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Mixed density wave state in quasi-2D organic conductor

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ABSTRACT

The density wave phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ was investigated by transport properties and magnetic susceptibility. The density wave transition was observed as a broad increase at $T_{DW}=9$ K by resistance measurement. Temperature dependence of the static magnetic susceptibility χ shows a large Curie tail below 100 K. By subtracting the Curie component, we found that the magnetic susceptibility increases like weak ferromagnetism with decreasing temperature below 7.4 K. The gradual increase of χ below T_{DW} is not expected in simple CDW or SDW, where the magnetic susceptibility decreases with decreasing temperature due to the reduction of Pauli paramagnetic component. To explain the weak ferromagnetic behavior, we consider the coexistence of CDW and SDW. We propose a model of the mixed density wave, where CDW exists with antiferromagnetically coupled canting spins.

1. Introduction

Density waves are known as the ground state in low-dimensional conductors. Density waves are classified into two types by the interaction. One is the spin density wave (SDW) due to the on-site Coulomb interaction. The other is charge density wave (CDW) due to the electron–phonon interaction. One can easily conceive the material, in which the electron–phonon and on-site Coulomb interactions are both comparable. It seems that the mixed density wave state of CDW and SDW can be realized as one of the ground state in low dimensional conductors. Actually, the coexistence of CDW and SDW has been reported in (TMTSF)$_2$PF$_6$ by X-ray diffraction [1]. The coexistence is stabilized only in the temperature range of $0.3T_{SDW}<T<T_{SDW}$ [2], where T_{SDW} is SDW transition temperature.

As a candidate for the material which has the coexistence of CDW and SDW for the ground state, then we focus on an organic conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$. α-(BEDT-TTF)$_2$KHg(SCN)$_4$ has two types of Fermi surfaces; a pair of one-dimensional sheet Fermi surfaces and two-dimensional columnar Fermi surfaces located at corners of the Brillouin zone [3]. The density wave occurs resulting from the nesting of the sheet Fermi surface. The density wave transition temperature T_{DW} was reported as $T_{DW}=8$ K [4]. As a character of the density wave of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, Sasaki et al. reported SDW from the anisotropic magnetic susceptibility below T_{DW} [5]. On the other hand, NMR measurement [6] reported no magnetic order, which suggested CDW. There is no clear evidence for the ground state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. We think this unclear situation comes from the coexistence of CDW and SDW.

In this paper, we report an anomaly of magnetic susceptibility which cannot be explained by simple CDW or SDW. Then we propose a model of the mixture of CDW and SDW. Preliminary result for scanning tunneling microscopy (STM) measurement at room temperature is also presented.

2. Experimental

Single crystals of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ were grown by electro-oxidation method with 1 μA for a month. Regents for synthesizing single crystals are K(SCN), Hg(SCN)$_2$ and 18-crown-6 ether. These regents were purified by recrystallization. 1-1-2 trichloroethane and ethanol was used as solvent. The typical shape of the single crystals is thin plate like along the $a-c$ plane. Crystal orientation was determined by the X-ray diffraction. Typical dimension of crystals was 1 mm × 0.5 mm × 0.1 mm.

The DC resistance was measured with the four probe configuration. Magnetic susceptibility was measured by SQUID magnetometer applying the magnetic field up to 1 T. Several crystals were attached to the sample holder.

For STM measurement, single crystals were flushed with 1-1-2 trichloroethane to obtain a fresh surface just before the measurement. A mechanically sharpened Pt–Ir wire was used as the STM tip.

3. Results and discussion

Fig. 1 shows temperature dependence of resistance from 4 K to 300 K at the conducting $a-c$ plane. The resistivity obeys T^2 law

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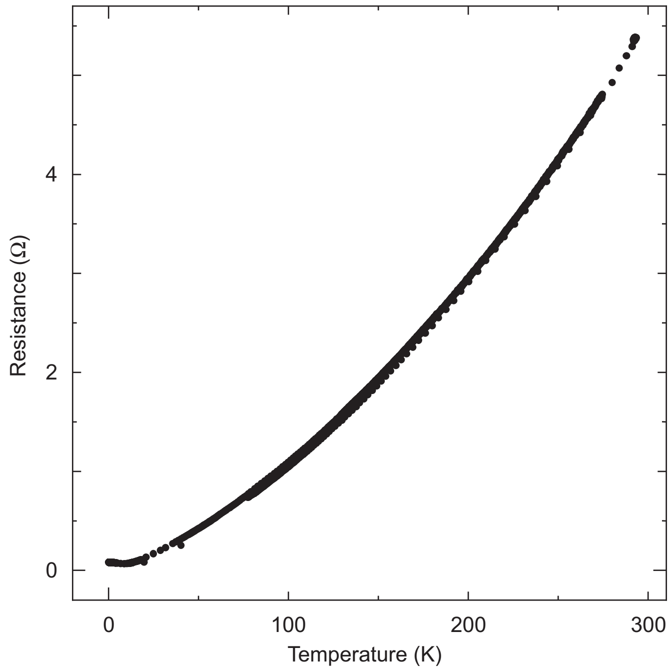

Fig. 1. Temperature dependence of resistance from 4 K to 300 K.

