Possible quantum Hall effect in a magnetic-field-induced phase transition in the quasi-one-dimensional CDW organic conductor, HMTSF–TCNQ

Keizo Murataa,*, Yuhei Fukumotoa, Keiichi Yokogawaa, Woun Kangb, Ryo Takaokaa, Ryota Tadaa, H. Hirayamaa, James S. Brooks c, David Grafc, Harukazu Yoshinoa, Takahiko Sasaki d, Reizo Kato e

a Graduate School of Science Osaka City University, Sumiyoshi-ku, Osaka 558-8585, Japan
b Ewha Womans University, 11-1, Daehyun-Dong, Seoul 120-750, South Korea
c National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32306-4005, USA
d Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan
e Riken, 2-1, Hirosawa, Wako, Saitama 351-0198, Japan

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A B S T R A C T
We have studied the angular dependence of magnetoresistance and Hall effect of the CDW organic conductor, HMTSF–TCNQ in order to see whether a magnetic-field-induced phase exists in the charge density wave (CDW) system, similarly to the magnetic-field-induced SDW phases in (TMTSF)2X. The anomaly in magnetoresistance was observed only around the pressure where the CDW is almost suppressed, i.e. around 0.8–1.1 GPa, but neither at low pressures (0 and 0.5 GPa) nor at high pressure above 2 GPa. This behavior is quite similar to that of (TMTSF)2X. At 1.1 GPa anomalies were found at fields of 0.2 T and 10 T. We speculate that at 1.1 GPa the field-induced phase is located between 0.2 T and 10 T, where 1D Fermi surface sheet and 2D Fermi-surface pocket are present. The Rxy shows plateau structure and Rxx was very small in the same region, suggestive of quantum Hall effect.

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1. Introduction

The first organic superconductors, (TMTSF)2X are remarkable not only because of the first superconductivity among the organic conductors [1] but also of the insulating state with spin density wave (SDW), which was rare at that time except in chromium. More attracted was the presence of so-called field-induced SDW, which appears by applying magnetic field in the metallic phase in the vicinity of the ambient pressure SDW. Further, this material drew attention by the fact that the FISDW states were accompanied by a new type of quantum Hall effect (QHE) [2] quite different from the conventional QHE seen in GaAlAs.

It is of great interests to find whether or not the magnetic-field-induced phase exists in the charge density wave (CDW) system and whether or not it is similar to FISDW if any. A FICDW were discussed previously on α-(BEDT-TTF)3KHg(SCN)4 [3,4] by Andres et al., but the system was a mixture of 1D and 2D and anomaly in magnetoresistance was not so distinct.

To achieve a combined high-field experiment with high pressure, we tried to find a suitable CDW system whose CDW is suppressed by a moderate pressure below 1.5 GPa. Fortunately, the CDW of HMTSF–TCNQ is reported to be suppressed by around 1 GPa [5] in 1970s, and we confirmed the same value of pressure [6]. The material, HMTSF–TCNQ is hexamethylene-traselenafulvalene-tetracyanoquino-dimethane. To examine precisely the possible FICDW and quantum Hall effect, we studied magneto-resistance, angular dependence of magnetoresistance oscillations (AMRO) and Hall effect [6,7].

The organic conductor, HMTSF–TCNQ is a charge transfer salt with charge transfer ratio of 0.74, which is verified by X-ray [8]. The stacking pattern of HMTSF donor molecule and TCNQ acceptor molecules in the crystal shown in Fig. 1 is not the same as that of TTF–TCNQ. In other words, in contrast to the case of TTF–TCNQ, where TTF stacks, as well as TCNQ stacks, are aligned to construct a sheet, in the case of HMTSF–TCNQ, HMTSF stack is surrounded by TCNQ stack like checker board [9]. However, the Fermi surface is proposed to be constructed with a pair (+kF and −kF) of 1-dimensional sheets of electrons and another pair of holes [10], in the same way as TTF–TCNQ. A new band calculation is currently on the way.

It is of worth mentioning that the high quality crystals of...
HMTSF–TCNQ were difficult to obtain i.e. most of the samples consisted of many small, oriented crystallites mimicking a single crystal morphology. Such crystals had broad Bragg spots, and the signature of the CDW transition in the resistivity was neither sharp nor large. More recently, one (R.K.) of the authors succeeded in synthesizing high-quality single crystals; and our understanding of the field-induced phase has drastically advanced or even renewed as a result [7]. This paper presents the state of art on the magnetic-field-induced phase in HMTSF–TCNQ using the most refined samples.

2. Experiment

The crystals of HMTSF–TCNQ were obtained by a diffusion method at a temperature of 40 °C which resulted in an improvement of the crystal quality. All the samples used for this transport study were examined by X-ray, confirming sharp Bragg spots. According to Phillips 3, the crystal is monoclinic, though nearly orthorhombic; \( a = 21.999(14), \ b = 12.573(8), \ c = 3.890(1) \) Å; \( \beta = 90.29(4) \). Since previously crystal axes have been assigned in different ways [9,10], to avoid confusion, we adopt the Phillips’s convention.

