Magnetotransport and Fermi-surface topology of $[\text{bis}(\text{ethylenedithio})\text{tetrathiafulvalene}]_2\text{KHg}(\text{SCN})_4$

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We have performed measurements of the angle-dependent magnetoresistance in the organic metal $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ [where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene] in fields of up to 30 T and at temperatures down to 0.4 K. Shubnikov-de Haas (SdH) oscillations were observed over a wide range of fields and field orientations; at fields below 22 T, the dominant oscillation for $\mathbf{B}$ perpendicular to the highly conducting layers has a fundamental field $B_F = 670 \pm 5$ T. However, we have observed that another series with a fundamental field which is sample dependent and some 100–200 T higher than the dominant series is also present, resulting in a beat structure in the envelope of the oscillations. In addition, significant hysteresis is seen in the magnetoresistance. At ~ 22 T, the resistance falls sharply, the hysteresis almost vanishes, the measured effective mass increases, and the SdH oscillations simplify to a single series, with a frequency $B_F = 656 \pm 10$ T similar to that predicted by band-structure calculations using the room-temperature crystal structure. On rotating about the a axis the transition at ~ 22 T slowly moves to lower field. $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ is known to order antiferromagnetically at ~ 8 K, and we suggest that the effects observed below the transition may result from a magnetic superlattice in the direction perpendicular to the highly conducting planes; using this model, we are able to account qualitatively for most of the features observed. The interplane interactions responsible for the ordering are overcome by the external magnetic field at ~ 22 T, resulting in the fall in resistance and the simpler high-field behavior.

I. INTRODUCTION

Charge-transfer salts of the molecule bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF) possess interesting electronic properties; by varying the anion X incorporated into the salt $(\text{BEDT-TTF})_nX$, the stoichiometry and band filling may be adjusted, producing compounds which are metallic, semimetallic, or semiconducting.¹ The electronic properties of the salts are quasi-two-dimensional, and electron-electron correlation effects are thought to be important.^{2,3} Much recent activity has centered on the superconducting properties of these materials, with salts such as $(\text{BEDT-TTF})_2\text{Cu}(\text{SCN})_2$ possessing critical temperatures above 10 K.^{3,4} The compound studied in this work, $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$, was originally synthesized as a likely superconducting modification of $(\text{BEDT-TTF})_2\text{Cu}(\text{SCN})_2$, but was found

to be metallic down to 0.4 K;⁵ the closely related salt $(\text{BEDT-TTF})_2\text{NH}_4\text{Hg}(\text{SCN})_4$ is, in fact, superconducting, with a critical temperature of 0.8 K.⁶

$(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ exhibits a number of features that distinguish it from other BEDT-TTF charge-transfer salts; for example, the material orders antiferromagnetically at ~ 8 K.⁷ In marked contrast to magnetically ordered charge-transfer salts of other molecules (e.g., TMTSF), $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_2$ remains metallic below 8 K. Around this point the temperature dependence of the electrical resistivity also changes slope and the magnetoresistance is dramatically enhanced.⁸ At low temperatures and in high magnetic fields, the Shubnikov-de Haas oscillations are anomalous in form and sample dependent (cf. Refs. 7–10). In addition, at around 22 T there is a region of strong negative magnetoresistance known as the “kink” transition.¹⁰ In this work we have studied the magnetoresistance of (BEDT-

FIG. 1. Two-dimensional Brillouin zone and Fermi surface of $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ (after Ref. 11). There are a pair of electronlike open Fermi surfaces running along the c direction, and a closed-hole-like 2D pocket around the V point.

$\text{TTF})_2\text{KHg}(\text{SCN})_4$ in fields of up to 30 T applied in a variety of orientations in order to elucidate such features. The high quality of our samples allows observation of Shubnikov–de Haas oscillations down to ~ 3 T at 0.5 K. As a result, we have been able to observe the presence of *two* series of Shubnikov–de Haas oscillations that interfere, causing strong beating below 10 T. These result from a band structure that is a modification of that calculated using the room-temperature crystal structure. On the other hand, a detailed study of the magnetoresistance above the “kink” transition shows rather good agreement with the predictions of the calculation. These results lead us to suggest a model for the internal magnetic order of the salt which modifies the band structure at low magnetic fields. The model accounts qualitatively for the experimental data, and indicates that the “kink” represents a field-induced change in internal magnetic order.

