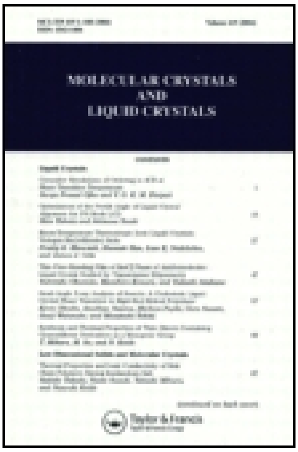

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Electrical and Magnetic Properties of Weak Ferromagnetic Organic Conductor, τ-(EDO-S,S-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$, (y≁0.75) and its Analogs

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Electrical and Magnetic Properties of Weak Ferromagnetic Organic Conductor, τ-(EDO-*S*, *S*-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$, ($y \sim 0.75$) and its Analogs

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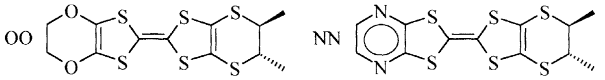
Abstract The electric resistivity was measured for τ- (EDO-*S*,*S*-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$ (1), τ-(EDO-*S*,*S*-DMEDT-TTF)$_2$- (I$_3$)$_{1+y}$ (2) and τ-(P-*S*,*S*-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$ (3), (y~0.75) under hydrostatic pressure and magnetic field. Pressure increases the M-I transition temperature of (2) and (3). The period of the angular dependence of the magnetoresistance of (1) at 5 T changes from 90° to 180° by pressure of 5 kbar, while that of (2) and (3) is not changed.

Keywords τ-type conductor; magnetoresistance; reflectance spectra

INTRODUCTION

Electron donors, ethylenedioxy-*S*,*S*-dimethylethylenedithio-tetrathiafulvalene (abbreviated as EDO-*S*,*S*-DMEDT-TTF or, in this paper, as OO) and pyrazino-*S*,*S*-dimethylethylenedithio-tetrathiafulvalene (P-*S*,*S*-DMEDT-TTF or NN) give an isostructural group of charge transfer salts known as τ-type conductors such as τ-$(OO)_2(AuBr_2)_{1+y}$ (abbreviated as OO-$AuBr_2$), τ-$(NN)_2(AuBr_2)_{1+y}$ (NN-$AuBr_2$) and τ-$(OO)_2(I_3)_{1+y}$ (OO-I_3). The crystal of OO-$AuBr_2$ is



made of alternating stack of conducting and insulating layers resulting in two-dimensional nature.[1] The conducting layer parallel to the *ab* plane consists of the 2:1 composition of OO and $AuBr_2^-$, where the OO molecules construct a kind of square lattice. The tight-binding calculation predicts the four-fold Fermi surface. The remaining anions of y~0.75 exist in the insulating layer with random vacancies.

The electric resistivity of OO-$AuBr_2$ is metallic for both parallel and perpendicular to the conducting layer below room temperature and has broad minimum around T_{min}~50 K. Recently some of the present authors reported that, for the magnetic field rotated within the conducting layer and the electrical current perpendicular to the layer, the angular dependence of the magnetoresistance of OO-$AuBr_2$ switches its periodicity between 180° at 0.5 T and 90° at 5 T at 1.3 K and, furthermore, changes the principal axis by 90° at 0.5 T depending on the direction of the magnetic field at 5 T.[2] The 'switching effect' was explained by assuming weak ferromagnetic domains aligned along the magnetic filed of 5 T.[2] In this paper the magnetoresistance is studied under hydrostatic pressure not only for OO-$AuBr_2$ but also for NN-$AuBr_2$ and OO-I_3 to understand the novel electronic states of the τ-type conductors.

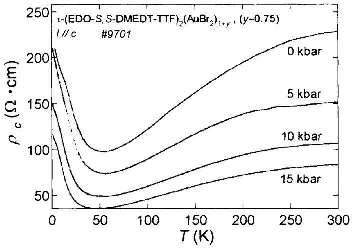
EXPERIMENTAL

Single crystals of the τ-type conductors were obtained by usual chemical oxidation method. The *c*-axis magnetoresistance under pressure up to 15 kbar was measured to extract the information on the anisotropy when the magnetic field is rotated within the most conducting *ab* plane. The pressure value in this study is that at the lowest temperature and about 1.5 kbar lower than that at room temperature. The high magnetic field measurement above 5 T was carried out using a hybrid magnet at Institute for Materials Research at

Tohoku University. At Institute for Solid State Physics at University of Tokyo, the reflectance spectra was measured for OO-$AuBr_2$.

