Isotropic magnetoresistance anomaly in the antiferromagnetic anisotropic conductor, \(\beta''-(EDO-TTFVO)_{2}FeCl_{4}\)

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Abstract. We report the magnetoresistance (MR) in 2D-organic conductor, \(\beta''-(EDO-TTFVO)_{2}FeCl_{4}\). We found an anomaly at 8 T in MR, which is field-angle-independent. We speculate that this behavior is associated with from antiferromagnetic to paramagnetic transition, occurring at the same magnitude of magnetic field irrespective of field direction. To realize this situation, comparable magnitude of \(J_{\pi d}\) and \(J_{d}\) is required, which is rather rare the case. The isotropic nature in the magnetic transition is remarkable in the anisotropic organic conductors.

1. Introduction
Organic conductors exhibit a variety of physical properties, which can be systematically understood on the bases of simple and clear electronic structure. So the study of organic conductors containing magnetic anions is one of the important issues to explore the novel phenomena related to an interaction between itinerant \(\pi\)-electrons and localized \(d\)-spins, so-called \(\pi-d\) interaction. The organic conductors with the \(\pi-d\) interaction show various interesting phenomena, for example a ferromagnetic metal of (ET)\(_3\)MnCr(C\(_2\)O\(_4\))\(_3\)[1], antiferromagnetic superconductivity of \(\kappa-(BETS)_{2}FeBr_{4}\)[2], and magnetic field induced superconductivity of \(\lambda-(BETS)_{2}FeCl_{4}\)[3] and \(\kappa-(BETS)_{2}FeBr_{4}\)[4]. These phenomena are discussed in terms of the \(\pi-d\) interaction between the conducting \(\pi\)-electrons and localized magnetic \(d\)-spins. In order to study the scattering mechanism in \(\pi-d\) system, we studied the magnetoresistance (MR) in \(\beta''-(EDO-TTFVO)_{2}FeCl_{4}\), where EDO-TTFVO stands for ethylenedioxy-tetrathiafulvalenoquinone-1,3-dithiolmethide (see the inset of figure 3).

The \(\beta''\)-type organic conductor, \(\beta''-(EDO-TTFVO)_{2}FeCl_{4}\) is the first salt which shows stable metallic behavior down to 0.3 K among Sugimoto-salts[5]. As for magnetic property, this salt shows antiferromagnetic ordering at \(T_{N} \sim 3\) K with an easy axis parallel to the \(a\)-axis (perpendicular to the conducting bc-plane). In this salt, metallic and antiferromagnetic phases coexist in the ground state. The band calculation in a tight-binding method suggests the existence of a two-dimensional (2D) Fermi surface (FS) and the anisotropy of exchange
interaction is $J_{\pi d}/J_{dd} \sim 2.4$, where $J_{\pi d}$ and $J_{dd}$ are $\pi$-$d$ and $d$-$d$ exchange interaction, respectively.

![Diagram](image)

**Figure 1.** 2D-FS (a) and dispersion relation (b) in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$.

2. Experiment

Single crystals of $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ were synthesized by electrochemical method. This salt was black needle-like crystal with a conducting $bc$ plane. The resistance measurements were performed by an ac four-terminal method along the $a^*$-axis ($\rho_a$) for a measurement of the quantum oscillation effects. Electrical leads of annealed gold wire (10 $\mu$m) were attached to samples with carbon paint. The typical sample size was $0.4 \times 0.2 \times 0.02$ mm$^3$. The experiments were carried out with a dilution refrigerator with a 14 T superconducting magnet at Osaka City Univ. and with a $^3$He cryostat with a 27 T hybrid magnet at the High Field Laboratory, IMR, Tohoku Univ.. The samples were mounted on the single-axis rotatable holder.