$(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ has a layered structure typical of metallic BEDT-TTF salts, resulting in highly conducting (ac) planes formed by zig-zag stacks of BEDT-TTF molecules separated by layers of polymeric anion sheets of $\text{K}^+[\text{Hg}(\text{SCN})_4]^{2-}$.⁴ The Fermi surface predicted by extended-Hückel-model band-structure calculations^{11,12} using the room-temperature crystal parameters^{5,11} is shown in Fig. 1; an approximately isotropic two-dimensional (2D) hole pocket lies at the corner of the Brillouin zone and a quasi-one-dimensional electron band runs down the zone center. As the anion sheet thickness is much larger (0.63 nm) than for most BEDT-TTF compounds, the interlayer transfer-matrix element t will be very small ($< \sim 0.5$ meV),¹⁰ resulting in a very small degree of warping of the cylindrical hole Fermi surface in the b (interplane) direction; i.e., the material is very two dimensional.

II. EXPERIMENT

$(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ crystals were grown electrochemically^{5,13,14} using BEDT-TTF prepared by the method of Larsen and Lenoir;¹⁵ the crystals are small black distorted diamond-shaped platelets. The plane of the platelets is defined by the highly conducting ac plane.⁵ Gold wires were attached on both faces of the

platelets using platinum paint, and the resistance was measured for currents in the b^* direction (perpendicular to the conducting planes). Standard ac-current lock-in techniques were used for the measurements; frequencies between 5 and 150 Hz were employed. Greater care was taken to check that the measured resistance was frequency independent, and, to avoid sample heating, currents were generally kept below $20 \mu\text{A}$. The crystals were studied over a wide range of fields, orientations, and temperatures in a ^3He cryostat which allowed the sample to be rotated *in situ*; magnetic fields were provided by a variety of superconducting, resistive, and hybrid magnets at Nijmegen and Oxford. Calibrated RuO and Ge resistance thermometers were used for temperature measurement. After a slow cool down to ~ 30 K lasting 2–3 h, the samples were cooled quickly to 1 K ($< \sim 1$ min), and then not warmed above 10 K for the duration of the experiments.

A. Magnetoresistance below 20 T

Figure 2(a) shows the magnetoresistance of two different crystals at 0.5 K, measured with the magnetic field applied perpendicular to the ac plane. Both samples exhibit a magnetoresistance that rises to a peak before decreasing, and superimposed on this are Shubnikov–de Haas oscillations. In each case there is hysteresis in the magnetoresistance; that recorded with the magnetic field sweeping upwards is generally larger than that recorded on the downsweep. Note also that the size of the hysteresis is sample dependent. Magnetization measurements reported by Sasaki *et al.*⁸ point to the onset of antiferromagnetic order at 8 K, with the dipole alignment occurring in the ac plane; the magnetoresistance hysteresis is a further indication of the presence of an effective internal magnetic field in this material.

Magnetoresistance data from the samples shown in Fig. 2(a) are plotted in Fig. 2(b) with the slowly varying background subtracted. Note that two primary SdH frequencies are present in both samples, causing beating, but that the beat frequency differs. The two frequencies, their harmonics, sums, and differences are plainly visible in the Fourier amplitude spectrum of the magnetoresistance [Figs. 3(a) and 3(b)]. One of the SdH frequencies, which we shall label B_{F1} , at $B_F = 670 \pm 5$ T, is present in all of the samples studied; its frequency is in very good agreement with values obtained in other studies.^{7–10} However, the other SdH frequency B_{F2} is sample or cooling method dependent, and ranges in our samples from $\sim B_{F1} + 50$ T to $\sim B_{F1} + 200$ T. In Fig. 3(a) sample 1 has $B_{F2} \sim 860$ T, whereas sample 2 [Fig. 3(b)] has $B_{F2} = 790$ T, leading to the different beat periodicities seen in the Shubnikov–de Haas oscillations [Fig. 2(b)]. Referring once more to Fig. 2(a), it will be seen that the degree of magnetoresistance hysteresis correlates with the size of the frequency difference between B_{F1} and B_{F2} , and we have found this to be true for all samples studied.

