Phase diagram of partially deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

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Abstract

The phase diagram of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br around the metal-insulator (MI) boundary controlled by partially deuteration and by cooling rate through the 80 K anomaly has been investigated by resistivity measurements under magnetic fields. According to approach to the critical region of MI transition from the metallic side by the increase of cooling rate, a) in addition to the deuteration and the cooling rate dependence of the resistance maximum, the hump of resistance, observed at 33.5 K in the partially deuterated sample for slowly cooled, is shifted towards a lower temperature, b) the temperature of the resistance hump at 9.5 K with the hysteresis of resistance increases. Our results suggest that these anomalies are related to the phase separation of the metallic and insulator phase around the MI boundary.

Key words: organic crystals; electric transport; κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br; metal-insulator transition

The quasi-two-dimensional organic compounds κ-(BEDT-TTF)$_2X$, where X=Cu[N(CN)$_2$]Cl, Cu[N(CN)$_2$]Br, Cu(NCS)$_2$, etc., show many interesting phenomena such as d-wave superconductivity (SC), antiferromagnetic (AF) ordered state, anomalous metallic state at low temperatures. A universal phase diagram for the κ-BEDT-TTF salts as function of U/W, where U is the on-site Coulomb interaction and W is the bandwidth, has been proposed. [1] In the metallic phase for the κ-BEDT-TTF salts, a cusp like enhancement of the spin-lattice-relaxation rate $(T_1T)^{-1}$ of ^{13}C-NMR, [2] a large softening of the ultrasound modes [3] and a local maximum of the thermal expansion coefficient [4] have been observed at about $T^* = 40$ K for the X = Cu[N(CN)$_2$]Br salt and at about $T^* = 50$ K for the X =Cu(NCS)$_2$ salt. At ambient pressure the hydrogened Cu[N(CN)$_2$]Br salt undergoes a metal-superconducting transition at 12 K. On the other hand, the deuterated Cu[N(CN)$_2$]Br salt undergoes a coexistence phase of AF and SC, where the first-order phase transition is suggested. [5]

In this paper, we describe the phase diagram of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br around the metal-insulator (MI) boundary controlled by cooling rate through the 80 K anomaly. We discuss the role of the cooling rate, and that of the deuteration in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br phase diagram.

Single crystals of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br were synthesized by the standard electrochemical method. The BEDT-TTF molecule has two ethylene groups, each of which contains four protons. We use the notation of d[n,n'] to present the number of the deuterium in each ethylene group. The resistance measurements along the conducting plane were carried out using a standard four-probe dc method.

It is well known that the resistivity of κ-(BEDT-TTF-d[0,0])$_2$Cu[N(CN)$_2$]Br shows a maximum around 100 K at ambient pressure and this maximum temperature decreases by the progressive deuteration and by the increase of cooling rate. [6] To avoid the effect of SC, resistivity measurements have been performed under magnetic field at 16 T perpendicular to the conducting plane. Figure 1 shows the temperature dependence of the transverse magnetoresistance

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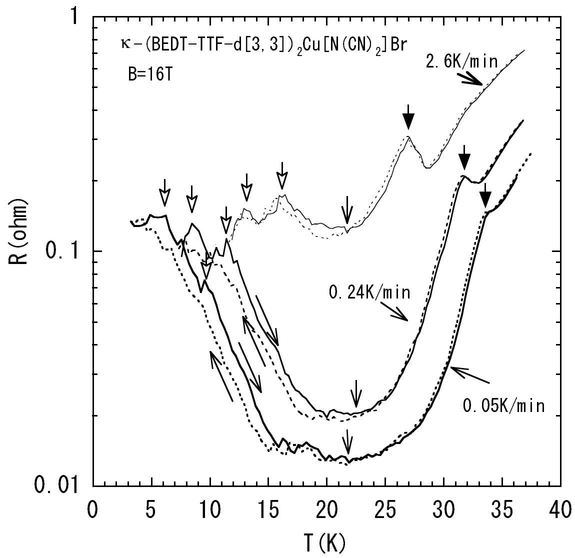

