High-pressure study of a doped-type organic superconductor, $\kappa$-(BEDT-TTF)$_4$Hg$_{2.89}$Br$_8$

T. Okuhata$^a$, T. Nagai$^a$, H. Taniguchi$^a$, K. Satoh$^a$, T. Itou$^b$, Y. Shimizu$^b$, K. Miyagawa$^b$, Y. Ishii$^c$, N. Tajima$^c$, and R. Kato$^c$

$^a$Faculty of Science, Saitama University, Shimo-okubo 255, Saitama 338-8570, Japan
$^b$Department of Applied Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan
$^c$RIKEN, JST-CREST, Wako, Saitama 351-0198, Japan

Physical properties of the organic charge-transfer complex that may be regarded as a doping system are presented. We measured resistivity of an ambient-pressure superconductor, $\kappa$-(BEDT-TTF)$_4$Hg$_{2.89}$Br$_8$, as functions of temperature, magnetic field, and pressure. Metallic and superconducting states of this salt are possibly attributable to the doping effect, which originates from incommensurability of Hg chain because band structure calculation predicts strong electron-electron correlation enough to localize the itinerant electrons. We uncovered anomalous pressure and magnetic field dependences of superconductivity as well as non-Fermi liquid behavior in the normal-state resistivity at low pressures. In addition, we observed a pressure-induced crossover to the Fermi liquid behavior, which is seen in non-doped $\kappa$-type salts at any pressures.

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1. INTRODUCTION

It is now widely accepted that the organic salts, $\kappa$-(BEDT-TTF)$_2$X (X=Cu(NCS)$_2$ and Cu[N(CN)$_2$]Br etc.), can be regarded as bandwidth-controlled superconductors at fixed band filling$^1$. In other words, the superconductivity in the $\kappa$-type salt is realized by pressurizing the Mott insulator. In this case, with increasing pressure, the electrons localized at dimers of BEDT-TTF experience the first-order insulator-metal transition$^2$ and show Fermi liquid behavior$^3$ just as they start to delocalize.

The (BEDT-TTF)$_4$Hg$_{2.89}$Br$_8$ is also known to crystallize into the $\kappa$-type structure$^4$. However, there is a structural characteristic not seen in other $\kappa$-type salts. This salt has incommensurate structure between lattice periodicity of mercury chains and that of halogens and donor molecules$^4$. 

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Due to this incommensurability, the electron filling is possibly shifted from the usual half-filled state of 2:1 salts, and therefore kind of hole-doped state may be realized. Indeed this salt shows metallic temperature variation and superconductivity at 4.3K\(^4\) in spite of highly enhanced U/W value (U/W=1.79)\(^5\). In order to clarify the difference in physical properties between doped and non-doped organic superconductors, we measured resistivity of \(\kappa\)-(BEDT-TTF)\(_2\)Cu[N(CN)\(_2\)]Br and \(\kappa\)-(BEDT-TTF)\(_4\)Hg\(_{2.89}\)Br\(_8\) as functions of temperature, pressure and magnetic field.

2. EXPERIMENTAL

Single crystals of \(\kappa\)-(BEDT-TTF)\(_2\)Cu[N(CN)\(_2\)]Br and \(\kappa\)-(BEDT-TTF)\(_4\)Hg\(_{2.89}\)Br\(_8\) were prepared by the electrochemical oxidation of BEDT-TTF in 1,1,2-trichloroethane in the presence of corresponding electrolyte\(^6,7\). The electrical resistivity measurements were performed with the standard four-probe method with dc current parallel to conducting layers under ambient and high pressures. In the magnetoresistivity measurement magnetic field and the current were applied perpendicular to the conducting layers.

3. RESULTS AND DISCUSSION

The electrical resistance of the Hg\(_{2.89}\)Br\(_8\) salt after subtracting residual resistance is plotted in a left panel in Fig. 1 as a function of temperature in the logarithmic scales. For comparison, the data of the Cu[N(CN)\(_2\)]Br salt are shown in Fig.2. We found that all the data were well fitted by \(R=R_0+T^n\) in the low-temperature region just before superconducting transition. Obtained \(n\) as well as onset-\(T_c\) are plotted as a function of pressure in the right panels. Although \(T_c\) of the Cu[N(CN)\(_2\)]Br salt strongly depends on pressure, the power, \(n\), is stable around 2. This value of the power is consistent with so-called Fermi liquid behavior with electron correlation. On the other hand, the power of the Hg\(_{2.89}\)Br\(_8\) salt shows the distinct variation from 1 to 2 with increasing pressure. The power of 1 in the low-pressure range is reminiscent of anomalous metallic state of underdoped cuprate or low-pressure range of CeCoIn\(_5\)\(^8\). Observed variation of the power from 1 to 2 may reflect that the system undergoes the crossover to the Fermi liquid behavior.

As shown in Fig.3, we also measured the resistivity of the Hg\(_{2.89}\)Br\(_8\) salt as a function of magnetic field perpendicular to the conducting layers at fixed temperatures under several pressures. Then we found that the superconductivity at low pressures, where the normal state showed the non-Fermi liquid behavior, had very large critical field, larger than the Pauli limit field. This behavior is quite unusual because other \(\kappa\)-type superconductors
High-pressure study of a doped-type organic superconductor

Fig. 1. (Left Panel) Temperature dependence of resistivity in the logarithmic scales for the Hg$_2$.89Br$_8$ salt at different pressures. (Right Panel) Pressure dependence of the power, $n$, and onset-$T_c$.

Fig. 2. (Left Panel) Temperature dependence of resistivity in the logarithmic scales for the Cu[N(CN)$_2$]Br salt at different pressures. (Right Panel) Pressure dependence of the power, $n$, and onset-$T_c$.

Fig. 3. Magnetic-field dependence of out-of-plane resistivity of the Hg$_2$.89Br$_8$ salt.
T. Okuhata et al.

never exceed the Pauli limit in this magnetic-field configuration.

4. CONCLUSIONS

In summary, we have demonstrated the comparative study of transport properties between doped and nondoped organic superconductors. While the nondoped system shows Fermi liquid behavior in the whole pressure-range that we measured, the doped one shows the strong violation from the Fermi liquid picture at low pressures and the crossover to Fermi liquid behavior with increasing pressure. The normal state behavior with $n \sim 1$, the positive $dT_c/dP$, and the large perpendicular $H_{c2}$ that exceeds Pauli limit are the characteristics of the present doped-type organics under low pressures, which is not observed in other organics but seen in underdoped cuprates and CeCoIn$_5$.

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