As mentioned in the Introduction, previous workers have observed Shubnikov–de Haas oscillations in $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ that appear to be strongly spin split.^{7–10} In the most sophisticated analysis of this split-

ting, Sasaki, Sato, and Toyota⁷ noted that the oscillations, supposedly due to spin splitting, changed in phase with magnetic field; a field-dependent exchange enhancement of the level splitting was proposed to explain the data. However, a simpler explanation in the light of our work is that the phase change is due to the two interfering series of Shubnikov-de Haas oscillations (B_{F1} and B_{F2}). Note that in sample 2, while the beats due to B_{F1} and B_{F2} are very prominent below 10 T, between 12 and 20 T the data are very similar to those obtained by Sasaki, Sato, and Toyota⁷ showing the splitting of the B_{F1} series apparently changing in relative phase. The two series are probably present but undetected in the data of Sasaki, Sato, and Toyota⁷ because for their sample the Shubnikov-de Haas oscillations are only well resolved

above ~ 10 T. Different values of B_{F2} will produce different apparent phase shifts in the region of splitting, leading to the rather diverse appearance of the published data (cf. Refs. 7–10).

Kang¹⁶ has attributed the splitting of the Shubnikov-de Haas oscillations at 17–20 T [see Fig. 2(b)] to beats between extremal areas of the warped Fermi tube, rather than to Pauli spin splitting. However, as will be shown in the next paragraph, the amplitude of the splitting of the oscillations varies with the orientation of the magnetic field in the manner expected for spin splitting, and not in the manner expected for the mechanism of Kang.¹⁶ Note also that Kang's model cannot account for the presence of both B_{F1} and B_{F2} ; the interplane transfer integral t would have to be ~ 10 meV to produce

FIG. 2. Magnetoresistance of two $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ crystals (labeled 1 and 2) for magnetic field parallel to b^* and $T=0.4$ K. (a) shows raw data for both upsweeps and downsweeps of the magnetic field; in each case the lower resistance is observed on the downsweep. (b) shows the Shubnikov-de Haas oscillations with the background magnetoresistance subtracted.

FIG. 3. Fourier transforms of magnetoresistance data shown in Fig. 2(b) [(a), data for sample 1; (b), data for sample 2]. Two primary frequencies (B_{F1} at ~ 670 T, and B_{F2} sample dependent at $B_{F1} + 100$ –200 T) are present, along with their sums, differences, and harmonics. (c) shows the Fourier amplitudes of the B_{F1} (squares) and B_{F2} (circles) Shubnikov–de Haas oscillation series of sample 1 obtained by transforming over a 0.01-T^{-1} range of inverse field, plotted against inverse field (see text).

such a large difference in Fermi-surface extremal area. As mentioned in the Introduction, t is, in fact, rather small.

Figure 3(c) shows the field dependence of the Fourier amplitudes of B_{F1} and B_{F2} . The points are obtained by Fourier-transforming magnetoresistance within a 0.1-T^{-1} -wide data window. Note that as one goes to higher field (lower inverse field), B_{F2} falls in amplitude, whereas B_{F1} follows the expected Dingle factor dependence (see, e.g., Ref. 17). B_{F2} also falls off in amplitude as the magnetic field is tilted away from the b^* direction. Typical magnetoresistance data with the magnetic field inclined at a variety of angles to the b^* direction ($\theta=0^\circ$ is defined to be field perpendicular to the conducting planes, i.e., parallel to b^*) are shown in Fig. 4(a); at higher angles the Shubnikov–de Haas oscillations become chiefly determined by B_{F1} . In addition to the fundamental fields of both Shubnikov–de Haas oscillation series shifting with angle as $1/\cos\theta$, as expected for a quasi-2D system, the amplitude of the B_{F1} series of Shubnikov–de Haas oscillations can be seen to oscillate as a function of tilt an-

