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High-field magnetotransport and Fermi-surface topology in the novel quasi-two-dimensional organic conductor bis(ethylenedithiolo)tetrathiafulvalenium mercuric potassium thiocyanate, (BEDT-TTF)$_2$KHg(SCN)$_4$

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Magnetotransport measurements have been carried out in the novel organic conductor (BEDT-TTF)$_2$KHg(SCN)$_4$. Some remarkable and unusual features were found in the high-field magnetoresistance measured under pulsed magnetic fields up to 40 T; a negative slope above 10 T, a sharp kink structure at 22.5 T, and large enhancement of the Shubnikov–de Haas oscillations above the kink structure. Angle-dependent quantum oscillations were also observed under static fields up to 12 T. The experimental results suggest strong two dimensionality and presence of a weakly warped cylindrical Fermi surface whose cross section is anisotropic and 16% of that of the first Brillouin zone. Vanishing of an overlap between neighboring Landau subbands is proposed as one of possible origins of the sharp kink structure.

The (BEDT-TTF)$_2X$ family of organic conductors [where BEDT-TTF denotes bis(ethylenedithiolo)tetrathiafulvalene] have attracted a great deal of attention because of their low dimensionality and superconductivity. Among these compounds (BEDT-TTF)$_2$Cu(SCN)$_2$, which has polymeric sheets of pseudohalide metal anion Cu(SCN)$_2$, has the highest critical temperature of superconductivity ($T_c = 10.4$ K).[1] The novel compound (BEDT-TTF)$_2$KHg(SCN)$_4$, which is studied in this work, was synthesized as a modification of (BEDT-TTF)$_2$Cu(SCN)$_2$.[2] This compound shows a metallic behavior down to 0.5 K without any superconducting or metal-insulator transitions. It has a layered structure consisting of polymeric anion sheets of $K^+[Hg(SCN)_4]^{2-}$ and donor sheets of zigzag-aligned BEDT-TTF molecules which form the two-dimensional (2D) conducting plane. The anion-layer thickness is much larger (6.8 Å) than other BEDT-TTF compounds because of the three-dimensional (3D) polymeric structure, so that the strong two dimensionality due to weak interlayer couplings is expected. In fact, the conductivity anisotropy σ_c/σ_b is larger than 2000 at 4.2 K, where σ_c is the conductivity along the **c** axis in the conducting plane (**a**-**c** plane) and σ_b is that along the **b*** axis perpendicular to the conducting plane. Since a unit cell contains four BEDT-TTF molecules on the same sheet, two holes per unit cell occupy four energy bands made from the highest occupied molecular orbitals (HOMO's) of BEDT-TTF. The observed metallic behavior suggests the presence of multiple Fermi surfaces (FS's) cut by the zone boundary. According to the 2D tight-binding band calculation using the HOMO obtained by the extended Hückel approximation, a 2D Brillouin zone (BZ) should have a closed FS and a pair of open FS's along the **c** direction.[3] For other BEDT-TTF compounds, several experimental Fermi-surface topology works have been recently reported; the Shubnikov–de Haas (SdH) effect,[4–8] and the newly discovered angle-dependent quantum oscillations of magnetoresistance.[6,9]

In this paper, we study the magnetotransport properties of (BEDT-TTF)$_2$KHg(SCN)$_4$ in order to investigate its electronic structure as a prototype of the quasi-2D system. We observed both the SdH effect and the angle-dependent oscillations. We discuss the Fermi-surface topology using the experimental results and compare it with the results of the band calculation. In addition, we report unusual magnetotransport properties at high magnetic fields.

The high-field magnetotransport measurements were carried out under pulsed magnetic fields up to 40 T. The angle dependence of magnetoresistance was measured in static magnetic fields up to 12 T. The typical size of samples was $2\times1\times0.3$ mm^3. Six gold lead wires (25 μm in diameter) were bonded on the sample by gold paint for both magnetoresistance and Hall resistance measurements. Most of the high-field measurements were done using the phase-sensitive-detection (PSD) technique with an ac bias current (typically $f = 200$ kHz, $I_{\text{peak}} = 1$ mA) in order to increase the signal-to-noise ratio and to prevent the destruction of the sample by the dc Lorentz force. Measurements using low dc bias currents were also done to check the accuracy of the ac measurements and to scale the measured ac data.

