Fermi surface and resistance anomalies in ET-TCNQ

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Abstract:
Recently, a new organic conductor ET-TCNQ, which exhibits two resistance anomalies at $T_1 = 80$ K and $T_2 = 20$ K, was synthesized. We present a systematic study of transport properties of ET-TCNQ under magnetic fields. Shubnikov-de Haas (SdH) oscillations are clearly observed and we find five distinct small Fermi surfaces (FS's). The cross-sectional areas of FS are estimated to be from 1$\%$ to 3.5$\%$ of the first Brillouin zone. We detect Lebed resonance and Danner-Chaikin oscillation in measurements of angular-dependent magnetoresistance oscillation (AMRO), suggesting the existence of the quasi-one dimensional FS related to ET molecules. The origins of the two resistance anomalies are discussed in terms of nesting-driven density-wave transition.

Keywords: Metal-insulator transition, Structural phase transition

1. Introduction
Recently, Yamamoto et al.\cite{1} synthesized a new organic conductor ET-TCNQ [bis(ethylenedithio)tetrathiafulvalene tetracyanoquinodimethane]. The band-structure calculation suggests that the dominant directions in overlap integral for ET molecules and TCNQ ones are $a$-axis and $c$-axis, respectively. ET-TCNQ possesses Fermi surfaces with rectangular cross sections. The temperature dependence of the resistance exhibits metallic behavior with two resistance anomalies, one at $T_1 = 80$ K and the other at $T_2 = 20$ K. One naively expects that two resistance anomalies are associated with partial nesting of Fermi surface. To investigate relationship between the two resistance anomalies and FS, we have measured Shubnikov-de Haas (SdH) and angular-dependent magnetoresistance oscillations (AMROs).

2. Experimental
To obtain single crystals of ET-TCNQ, a CH$_3$Cl(or CH$_3$Br) solution of ET, TCNQ, and TIE(=tetradiamidobutylenene) was placed at room temperature, and the solvent was allowed to evaporate slowly to dryness within 24 hours. The residual resistance ratio $R(300$ K)/$R(1.8$ K) of the single crystals are 3-10. The resistance was measured by a conventional four-probe technique with electric current along the $b$-axis (normal to the 2D plane). Four gold wires (10$\mu$m) were attached to the sample by silver paint. The experiments were made with a $^3$He cryostat with 14 T superconducting magnet (National Institute for Materials Science) and with a $^3$He cryostat with 33 T resistive magnet (National High Magnetic Field Laboratory, Florida State University).

3. Results and Discussion
Figure 1 shows AMRO for the magnetic field tilted in the $b$-c plane under various temperatures. At 1.8 K, the Lebed structure is clearly observed, suggesting the presence of FS from the ET origin. We can see that the Lebed structure is reduced with increasing temperature and finally the rotation curve assumes the simplest sine behavior. However, dip positions do not hold the usual resonant condition\cite{2} $\tan \theta \sim (p/q)(c/b^*)$, instead but $\tan \theta \sim (p/q)(a/b^*)$, where $p$ and $q$ are integer. The origin is unclear. On the other hand, we detect no Lebed structure for magnetic field tilted in the $b^*$-a plane, suggesting the absence of TCNQ's FS. Therefore, the origin of resistance anomaly at $T_2$, may be attributable to imperfect nesting of TCNQ's FS. However, the origin of $T_1$ anomaly is still unclear.

Figure 2(a) shows the magnetoresistance of ET-TCNQ as a function of magnetic field in the range 0 T -32 T at 0.63 K. Fourier transform spectrum, we find five distinct SdH oscillations as seen in Fig. 2(b).
The areas of FS are very small. For example, the area of α orbit is estimated to be 1% of the first Brillouin zone. Band-structure calculation does not predict such small Fermi surfaces. Therefore, these pockets may be created by an imperfect nesting of FS.

Figure 3 shows resistance as a function of temperature under zero magnetic field. The transition at \( T_2 \) is hysteretic, and therefore first order. It is likely that structural phase transition such as charge-density-wave transition with lattice distortion occurs.

We consider that the origin of the resistance anomalies at \( T_2 \) is attributable to the charge-density-wave transition associated with the TCNQ's FS. Therefore, the change of periodic potential in the \( c \)-axis direction will be caused by nesting of the FS associated with TCNQ molecules because of the weak interaction between ET molecules and TCNQ ones. The origin of the dip positions in the Lebed structure for the magnetic field tilted in the \( b^* - c \) plane may be related to the lattice periodicity parallel to the \( b^* - a \) plane such as \( \tan \theta \sim (p/q)(a/b)^* \).

4. Summary

We have measured the SdH and AMROs for new organic conductor ET-TCNQ. We find small FS pockets and quasi-one dimensional FS related to ET molecules. The resistance anomaly at \( T_2 \) may be attributable to the nesting of TCNQ Fermi surface. On the other hand, the origin of \( T_1 \) anomaly is still unclear.

In the Lebed structure for the magnetic field tilted in the \( b^* - c \) plane, we find that the dip positions do not hold the usual resonant condition \( \tan \theta \sim (p/q)(c/b)^* \), instead but \( \tan \theta \sim (p/q)(a/b)^* \), suggesting the importance of the weak interaction between ET molecules and TCNQ ones. To confirm whether charge-density-wave phase exists or not, \( x \)-ray scattering study at low temperature is required.

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References
