terms of orbital motion of electrical carriers on a Q2D Fermi surface. The positions are not affected by temperature as in Fig. 2(b) showing these anomalies are not caused by a kind of field-induced phase transition like that in $(TMTSF)_2PF_6$ (TMTSF = tetramethyltetraselenafulvalene) [12]. The angular position θ_N of the N-th anomaly of Kajita-Yamaji oscillation satisfy the following equations [11],

$$k_F d \tan\theta_N = \pi(N \mp \delta), \qquad (1)$$

where N is ±1, ±2, ±3 ..., δ = 1/4 and -1/4 for positive and negative N, k_F is the Fermi wave vector and d = 20.078 Å is the distance between neighboring conducting layers. The angular positions of the arrows are plotted as $\tan\theta_N$ against $N \pm \delta$ in the inset of Fig. 2(a).

Since the data points satisfy the eq. (1), we conclude that 1) the anomalies are due to Kajita-Yamaji oscillation and 2) at least one Q2D Fermi surface exists even below T_N. We could estimate $k_{F,b'}$ as 0.19 Å^{-1}. The calculated $k_{F,b'}$'s based on tight-binding approximation are 0.11 Å^{-1}, 0.05 Å^{-1} and 0.05 Å^{-1} for the three Q2D Fermi surfaces in the inset of Fig. 2(b), respectively. At present, we can not explain the difference between observed and calculated Q2D Fermi surfaces in number and size.

It should be noted that Kajita-Yamaji oscillation and other AMRO phenomena are usually observed in the magnetoresistance measured along the direction perpendicular to the most conducting plane. We studied the in-plane resistivity ρ_{ab} as shown in Fig.2, although the inter-plane ρ_{c*} is more commonly studied. Semiclassical calculations based on the solution of Boltzmann equation with relaxation time approximation show that the AMRO anomalies are distinct for the inter-plane resistivity. Due to the difficulty in measuring pure in-plane resistivity of low-dimensional materials, it is possible that observed ρ_{ab} reflects the behavior of ρ_{c*}. The positions of the AMRO anomalies are, however, independent of the current directions.

4. Conclusion

β”-$FeBr_4$ with 2D Fermi surface possesses a metallic behavior in spite of a slight upturn observed below 4 K. With lowering temperature, resistivity of upturn increases as a function of log$T$. Rather weak (-1.7% at 2 T) and magnetic field orientation dependent negative magnetoresistance was observed at 1.4 K. Besides π-d interaction screened by conducting electrons in metallic state of β”-$FeBr_4$, both weak localization and Kondo effect are possible origins for explaining this negative magnetoresistance. Based on these proceeding works at Osaka City University, we studied high field properties at IMR, Tohoku University. The oscillatory behavior due to Kajita-Yamaji oscillation was observed in angular dependent magnetoresistance and $k_{F,b'}$ was estimated to be 0.19 Å^{-1}. However, no Shubnikov-de Haas oscillations were observed.

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