Figure 1 shows a typical example of the magnetoresistance $[R(B)-R(0)]$ traces under magnetic fields perpendicular to the conducting plane (**a**-**c** plane). Although the current direction was almost parallel to the **c** axis, the measured resistance possibly contains the **b*** axis component because of the irregular sample shape and the large anisotropy of the conductivity. As shown in the figure, the magnetoresistance exhibits remarkable features in this arrangement: With increasing magnetic fields, the magnetoresistance increases sublinearly and saturates around 10 T. It shows an unusual negative slope above 10 T. A sharp "kink structure" appears around

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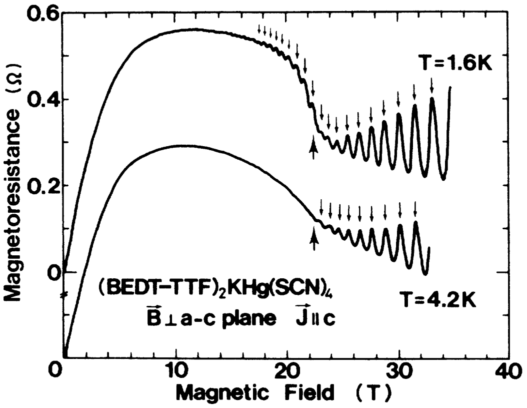

FIG. 1. The transverse magnetoresistance under the fields applied perpendicular to the conducting plane. The kink structure is indicated by the large arrow. The oscillation peaks of the SdH effect are marked by small arrows.

22.5 T as indicated by large arrows in Fig. 1. SdH oscillations are also seen superposed on this background magnetoresistance. The amplitude of the SdH oscillations are largely enhanced above the kink structure. The field position of the kink structure shows no explicit temperature dependence. Although the overall shape of the magnetoresistance trace and the amplitude of the SdH oscillations were slightly different from sample to sample, the qualitative features mentioned above and the field positions of the kink and the SdH peaks were the same in all the samples. Neither the explicit kink structures nor the SdH oscillations were found when magnetic fields were applied parallel to the **a**-**c** plane.

The dependence of the magnetoresistance on the direction of the magnetic field was studied at lower fields in detail. Figure 2(a) shows the angle dependence of the magnetoresistance at several fixed field strengths when the magnetic fields are tilted from the **b*** direction in the **a**'-**b*** plane (the **a**' axis is perpendicular to both the **b*** axis and the **c** axis), while Fig. 2(b) shows the angle dependence when the fields are tilted in the **b***-**c** plane. The angle-dependent quantum oscillations were observed as a series of dips indicated by arrows in Figs. 2(a) and 2(b). The dip positions are almost periodic against the tangent of the angle θ between the field direction and the **b*** axis (normal to the conducting plane). Generally, the oscillation period depends on the direction to which the field is tilted from the normal, as Figs. 2(a) and 2(b) show different periods. These angle-dependent oscillations are considered to originate essentially from the same origin as those observed in θ-(BEDT-TTF)$_2$I$_3$ and β-(BEDT-TTF)$_2$IBr$_2$.[6,9] However, the angle dependence of the magnetoresistance shows some different features from these two compounds: As shown in Fig. 2(a), the background magnetoresistance takes a maximal value when the field is applied perpendicular to the conducting plane and takes a minimal value for the field parallel to the conducting plane, whereas it shows the minimum for the normal direction and the maximum for the parallel direction in θ-(BEDT-TTF)$_2$I$_3$ and β-(BEDT-TTF)$_2$IBr$_2$. In addition, the angle-dependent oscillations of these materials appear as a series of peaks in contrast to dips in (BEDT-TTF)$_2$KHg(SCN)$_4$.

First, we discuss the electronic structure of the quasi-2D conductor (BEDT-TTF)$_2$KHg(SCN)$_4$ using the results of the SdH effect and the angle-dependent quantum oscillations. The period of SdH oscillations $\Delta(1/B)=0.0015$ T^{-1} gives the cross section of FS: $S=0.065$ Å^{-2}. This area corresponds to 16% of the first BZ. The temperature dependence of the oscillation amplitude reflects the Landau-level spacing. Numerical fitting of the conventional formula of the SdH effect gives the cyclotron mass $m_c/m_0=1.4$. The Dingle temperature is deduced from the field dependence of the oscillation amplitude. Employing the theoretical formula of the SdH effect in the 2D system, we obtain the Dingle temperature $T_D=4.0$ K and the relaxation time $\tau=0.3$ ps. The mean free path of $l\sim 350$ Å is estimated from m_c and τ.