RESULTS AND DISCUSSION

Figure 1 shows the *c*-axis electric resistivity of OO-$AuBr_2$. Although the magnitude of the resistivity is decreased by applying pressure, T_{min} are not changed very much. The angular dependence of the magnetoresistance at 1.2 K under pressure is, however, very different from that observed at ambient pressure as in Fig. 2. The period of the magnetoresistance changes from 90° to 180° between 0 and 5 kbar. The switching effect was not detected above 5 kbar, while the magnitude of the negative magnetoresistance is almost the same at any pressures. It must be noted that the *a*- and *b*-axes are equivalent in the crystal structure at room temperature. In fact, no anisotropy was detected by the reflectance spectra measurement. Figure 3 shows the result obtained for the polarization parallel to the *a*-axis at ambient pressure. The Drude-Lorentz fit gives the plasma frequency, ω_p~5.1×10^3 cm^{-1}. The Drude dispersion develops on lowering temperature down to 22 K. No anomaly has been observed in the specific heat between 4 and 80 K.[3] Both the results suggest that OO-$AuBr_2$ does not undergo any phase transitions at ambient pressure. Since the pressure does not change the overall temperature dependence of the resistivity, no phase transition is expected also under pressure.



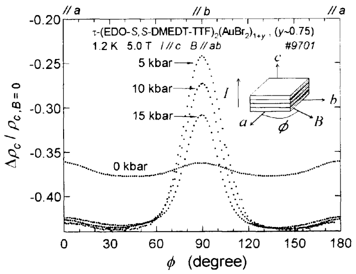
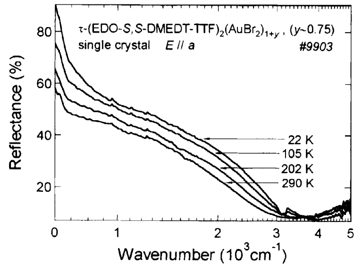


FIGURE 1 Temperature dependence of *c*-axis electric resistivity of τ-(EDO-*S*,*S*-DMEDT-TTF)$_2$($AuBr_2$)$_{1+y}$, (y~0.75, #9701).

FIGURE 2 Angular dependence of *c*-axis magnetoresistance of τ-(EDO-*S*,*S*-DMEDT-TTF)$_2$($AuBr_2$)$_{1+y}$, (y~0.75, #9701) for the magnetic field rotated within the most conducting *ab* plane.

Static magnetic susceptibility measurement for polycrystalline OO-$AuBr_2$ at 0 kbar, however, revealed that weak spontaneous magnetization develops below 30 K and amounts to $1.0\times10^{-3}\mu_B$/formula at 0 K.[4] This supports the magnetic domain model to explain the switching effect, though the origin of the magnetic moment is unclear at present. In this model, the 90° period of the magnetoresistance at 5 T is due to rearrangement of the domains following the rotated magnetic field and reflects the crystal symmetry (the space group $I4_122$).[2] The 180° period at 0.5 T at 0 kbar reported previously[2] shows the lower symmetry of the electronic system than that expected from the crystal structure. The 180° period under pressure suggests that the magnetic domains become hard to be moved even by the magnetic field of 5 T.



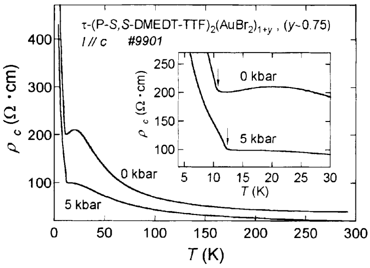
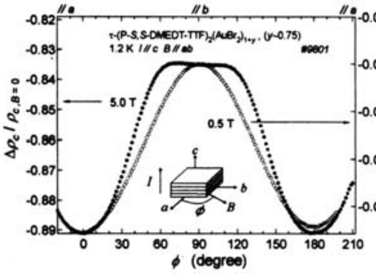


FIGURE 3 Polarized reflectance spectra (//*a*) of τ-(EDO-*S*,*S*-DMEDT-TTF$)_2$($AuBr_2$$)_{1+y}$, ($y$~0.75, #9903).