FIG. 4. (a) shows the oscillatory component of magnetoresistance for magnetic field applied at various angles to the b^* direction (rotation in the b^*c plane). (b) shows harmonic ratios $M_1(\theta)/M_2(\theta)$ of the primary SdH frequency B_{F1} for the field range 17–20 T plotted as a function of tilt angle; data are points, and the line is a fit using Eq. (1).

gle, and the splitting of the oscillations discussed above varies in amplitude, disappearing at some angles. This type of behavior has been reported in a number of BEDT-TTF salts (see, e.g., Ref. 18 and references cited therein), and is interpreted as a direct observation of Pauli spin-splitting effects. The Pauli spin splitting of the Landau levels, which depends to first order only on the total magnetic field, is roughly angle independent, whereas the cyclotron splitting is determined by the component of the magnetic field perpendicular to the conducting planes; hence, at fixed field, the Pauli and Landau splittings change in relative size as the field is tilted, leading to oscillations in the Shubnikov–de Haas amplitude.¹⁹ When pairs of levels overlap, the amplitude of the fundamental is large; when the levels are equally spaced, splitting of the oscillations is apparent and the second-harmonic amplitude is large. These changes in amplitude with tilt angle may be used to determine the product of the g factor and the effective mass; this so-called harmonic-ratio technique has been treated in detail by many authors^{17,18} and we shall only quote the result for the ratio of the amplitude M_1 of the fundamental Shubnikov–de Haas frequency to that of the second harmonic M_2 :

$$\begin{aligned} M_1(\theta)/M_2(\theta) \\ = \{ \cos[\pi g^* m^*(\theta)/2] / \cos[\pi g^* m^*(\theta)] \} G(T, H), \end{aligned} \quad (1)$$

where g^* is the effective g factor, and $m^*(\theta)$ is the effective mass, assumed to vary with tilt angle θ as $m^*(\theta) = m^*(0^\circ)/\cos\theta$; G is a slowly varying function of temperature and field, which may be taken as a constant over a restricted range of field.^{17,18} Magnetoresistance data were Fourier transformed over a restricted magnetic-field range, and the ratio of the fundamental frequency Fourier amplitude of the B_{F1} series to that of its second harmonic as a function of angle was compared with Eq. (1), using $g^* m^*(0^\circ)$ as an adjustable parameter. Ratios for the field range 17–20 T are plotted in Fig. 4(b), along with the curve generated by Eq. (1): the best fits to all data below 20 T were obtained using $g^* m^*(0^\circ) = 3.07 \pm 0.02$. If one assumes that $g^* = 2$, as measured by ESR,^{8,20} this indicates an effective mass of $\sim 1.5m_e$, in reasonable agreement with the values $\sim 1.4m_e$ obtained by others¹⁰ using the temperature dependence of the Shubnikov–de Haas amplitude of the B_{F1} series.

Figure 5 shows the angle dependence of the background (i.e., nonoscillatory) magnetoresistance at fixed field and at a temperature of 1.5 K for currents in the b^* direction. Data obtained for the magnetic field in the ab^* plane [Fig. 5(a); axis of rotation $\sim c$] and in the b^*c plane [Fig. 5(b); axis of rotation $\sim a$] are shown, and it can be seen that the magnetoresistance is rather isotropic, in contrast to many other BEDT-TTF salts (see, e.g., Ref. 14); this is perhaps surprising in view of the presence of the quasi-1D electron band (see Fig. 1). In addition to the major magnetoresistance extrema for the magnetic field perpendicular to and in the conducting plane, small-

FIG. 5. Angle dependence of the background magnetoresistance at 1.5 K for a magnetic field of 17 T rotated in the ab^* (a) and b^*c (b) planes. Weak maxima in the magnetoresistance are indicated by arrows.