The most plausible mechanism of the angle-dependent oscillations is the following model proposed by Yamaji[10]. The energy spectrum of the quasi-2D system with a weakly warped cylindrical FS under magnetic fields is a set of Landau subbands with the dispersion along the field direction. When the magnetic field is tilted from the normal to the 2D plane, the width of the Landau subband around the Fermi level oscillates against the tilted angle, so the conductivity oscillates as a function of the angle through the oscillation of the subband mass. This model is very similar to that for the new type of quantum magnetic oscillations discovered recently in the 2D electron gas formed in a GaAs/Al$_x$Ga$_{1-x}$As heterostructure with the weak lateral periodic potential.[11,12] Assuming this model, we can directly obtain the Fermi wave number k_F for the special direction in the 2D plane from the oscillation period. According to this model, k_F along the main axis is given by $\Delta(\tan\theta)=\pi/bk_F$ in the case of the elliptical cross section of FS. Here, b (~ 20 Å) is the interlayer spacing. The observed period $\Delta(\tan\theta)\sim 1.5$ in the case that the magnetic fields are tilted in the **a**'-**b*** plane gives the Fermi wave number along the **a** axis $k_{F\mathbf{a}}\sim 0.1$ Å^{-1}. For the **c** direction, the period $\Delta(\tan\theta)\sim 3$ gives $k_{F\mathbf{c}}\sim 0.05$ Å^{-1}. Therefore, the cross section of the cylindrical FS has a strong anisotropy (at least 2:1) in the conducting plane. Though the value of k_F is smaller than that expected from the SdH period, their order of magnitude shows good agreement.

The presence of a closed elliptical FS in the 2D BZ has been predicted by the band calculation by Mori and Inokuchi.[3] The cross-sectional area of this FS corresponds to 19% of the first BZ, and this FS has a smaller Fermi wave number along the **c** axis. Qualitative features are very consistent with the experimental results mentioned above although a small discrepancy is found in quantitative details.

Next, we discuss the origin of the anomalous kink structure of high-field magnetoresistance. The field position of the kink structure seems to be independent of temperature. This fact suggests that the mechanism of the kink structure has nothing to do with the many-body effect such as the phase transition, but is ascribed to the single-particle effect related to the electronic structure.

