Metal Instability of (DMe-DCNQI)<sub>2</sub>Cu Induced by Uniaxial Stress and Enhancement of Electron Mass

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The metal instability of (DMe-DCNQI)<sub>2</sub>Cu was induced by applying uniaxial stress or coating the crystal with Apiezon L grease. The quadratic temperature dependence of the resistivity gave an indication of the large enhancement of the effective mass of the conduction electrons.

It has been pointed out that the DCNQI-Cu system is a new type of molecular metal with  $p_{\pi}$ -d mixing band (DCNQI= dicyanoquinonediimine). There are two types of the DCNQI-Cu systems  $^{1,2}$ : (1) compound with stable metallic state at ambient pressure ((DMe-DCNQI)2Cu, (DMeO-DCNQI)2Cu,...), (2) compound exhibiting a sharp metal-insulator(MI) transition ((MeBr-DCNQI)2Cu, (DBr-DCNQI)2Cu...).

It is well known that the metallic state of  $(DMe-DCNQI)_2Cu$  becomes unstable at the critical pressure  $(P_C)$  as small as 100 bar. We tried to reproduce the metal instability without use of the He gas pressure technique. In this paper, we report simple methods to induce the metal instability and the  $T^2$ -dependence of the low-temperature resistivity indicating the mass enhancement of the conduction electrons.

A needle crystal of (DMe-DCNQI) $_2$ Cu with four gold wires (15  $\mu m \phi$ ) bonded by Au paint was fixed on a small Teflon block by an adhesive agent and a small weight (W= 0-25 g) was put carefully on the top of the needle (Fig. 1). The typical dimensions of the crystals used are 2x0.02x0.02 mm $^3$ . The resistivity was measured by a four-probe method down to 4.2 K. The uniaxial pressure was estimated roughly as W/S, where S is the cross sectional area of the crystal. As seen from Fig. 1 (sample #8), the

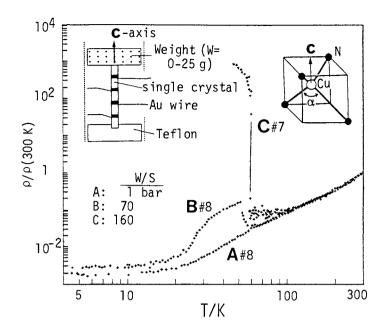


Fig. 1.
Temperature dependence of resistivity of (DMe-DCNQI)<sub>2</sub>Cu under uniaxial stress and schematic drawing of the setting of the measurement.

resistivity showed an anomaly around the uniaxial pressure of 70 bar. At 160 bar, a sharp MI transition was observed ( $T_{\rm MI} \simeq 60$  K). At first, the uniaxial pressure along [001] was supposed to be more effective to induce the metal instability than the hydrostatic pressure, because the increase of the bond angle N-Cu-N ( $\alpha$ ) (Fig. 1) was considered to be a key factor for the metal instability. But the results were in good agreement with those obtained by the He gas pressure technique. In order to investigate further the effect of non-hydrostatic pressure, another experiments was made.

As reported recently, an organic superconductor,  $\kappa$ -(ET)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Cl  $(T = 12.8 \text{ K at } 0.3 \text{ kbar})^5)$  becomes an "ambient-pressure superconductor" by coating the crystal with grease.  $^{6}$ ) In the case of the experiment on the  $\kappa$ type ET superconductor, the "effective pressure" produced by freezing of grease was estimated roughly to be about 0.2 kbar, 6) which is large enough to make (DMe-DCNQI)2Cu an insulator. The needle crystal was coated by Apiezon L grease as illustrated in Fig. 2. The freezed grease was compress the crystal mainly along perpendicular to [001]. The resistivity showed a small anomaly around 50 K, indicating that the crystal felt the effective pressure approximately equal to Pc.