3. Results and Discussion

Temperature dependence of the out-of-plane resistivity ($\rho_o$) of the $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ is shown in figure 2 for three samples. Samples 1 and 2 are from the same batch, showing a metallic behavior in resistivity from room temperature down to 0.5 K. However, Sample 3 showed a slight upturn below 4.0 K. Residual Resistance Ratio (RRR) is 113, 62 and 34 for Samples 1, 2 and 3, respectively. The MRs in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ (for Samples 1, 2 and 3) are shown in figure 3. Below 8 T the hump is sample dependent, which have a tendency to be enhanced as the RRR is small. Thus the difference in shape of the anomaly must be caused by the effect of impurities. On the other hand, the appearance of the kink anomaly around 8 T is common to all samples. The anomaly of this kind is observed in $\kappa$-(BETS)$_2$FeBr$_4$[7], where the kink anomaly is understood as the transition from the antiferromagnetic state to the paramagnetic state. In $B < 1.2$ T, we can not compare directly because the easy axis in $\kappa$-(BETS)$_2$FeBr$_4$ is parallel to the conducting layers while in the title compound, it is perpendicular to the layers. However, since this $\beta''$-salt undergoes the spin-flop around 1.2 T, spin orientation becomes parallel to the plane. In $\kappa$-(BETS)$_2$FeBr$_4$, the kink anomaly is observed around 5 T with $B \perp$ plane. Thus the background orientation of $B$ and $d$-spins are the same between the two kind of salts, i.e. for present salt ($B > 1.2$ T) and $\kappa$-salt ($B > 0$ T).

Shubnikov-de Haas (SdH) oscillations are observed at 0.5 K above 17 T in Samples 2 and 3 as shown in figure 3. Fourier transform spectrum of the SdH oscillations shows the frequency of 602 T, whose cross-sectional area corresponds to 13% of the first Brillouin zone (FBZ). This value is about twice as large as the calculated value (7.2%). At present, the reason of this difference
Figure 2. Temperature dependence of the out-of-plane resistivity ($\rho_a$) from 150 K down to 0.5 K in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ (for Samples 1, 2 and 3). The upturn was observed around 4 K in Sample 3 and no hysteresis was seen in all samples.

Figure 3. MRs in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ (for Samples 1, 2 and 3). The upper inset shows the structure of EDO-TTFVO. The batch of Samples 1 and 2 is the same. The kink anomaly appears at 8 T in all samples.

is unknown. The frequencies of the oscillations as a function of $\theta$ follow $F(\theta) = F_0 / \cos \theta$, which is typical of the 2D FS. The cyclotron effective mass $m_c$ is estimated to be $m_c \sim 3.3 m_e$ by Lifshitz-Kosevich formula[8], where $m_e$ is the free electron mass.

Figure 4 presents the temperature dependence of MRs in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ (for Sample 1). The anomaly around 8 T appears below 3.5 K and shifts slightly to higher field by 1 T between 3.5 K and 0.3 K with decreasing temperature as the inset shows. Since no hysteresis is observed, this anomaly corresponds to a second order transition from the antiferromagnetic to paramagnetic states taking into account that this material shows antiferromagnetic transition at 3K. The angular dependence of MRs at 0.5 K is shown in figure 5. The inset shows that the peak appears at 8 T irrespective of the direction of the magnetic field. In $\kappa$-(BETS)$_2$FeBr$_4$, however, the kink anomaly shifts to lower field as increasing the angle. This $\kappa$-salt shows the metamagnetic transition. In this case the magnetic field component parallel to the easy axis is effective. The spin-flop is observed in $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ around 1.2 T. When the magnetic anisotropy is small, the spin-flop occurs. In $\beta''$-(EDO-TTFVO)$_2$FeCl$_4$ the anisotropy of exchange interactions $J$ is estimated $J_{\pi d}/J_{dd} \sim 2.4$. This value is very much isotropic in comparison with $\kappa$-(BETS)$_2$FeBr$_4$, which is calculated $J_{\pi d}/J_{dd} \sim 22$[9].

4. Summary
In order to study its electronic structure and the $\pi$-$d$ interaction, we carried out the MR study in this salt. We observed the kink anomaly near 8 T below 3.5 K (close to $T_N$) and the SdH oscillations with the frequency of 602 T, whose cross-sectional area corresponds to 13% of the FBZ. The cyclotron effective mass of the $\pi$ electron is estimated to be $m_c \sim 3.3 m_e$. The anomaly becomes more prominent and shifts slightly to higher field by 1 T between 3.5 K and 0.3 K with decreasing temperature. The angular-dependence of the MR shows that the 8 T dip anomaly is independent of angle between the $a^*$-axis and the magnetic field in the $ac$-plane at least in the $ac$-plane. To realize this situation, comparable magnitude of $J_{\pi d}$ and $J_{dd}$ is required, which
is rather rare the case in the organic conductors. This feature can be understood in terms of two kinds of the AF exchange interactions, $J_\pi d/J_{dd} \sim 2.4$. The isotropic nature in the magnetic transition is remarkable in the anisotropic organic conductors.

Acknowledgments
This work was partly supported by a Grant-in-Aid for Scientific Research on Priority Areas of Molecular Conductors (No. 15073220) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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