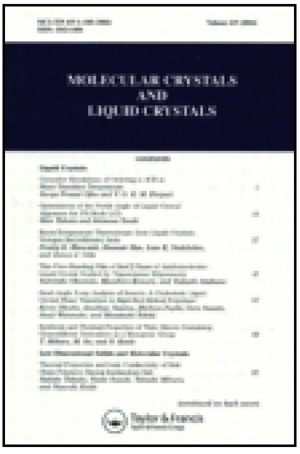

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Effects of Uniaxial Strain on α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, NH$_4$]

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The effects of uniaxial strain on α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, NH$_4$] were systematically studied in order to clarify the difference of the electronic properties between K-salt and NH$_4$-salt. Superconductivity like NH$_4$-salt was induced in K-salt by the *c*-axis uniaxial strain, while the density wave state like K-salt was induced in NH$_4$-salt by the *a*-axis uniaxial strain. The latter was confirmed by the angle dependent magnetoresistance measurement. It was found that the ratio of the in-plane lattice constant *c*/*a* plays a key role in dominating their electronic properties.

Keywords BEDT-TTF; Organic Superconductor; Uniaxial Strain; Transport Measurements

INTRODUCTION

A series of quasi-two dimensional organic conductors α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, Rb, Tl, NH_4] has attracted much attention, since they have afforded a wide variety of unique electronic properties. These compounds are isostructural and their lattice parameters are almost same. However, only NH_4-salt shows superconductivity below about 1K [1], while the others with M=K, Rb, Tl undergo a transition to density wave (DW) state at about 10 K [2]. In order to clarify what causes the difference in their electronic properties and to search for novel electronic states, we systematically studied the effects of uniaxial strain on the title compounds α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, NH_4] having the different ground state.

EXPERIMENTAL

The out-of-plane resistance of these compounds was measured down to 0.6 K under the uniaxial strain applied along the *a*-, *b**- and *c*-axes independently using four probe dc method. Details of the experimental technique of the uniaxial strain method have been described in our previous paper [3]. We used the oil (Demnum S-20) and the epoxy (Stycast 1266) as a pressure medium. The crystallographic axes of these compounds were identified by the X-ray diffraction measurement.

RESULTS AND DISCUSSION

The anisotropy of the material was directly controlled at low temperatures using the frozen oil method. We found the electronic properties of these compounds systematically changed in relation to the direction and magnitude of the in-plane uniaxial strain. Fig.1 depicts the temperature dependence of the resistance of the K-salt (a) and the NH_4-salt (b) under the in-plane uniaxial strain. The *c*-axis uniaxial strain suppressed the DW state and induced the superconductivity above

5kbar in the K-salt, whereas the superconductivity was strongly enhanced in the NH_4-salt. The superconducting transition temperature T_C of the latter compound was increased up to about 6K under 5 kbar of the external pressure, followed by the substantial decrease in the T_C by the further increasing of the pressure. It has been reported that among the series of these isostructural compounds only the superconducting NH_4-salt shows a significant decrease in the ratio of *c*/*a* below about 200 K [4]. We found, in accordance with the anisotropic thermal contraction, the reduction in *c* plays a dominant role in the appearance of the superconductivity.

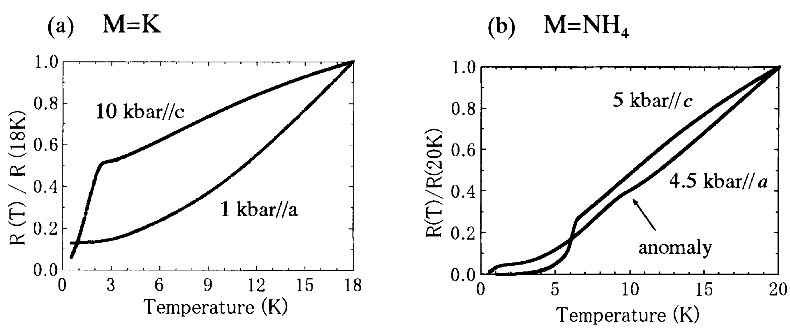

FIGURE 1. The temperature dependence of the out-of-plane resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ (a) and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ (b) under the uniaxial strain along the *a*- and *c*-axes. The external pressure was applied at 20K using the frozen oil method.