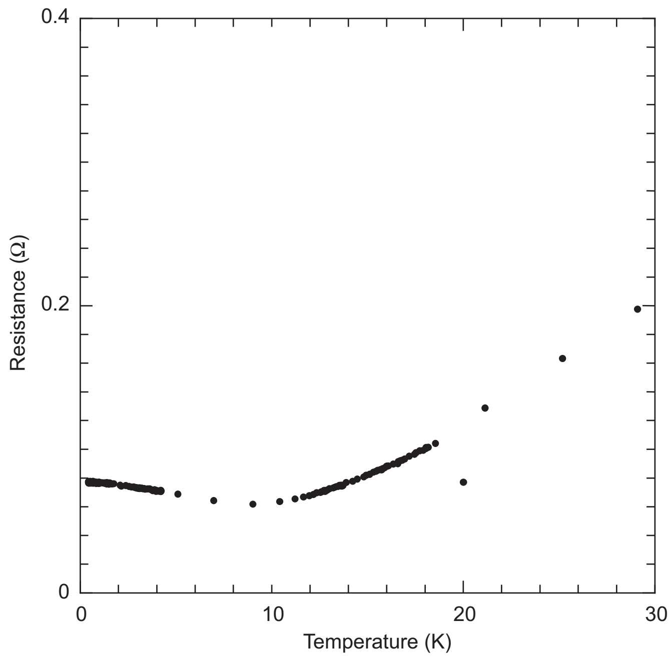

Fig. 2. Temperature dependence of resistance from 1 K to 30 K.

with T-linear component. It suggest that both of electron correlation and electron–phonon interaction influence the electronic state. Fig. 2 shows temperature dependence of resistance at low temperature region. Below about 10 K resistance increases with decreasing temperature due to the density wave transition. We determine the density wave transition temperature T_{DW} as $T_{DW} = 9$ K from the minimum of the resistance. Anomaly at T_{DW} was reported as a shoulder [4]. In the present study the density wave transition was observed as the increase of resistance. Residual resistance in our sample is slightly larger than that in previous report [4]. It suggests that the present sample contains a small amount of impurity which causes the Curie paramagnetism as discussed below. However, the impurity effect is too restrictive to suppress the density wave transition.

Fig. 3 shows temperature dependence of magnetic susceptibility applying the magnetic field of 1 T. Overall magnetic susceptibility χ is negative due to the molecular diamagnetism of BEDT-TTF. The magnetic susceptibility χ is almost constant in high temperature region. Below 100 K, χ increases with decreasing temperature. The temperature dependence shows the Curie paramagnetic behavior due to some impurity as discussed above. In order to estimate the Curie component, we plot $1/\chi$ versus T as shown in Fig. 4. Before the analysis we subtracted the diamagnetic component, χ_{dia}, which is

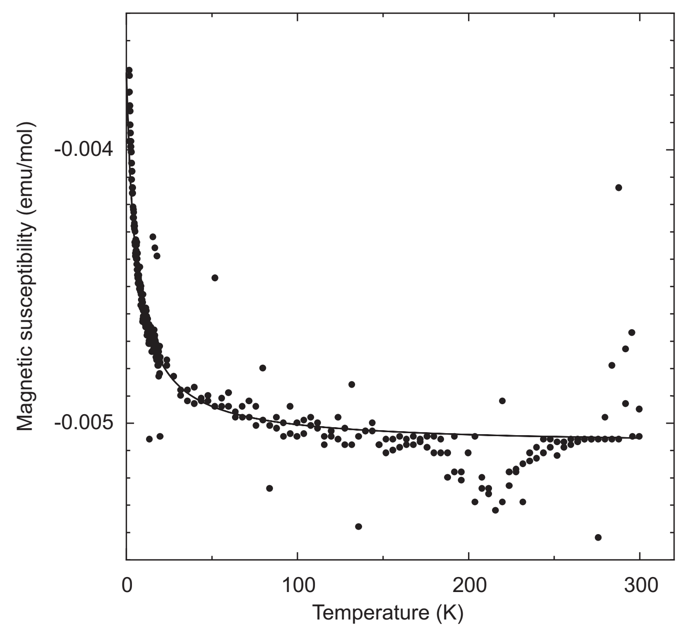

Fig. 3. Temperature dependence of magnetic susceptibility. Solid line represents the Curie–Weiss law.

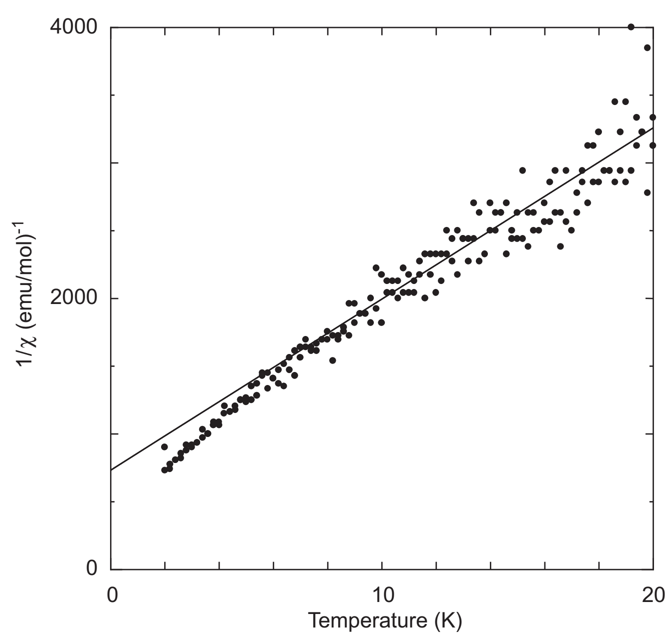

Fig. 4. Inverse of magnetic susceptibility versus temperature. Diamagnetic component was subtracted before obtaining $1/\chi$. The solid line represents the Curie–Weiss law.