The sample sizes were typically 0.87 × 0.14 × 0.06 mm³ and 0.92 × 0.26 × 0.13 mm³. Electrical contacts were attached by carbon paste and gold wires 10 or 20 μm in diameter sometimes with and sometimes without gold deposit on samples. Two samples of HMTSF–TCNQ, along with Sn and manganin pressure monitors were mounted together in a miniature pressure clamp cell, which allows rotation in the cryostat in a high magnetic field. The pressure medium was Daphne 7373 [11] and pressure at low temperature was determined with the superconducting transition of Sn. Most of the AMRO experiments reported here were performed at 1.1 GPa. Studies in high magnetic field were carried out with the clamp type pressure cell, with samples immersed in the pressure medium Daphne 7373. Four terminal methods were used with ac current from 10 μA to 100 μA. Pressure at low temperature for later works was calibrated against the superconducting transition temperature.

3. T–P phase diagram

In the temperature–pressure phase diagram, the quasi-one-dimensional conductor, HMTSF–TCNQ, the ground state at ambient pressure is an insulator of charge density wave (CDW) below 30 K, while it shows a good metallic nature at higher temperature. The CDW insulating state is almost suppressed by a pressure of 1 GPa [6].

4. Results and discussions of magnetoresistance at 1.1 GPa

The result of the 1.1 GPa measurements at low temperature is the following. Most of the studies were with \( R_{zz} \)-measurements configuration. Fixing the temperature (0.4 K for instance) and magnetic field (2 T, 4 T, through 31 T), we rotated magnetic field in the \( xy \)-plane, which is called Lebed–Osada oscillations-configuration in (TMTSF)₂X salts. It turned out that there were two regions separated at different border of magnetic field as shown in Fig. 2. The two regions were extremely high magnetoresistance (HMR) region and low resistance (LMR) one. Further it was found that the field-value that separates two regions is ruled by \( z \)-component of magnetic field, which was 10 T.

Next, we rotated the magnetic field in the \( xz \)-plane keeping other conditions the same. This rotation is called Danner–Kang–Chaikin configuration in (TMTSF)₂X salts. Again we obtained the same results. That is to say, there were two regions separated at different borders of magnetic field. And it was found that the field-value that separates two regions is ruled by \( z \)-component of magnetic field, which was 10 T.

Summarizing the above two rotations, \( B_z = 10 \) T suggests the existence of 2D straw-shaped Fermi surface below 10 T. By taking the magnetoresistance \( R_{zz} \) up to \( B_z = 45 \) T, no structure in \( R_{zz} \) was observed, but with continuous increasing of \( R_{zz} \). Then we concluded that 10 T is the Landau quantum limit. The lowest Landau level of the straw Fermi surface runs over the Fermi level resulting in an insulator. And we concluded that the HMR must be corresponding to this insulating state.

Coming back the Lebed–Osada configuration, there are rapid oscillations on the large background which show HMR and LMR in \( R_{zz} \) against the angles. The rapid oscillations are very clear in the HMR state and less clear but present in the LMR state. We could relate these oscillations Lebed–Osada oscillations themselves. It means in both HMR and LMR states there is a one-dimensional (1D) Fermi-surface sheet or pair of Fermi-surface sheets, which was found by the fine structure of the angular dependence of magnetoresistance oscillations Fig. 3.

Again summarizing up to now, in the LMR state below \( B_z = 10 \) T, there coexist 2D-Fermi surface straw and 1D-Fermi surface sheet, and in the HMR state above 10 T only 1D-Fermi surface sheet is present.

We should note finally that a careful view of magnetoresistance
5. Results and discussions of Hall effect at 1.1 GPa

By applying magnetic field of $B_z$, we studied $R_{xy}$ and $R_{xx}$. Between 0.2 T and 10 T, $R_{xy}$ is very flat and $R_{xx}$ is very low, which is consistent with quantum Hall effect. Below 0.2 T, $R_{xy}$ is negative and proportional to $B_z$. The fact that $R_{xy}$ is proportional to $B_z$ shows that a field-induced state is not present at least below 0.2 T. These data are published elsewhere [7].

The features of the constant $R_{xx}$ between 0.2 and 10 T without SdH oscillations in $B_z$ favor quantum Hall effect. But the two features (i) the only one plateau in $R_{xy}$ and (ii) the absolute value as a sheet resistance for $R_{xy}$ do not explain in a clear way the quantum Hall effect. The coexistence of 1D and 2D Fermi surfaces may smear to some extent.

6. Results and discussions at other pressures than 1.1 GPa

We studied the anomalous magnetoresistance $R_{zz}$ in magnetic field of $B_z$ at lower pressures than 1.1 GPa. At about 0.85 GPa, the field of transition which appeared at 10 T for 1.1 GPa moved down to 5 T. Further at about 0.8 GPa, the field of transition moved down to 2 T. We note that these lower pressure is the region where ambient pressure CDW is still present to some extent. These ideas are shown in Fig. 4.

7. Conclusion

Since it is clarified that the anomaly in magnetoresistance is not seen neither below 0.5 GPa nor above 2 GPa, we conclude that we have observed a magnetic-field-induced state in HMTSF–TCNQ. The quantum Hall effect seems to be present but the number of plateau is only one which is not similar to that of (TMTSF)$_2$X. As for Fermi surface, at least around 1 GPa, from 0 to 0.2 T of $B_z$, there must be two pairs of 1D Fermi surface. Between 0.2 and 10 T, a pair of 1D Fermi surface and a straw shaped 2D Fermi surface due to imperfect nesting. And above 10 T, only one pair of 1D Fermi surface remains due to a disappearance of 2D Fermi surface caused by Landau quantum limit.

Devoted note to Professor J.S. Brooks

During completing this paper, one of the authors of this paper, Professor J.S. Brooks passed away on September 27, 2014. He was really a great friend and leader as well as the person who kept
mind of specialist of low temperature. We miss him deeply.

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References