The resistivity anomaly around 50 K presumably associated with the appearance of the magnetic ions (Cu<sup>2+</sup>) and the reentrance of the metallic state at low temperature remind us the resistivity behavior of "heavy Fermion system". <sup>7-9</sup>) So that, the low-temperature resistivities ( $\rho$ ) were analyzed according to the relation commonly observed in heavy (or semiheavy) Fermion compounds :  $\rho$ = AT<sup>2</sup>+  $\rho$ 0 and A/ $\gamma$ <sup>2</sup>=1.0 x 10<sup>-5</sup>  $\mu\Omega$ cm(mol K/mJ)<sup>2</sup>,

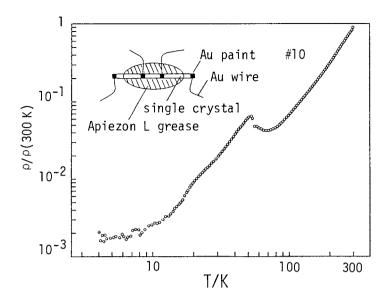
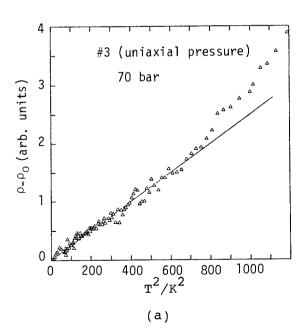


Fig.2.
Resistivity of a grease-coated crystal of (DMe-DCNQI)<sub>2</sub> Cu.

where  $\gamma$  is the linear term in the temperature dependence of low-temperature specific heat. The form the A-value obtained by the least-squares fitting of the T<sup>2</sup>-dependence of  $\rho$  shown by the straight lines in Fig. 3,  $\gamma$  was estimated as, 120 mJ mol<sup>-1</sup> K<sup>-2</sup> (uniaxial pressure (W/S) of about 70 bar), 110 mJ mol<sup>-1</sup> K<sup>-2</sup> (Apiezon L grease).

Considering that  $\gamma$  of  $(DMeO-DCNQI)_2$  Cu is 9 mJ mol<sup>-1</sup> K<sup>-2</sup> (1 bar), the obtained  $\gamma$  is anomalously large. Note that  $\gamma$  of heavy (or semi-heavy) Fermion system is 50-1000 mJ mol<sup>-1</sup> K<sup>-2</sup>. Enhanced  $\gamma$  was also suggested by the similar analyses on the resistivities of  $(DMe-DCNQI)_2$ Cu ( $\gamma$ = 40



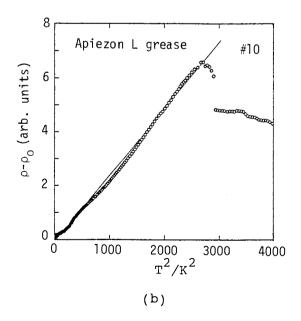


Fig. 3. The  $T^2$ -dependence of the resistivity of (DMe-DCNQI) $_2$ Cu. (a) Crystal under uniaxial stress (b) Grease-coated crystal

mJ mol<sup>-1</sup> K<sup>-2</sup>(1 bar)) and (DMeO-DCNQI)<sub>2</sub>Cu (45 mJ mol<sup>-1</sup> K<sup>-2</sup>(6 kbar), 200-400 mJ mol<sup>-1</sup> K<sup>-2</sup>(6.5 kbar)). The  $\gamma$ -value of (DMe-DCNQI)<sub>2</sub>Cu (= 40 mJ mol<sup>-1</sup> K<sup>-2</sup>) agrees well with that obtained from the specific heat measurements. 13,14) Extraordinarily large  $\gamma$  suggests that the electron becomes very heavy in the low-temperature metallic state around the critical condition where metal instability begins to appear. 9) Good agreement between  $\gamma$ -values determined from the resistivity data and those from specific heats in the alloyed system, [(DMe)<sub>1-x</sub>(MeBr)<sub>x</sub>-DCNQI]<sub>2</sub>Cu will be reported elsewhere. 12)

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