Charge Disproportionation and Weak Localization in $\theta$-(BEDT-TTF)$_2$MZn(SCN)$_4$ [M=Cs,Rb]

Keizo Murata$^a$, K. Iwashita$^a$, H. M. Yamamoto$^b$, H. Yoshino$^a$, J. S. Brooks$^c$, K. Hiraki$^d$, T. Takahashi$^d$

$^a$ Dep. of Mat. Sci., Graduate School of Sci., Osaka City Univ., 558-8585, Osaka, Japan

$^b$ Riken, Wako-shi, 351-0198, Saitama, Japan

$^c$ Dept. of Physics, Florida State Univ./NHMFL, Tallahassee Fl, 32306-4005, USA

$^d$ Dep. of Physics, Gakushuin University, Mejiro, Toshima, 171-8588, Tokyo, Japan

Abstract

The metallic state of $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$ and $\theta$-(BEDT-TTF)$_2$CsZn(SCN)$_4$ was stabilized at low temperature by uniaxial strain along the a-axis and its magnetoresistance was examined. What we observed was the negative magnetoresistance, which could be explained by weak localization. We discuss this effect in terms of charge fluctuation associated with charge disproportionation.

Keywords: $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$, $\theta$-(BEDT-TTF)$_2$CsZn(SCN)$_4$, weak localization, charge fluctuation

1. Introduction

One of the $\theta$-materials shows superconductivity (SC). But most of the other $\theta$-materials exhibit metal-insulator (M-I) transition, associated with charge ordering (CO). It is of interest whether or not SC is realized by suppressing CO. By means of uniaxial pressure, we extended the metallic region below ~5 K and found the phenomena of weak localization but no SC. We propose that the origin of the weak localization is the fluctuation of CO.

H. Mori [1] has proposed that the $\theta$-type BEDT-TTF salts could be systematically assigned in a single diagram by the dihedral angle ($\theta$) between the donor-molecules in the crystals. In other words, larger the dihedral angle, more insulating the electronic states, i.e. the higher the metal-insulator transition temperature. When the dihedral angle is narrow enough, metallic state is stabilized, and even the superconductivity (SC) is observed. The Fermi surface in $\theta$-type is calculated to be of 2-dimension, quarter-filled and closed in a 1st Brillouin zone.

Recently, in $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$ (abbreviated as $\theta$-RbZn), the evidence of charge ordering, which is driven by the intersite Coulomb interaction ($V$) below the M-I transition temperature (190 K) and a fluctuation of charge order pattern above $T_{sc}$ were reported [2]. Also, in the $\theta$-(BEDT-TTF)$_2$CsZn(SCN)$_4$ (abbreviated as $\theta$-CsZn) which is located closer to the metallic phase than $\theta$-RbZn, the charge fluctuation among the BEDT-TTF molecules along the c-axis just above the M-I transition temperature (20 K) has been pointed out [3]. In this work, we shed light on this M-I transition and/or charge fluctuation associated with charge disproportionation by means of transport measurement.

Our idea is that the charge fluctuation might appear in the similar way as electron weak localization phenomenon in magnetoresistance. However, the effect of the localization is usually more appreciable at low temperature and in the metallic state. In order to combine the metallic state with low temperature, the uniaxial compression along the a-axis (strain method), which suppresses the dihedral angle, was indispensable to suppress the M-I transition, because hydrostatic pressure only gives rise the M-I temperature.

From the temperature dependence of the resistivity under uniaxial strain $/a$, the M-I transition is controlled to be suppressed as expected. We note that, even at 12 kbar, the SC is not observed down to 1.2 K, instead the resistivity increases as $\log(T)$ with decreasing $T$ below about 5 K as shown in Figs. 1 and 2.

In the temperature region where the resistivity behaves as $dR/dT<0$, negative magnetoresistance (N-MR) was observed as shown in Fig.3. Further, as seen in Fig. 3, the magnitude of N-MR is strongly angular dependent. The magnetoresistance which is in a weakly localized state can be expressed as a sum of positive MR, due to Lorentz force, and negative MR, due to the break of the time reversal symmetry of electron interference by magnetic field. The angular dependence of MR (Fig. 4) can be fit to the

---

1 Corresponding author. Tel: +81-(0)96-6605-2509; fax: +81-(0)96-6600-2710; E-mail: muratak@sci.osaka-cu.ac.jp
experimental data using the $p_0$ value of $2.28 \times 10^{-6}$ ohm-cm, for the uniaxially strained (9 kb //a for example) sample of $\theta$-CsZn as shown in Fig. 5.

We summarize our result in the following way. At least we could successfully stabilize the metallic state by the uniaxial strain/a. Since the N-MR is observed in both $\theta$-CsZn and $\theta$-RbZn (not shown), we assume that the origin of N-MR is common to both salts. From previous reports, structural disorder seems to be present in $\theta$-RbZn, but not in $\theta$-CsZn. Therefore we discard this possibility of negative magnetoresistance caused by the structural disorder. It is reminded that in both materials, fluctuation in charge order is observed above $T_{\text{fl}}$ by X-ray or NMR. The fluctuation might be induced by frustration of two kinds charge order patterns (vertical and horizontal) due to balance of the nearest neighbor Coulomb interaction. The time scale of the fluctuation of charge order is slow when viewed by conduction electron. Therefore conduction electrons might see the frustration of charge order as random Coulomb potentials.

This work was supported by 'Research for Future Project', JSPS-RFTF97P00105, of Japan Society for the Promotion of Science.

References