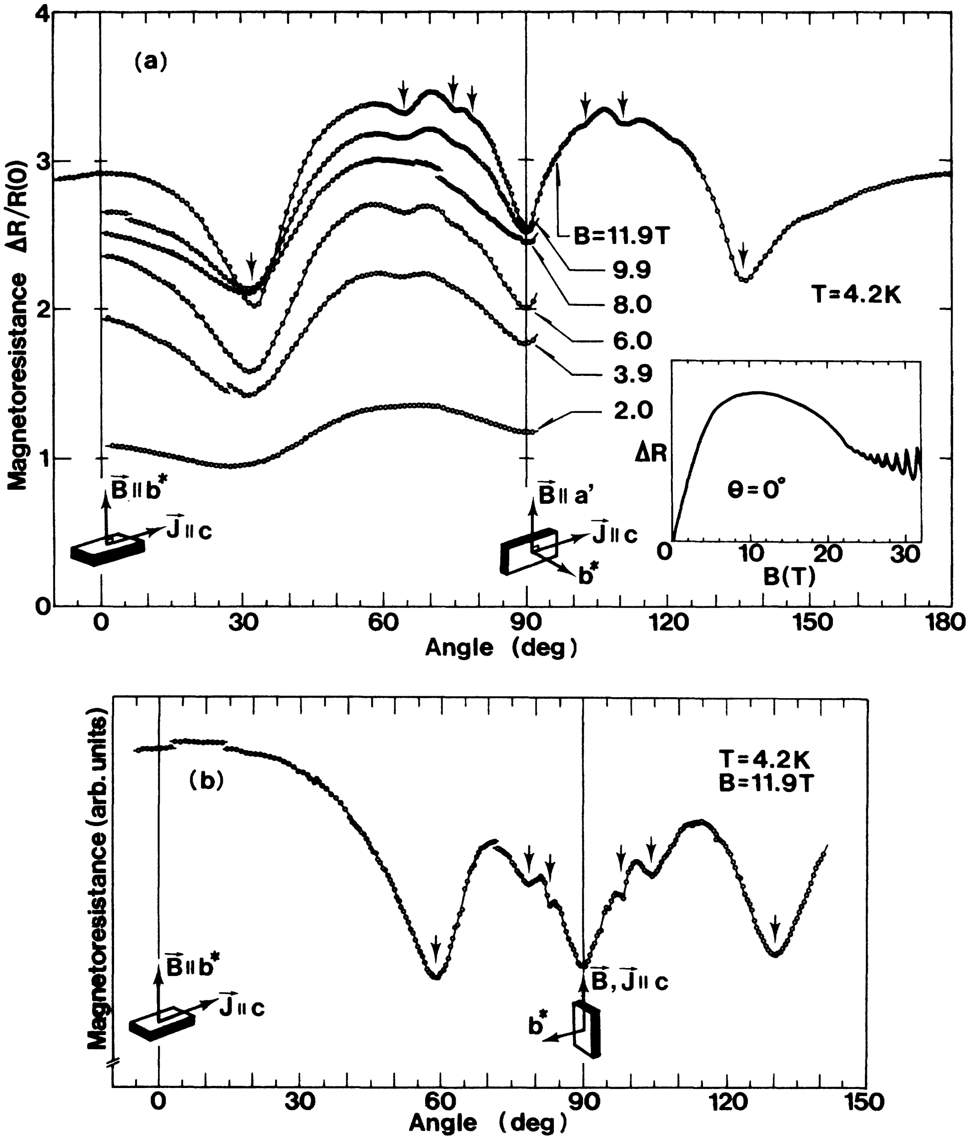

FIG. 2. (a) The angle dependence of magnetoresistance when the field is tilted in the b*-c plane at several fields. The inset shows the magnetoresistance under the fields parallel to the b* axis. Arrows indicate the dip positions of the angle-dependent oscillations. (b) The angle dependence when the field is tilted in the a'-b* plane.

The magnetoresistance behavior, that is, the large negative slope with SdH oscillations, is reminiscent of those observed in the hexagonal divalent metals such as Mg or Zn in which the magnetic breakdown occurs.[13] As the FS calculated by Mori can be regarded simply as a linear chain of elliptical orbits, the magnetic breakdown is possible in this compound. The energy gap E_g at the zone boundary is estimated as about 100 meV from Mori's calculation. Using the effective mass $m^* = 1.4m_0$ and the Fermi energy $E_F = \hbar^2 k_F^2/2m^* \sim 50$ meV, we estimate the characteristic magnetic field for the magnetic breakdown as $B_0 \sim E_g^2/(2e\hbar E_F/m^*) \sim 1000$ T. Therefore, the magnetic fields used in the present work are too small to cause the magnetic breakdown. In addition, it is hard to imagine that the tunneling process, like the breakdown, causes such an abrupt change, like the kink structure. Therefore, the magnetic breakdown is unlikely to occur in the present case.

The vanishing of an overlap of the Landau subbands might be a more plausible single-particle mechanism responsible for the origin of the kink structure. The system has a weak dispersion along the normal axis of the conducting plane with the bandwidth $4t$, where t is an interlayer transfer integral. Under magnetic fields perpendicular to the conducting plane, the system is quantized to Landau subbands with the width $4t$. At low fields their width is larger than the cyclotron energy $\hbar\omega_c = \hbar eB/m^*$, and thus the density of states (DOS) at the Fermi level involves many overlapping Landau subbands. Therefore, the system is regarded as 3D-like. In this case, beating of the SdH oscillations is expected. At the critical field given by the condition $4t = \hbar\omega_c$, this overlap between the neighboring Landau subbands disappears, and the spectrum above the critical field is discretized to a set of Landau subbands separated by gaps. In this sense, the system becomes 2D-like. The perfect discretization of DOS in energy causes large SdH oscillations. The observed enhancement of the SdH amplitude above the kink field can be understood well, if we ascribe the kink field to the critical field of the "dimensional crossover" from 3D to 2D. It is considered that in the present case the SdH oscillations are almost smeared out in the 3D region by the collision broadening of overlapping Landau subbands and they are enhanced in the 2D region reflecting the large oscillation of DOS in energy. Assuming this mechanism, we can estimate the interlayer transfer integral as $t = 0.5$ meV using $m^* = 1.4m_0$. This value is very close to that estimated from the beating of the giant SdH oscillations in β_H-(BEDT-TTF)$_2$I$_3$.[8]

In conclusion, we observed both the large SdH oscillations and the angle-dependent oscillations in the novel organic conductor (BEDT-TTF)$_2$KHg(SCN)$_4$. The results suggest the strong two dimensionality and the existence of the weakly warped cylindrical FS which is anisotropic in the conducting plane. We also found the anomalous transport behavior at high magnetic fields; the negative slope of magnetoresistance, the remarkable kink structure, and the large enhancement of the SdH effect above the kink structure. We ascribe the kink field to the field where the overlap between neighboring Landau subbands vanishes, and obtain a reasonable value for the interlayer transfer integral. It would be interesting to measure the angle dependence of the magnetoresistance in the high-field regime: The coexistence of the SdH and angle-dependent oscillations should be observed, and the angle dependence of the kink field would be a good test for the model presented here.

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