Fig. 1. Temperature dependence of the transverse magnetoresistance in κ-(BEDT-TTF-d[3,3])$_2$Cu[N(CN)$_2$]Br under magnetic field at 16 T perpendicular to the conducting plane for various cooling rates. The closed, bold and open downward arrows indicate the hump1 corresponding to the T^* anomaly, the starting temperature of the hysteresis and the hump 2,3 with the hysteresis, respectively.

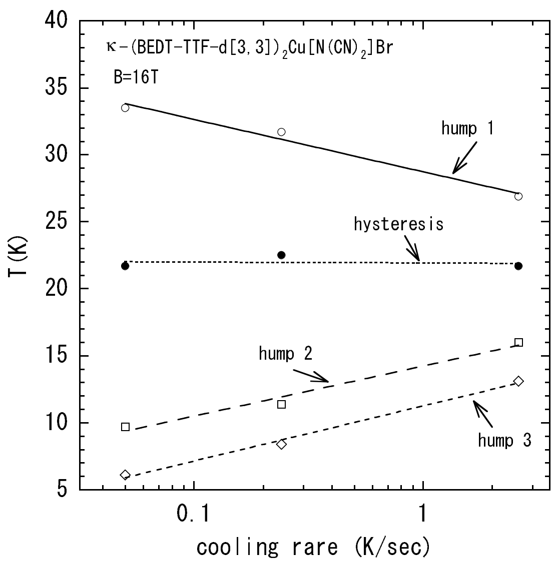

Fig. 2. Temperature of the hump 1, hump 2, hump 3 and the start of hysteresis in the resistance of κ-(BEDT-TTF-d[3,3])$_2$Cu[N(CN)$_2$]Br as a function of cooling rate.

in κ-(BEDT-TTF-d[3,3])$_2$Cu[N(CN)$_2$]Br, in which the cooling rates through the 80 K anomaly are 0.05, 0.24, and 2.6 K/min. With decreasing temperature from 40 K, the resistance decreases and has a small hump 1 at 33.5 K. With increasing cooling rate, this small hump 1 becomes large and the temperature of the hump 1 decreases. We expect that the hump 1 around 30 K and the T^* anomaly cause by the same origin, because the cooling rate and the deuteration dependence of the hump seem to smoothly connected to the T^* anomaly in the universal phase diagram for the κ-BEDT-TTF salts. The resistance shows hysteresis below around 20 K, irrespective of the cooling rate. With decreasing temperature, the resistance shows the rapid increase at 15 K for 0.05 K/min and 18 K for 0.24 K/min and almost saturate with the hump 2 and the hump 3 at low temperature. With increasing cooling rate, the temperature of the hump 2 and the hump 3 increases. The origin of the hump 2 and the hump 3 is unsolved. The cooling rate dependence of the hump 1 2 3 and the starting temperature of the hysteresis are summarized in Fig. 2. It is reasonable to consider that a) the resistance hysteresis corresponds to the first-order phase transition due to the phase separation of AF and the metallic state [5], b) the rapid increase of the resistance corresponds to the rapid decrease of the metallic part in the sample, and c) the saturation of the resistance correspond to one of the decrease of the metallic part. These results are consistent with the ^{13}C-NMR experimental results.

In conclusion, we have measured the magnetoresistance in the partially deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br for various cooling rates through the 80 K anomaly. We have found that, according to approach to the critical region of MI transition from the metallic side by the increase of cooling rate, a) the hump of resistance corresponding to the T^* anomaly is shifted towards a lower temperature, b) the temperature of the hump at 9.5 K with the hysteresis, which appears below around 20 K, increases. These results suggest that these anomalies are related to the phase separation of the metallic and insulator phase around the MI boundary.

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