FIGURE 4 Temperature dependence of *c*-axis electric resistivity of τ-(P-*S*,*S*-DMEDT-TTF$)_2$($AuBr_2$$)_{1+y}$, ($y$~0.75, #9901).

Figure 4 shows the *c*-axis resistivity of NN-$AuBr_2$. The sharp M-I transition at $T_{M\text{-}I}$=11 K previously reported[5] was reproduced, though the *c*-axis resistivity is semiconducting even just below room temperature. It was found that $T_{M\text{-}I}$ is increased up to 12 K at 5 kbar and the anomalous hump just above $T_{M\text{-}I}$ is suppressed. The angular dependence of the magnetoresistance at 0 kbar at 1.2 K is shown in Fig. 5. The distinct negative magnetoresistance ($\Delta\rho_c/\rho_{c,B=0}$ ~ -0.9 at 5 T) and its 180° period was observed. It was found that the periodicity is not changed even at 24 T (Fig. 6), where the magnetic field directions parallel and perpendicular to one crystal edge are tentatively noted as *B*//*a* and //*b*, respectively, and the magnetoresistance saturates above about 8 T. The overall field dependence is similar to that of OO-$AuBr_2$ except for the hysteresis below 5 T for OO-$AuBr_2$.[2]



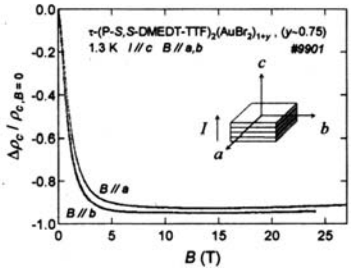
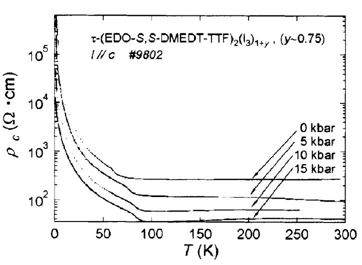


FIGURE 5 Angular dependence of c-axis magnetoresistance of τ-(P-S,S-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$, (y~0.75, #9801) for the magnetic field rotated within the ab plane.

FIGURE 6 Magnetic field dependence of the magnetoresistance of τ-(P-S,S-DMEDT-TTF)$_2$(AuBr$_2$)$_{1+y}$, (y~0.75, #9901).

The c-axis electric resistivity of OO-I$_3$ is almost independent of temperature below room temperature and shows rapid increase below $T_{\text{M-I}}$~70 K at 0 kbar and $T_{\text{M-I}}$ is increased up to about 90 K at 15 kbar (Fig. 7). The angular dependence of the magnetoresistance shows the period of 90° at 1.2 K and 5 T (Fig. 8). Rather large resistivity and small negative magnetoresistance prevented us to obtain its accurate angular dependence at lower magnetic field. No hysteretic behavior of the field dependence of the magnetoresistance below 5 T (not shown) suggests that OO-I$_3$ does not show the switching effect and the four-fold symmetry of its electronic system is not changed on the occasion of the M-I transition.



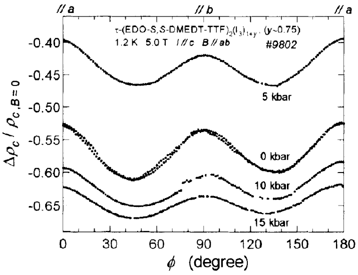


FIGURE 7 Temperature dependence of c-axis electric resistivity of τ-(EDO-S,S-DMEDT-TTF)$_2$(I$_3$)$_{1+y}$, (y~0.75).

FIGURE 8 Angular dependence of c-axis magnetoresistance of τ-(EDO-S,S-DMEDT-TTF)$_2$(I$_3$)$_{1+y}$, (y~0.75).

SUMMARY

The electric resistivity was measured for OO-$AuBr_2$, NN-$AuBr_2$ and OO-I_3 under hydrostatic pressure and magnetic field. It was found that the M-I transition or upturn of the resistivity is not suppressed by pressure for all the compounds studied in spite of the decrease in the resistivity. Since the τ-type donor packing is highly symmetric and considered to give larger bandwidth under pressure than that at ambient pressure, the mechanism of the M-I transition is probably different from that of the Mott insulator. Furthermore, the different angular dependence of the magnetoresistance of OO-$AuBr_2$, NN-$AuBr_2$ and OO-I_3 suggests that these compounds are in different electronic states from one another at least at low temperature.

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