In contrast to the above results, the *a*-axis strain stabilized the normal metallic state. In the K-salt, the DW state was entirely suppressed by the low external pressure of 1kbar along the *a*-axis and the normal metallic behavior was observed. In case of the NH_4-salt, the *a*-axis strain suppressed the superconductivity and the normal metallic behavior was observed above 7 kbar. It should be noted that the step like anomaly in the resistance was observed at about 10 K in the intermediate pressure range (4-6kbar), reminiscent of the DW transition. In order to characterize this low temperature state, we performed the study of the angle dependent magnetoresistance oscillations (AMRO) of NH_4-salt under the *a*-axis uniaxial strain, where the epoxy-crystal method was employed. Fig. 2 (a) represents the AMRO pattern of NH_4-salt under the *a*-axis uniaxial strain. The magnetic field was rotated from the *b**-axis to the *a*-axis. Under the external pressure of 2.5 kbar,

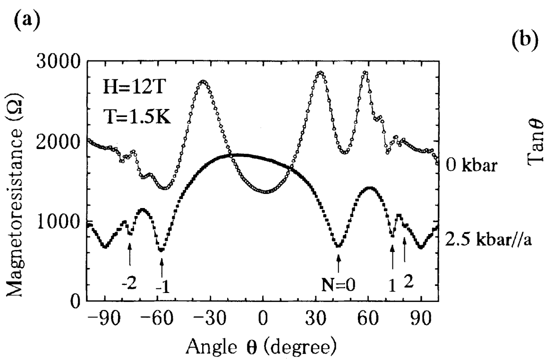

FIGURE 2. (a) Angle dependent magnetoresistance oscillation of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the *a*-axis uniaxial strain. The external pressure of 2.5 kbar, which correspond to about 5kbar by the frozen oil method, was applied at room temperature using the epoxy-crystal method. (b) The positions of the dips in the magnetoresistance under 2.5 kbar plotted against tanθ.

the anomaly in the temperature dependence of the resistance was observed around 10 K, below which the pattern of the AMRO drastically changed as shown in Fig. 2 (a), indicating the reconstruction of the Fermi surface. The positions of the dips in the magnetoresistance under 2.5 kbar were found to be periodic in tanθ without phase sift between θ>0 and θ<0 as shown in Fig. 2 (b). These features are characteristic of the AMRO due to the quasi-one dimensional open

Fermi surface. More detailed studies revealed that the topology of the Fermi surface in this low temperature state is essentially the same as-that of K salt in the DW state [2,5]. Therefore, this low temperature state is regarded as a DW state.

From these systematic studies, we obtained the unified phase diagram of α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, NH_4] as shown in Fig. 3. It was verified that both K-salt and NH_4-salt have essentially the same electronic properties, which can be systematically understood as a function of c/a. The reduction in c/a enhanced the superconductivity, while the increase in c/a stabilized the normal metallic state. The DW state appeared between them.

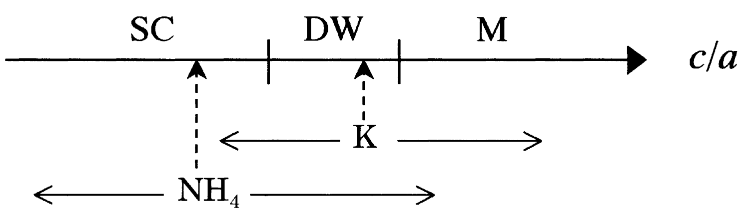

FIGURE 3. The schematic unified phase diagram of α-(BEDT-TTF)$_2$MHg(SCN)$_4$ [M=K, NH_4]. The superconducting state, the density wave state and the metallic state are abbreviated as SC, DW and M, respectively. The dashed arrows indicate the locations of the K- and NH_4-salts at ambient pressure.

The suppression of the DW state in the K-salt by the c-axis compression and the inducement of the DW state in the NH_4-salt by the a-axis compression can be ascribed to the changes in the nesting condition of the open Fermi surfaces running perpendicular to the a-axis. However, in order to account for the suppression of the DW state by the further a-axis compressions, one should consider other effects such as the reduction of the density-of-states.

It is still unclear why the reduction in c strongly enhances the superconductivity. The external force possibly causes not only the translations of molecule parallel to the force but also some changes in the molecular orientation. Either of these effects can cause dramatic changes in the electronic structure in these anisotropic materials. For more quantitative discussions, we are planning further investigations such as AMRO and the X-ray diffraction.

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