er maxima and minima are visible at certain angles. For both rotation axes, maxima occur at angles $\pm \sim 39^\circ$ and $\pm \sim 70^\circ$ and minima at $\sim 63^\circ$. Similar phenomena have been observed by other groups using fields applied in the bc plane and currents in the c direction; Osada *et al.*¹⁰ noted a minimum at $\sim +60^\circ$, and very weak maxima may be discerned at $\pm \sim 70^\circ$ in the data of Sasaki *et al.*⁸ The origins of these weak oscillations in the background magnetoresistance will be discussed in more detail below. However, the presence of the weak maxima and minima at very similar angles for both axes of rotation suggests that the portion of Fermi surface dominating the magnetotransport is quasi-two-dimensional and roughly isotropic in the ac plane.

B. Magnetoresistance above 20 T

Figure 6(a) shows the high-field magnetoresistance for several angles θ between the field and the sample surface normal. For $\theta < \sim 50^\circ$, a large drop or “kink”¹⁰ in resistance occurs at ~ 22 T; at higher fields only one series of Shubnikov–de Haas oscillations is observed, with a frequency $B_F = 656 \pm 10$ T (the same as B_{F1} to within the experimental errors) and the hysteresis between upswing and downswing almost vanishes. However, there is an apparent 180° phase change between the B_{F1} series of Shubnikov–de Haas oscillations and those above the “kink” transition, also noted by Osada.¹⁰ As the tilt angle increases, the “kink” transition gradually moves to a lower total field with the approximate functional form $B = B_0 \cos^{1/2}\theta$ [see Fig. 6(b)], until it vanishes at $\theta \sim 57^\circ$.

On tilting, the high-field (> 23 T) Shubnikov–de Haas oscillation frequency follows a $1/\cos\theta$ dependence as ex-

pected of a 2D metal, and there are again the amplitude and harmonic content oscillations expected from the varying spin to cyclotron splitting ratio as the tilt angle increases. Using Eq. (1), the angle dependence of the harmonic ratio for the oscillations above the “kink” transition is fitted by $g^*m^*(0^\circ)=4.02\pm0.02$; if one assumes that the g factor remains constant at $g^*=2$, this would seem to indicate that the effective mass has increased to $\sim 2m_e$. Figure 6(a) also illustrates that there are again oscillations in the background magnetoresistance. The background resistance between 23 and 30 T exhibits maxima at the angles $33^\circ\pm1^\circ$, $59^\circ\pm1^\circ$, and $70^\circ\pm1^\circ$. This is a common phenomenon in BEDT-TTF charge-transfer salts, and has been successfully explained by Yamaji,²¹ who showed that at certain angles, all the semiclassical k -space closed orbits around the warped cylindrical Fermi surface have approximately the same area. This means that the density of states at the Fermi energy is greatly enhanced, resulting in a maximum in the background magnetoresistance. The angles θ_y at which the maxima occur are given by²¹

$$bk_F \cos \theta_y = \pi(i - 1/4), \quad (2)$$

FIG. 6. (a) shows the angular dependence of the high-field magnetoresistance showing a drop (“kink”) in resistance around 22 T; the traces are offset for clarity and the temperature is 490 mK. The inset [(b)] shows the angular dependence of the “kink” (points) fitted to a $\cos^{1/2}\theta$ dependence (line). (c) is a plot of the tangent of the angle at which the background resistivity at 30 T exhibits maxima vs peak index minus one quarter. Data are points; the straight-line fit is shown as a solid line.

where b is the interplane spacing (i.e., the unit-cell height), k_F is the mean radius of the warped cylindrical Fermi surface, and i is an integer. Figure 6(c) shows a plot of the experimental values of $\cos \theta_y$ ($\theta_y = 33^\circ$, 59° , and 70°) versus index i ; the data lie on a straight line, of gradient π/bk_F . If we assume that the 2D hole pocket section of the Fermi surface is approximately isotropic (as predicted in the band-structure calculations shown in Fig. 1) and that the transfer energy t is very much smaller than E_F (a reasonable assumption discussed in the Introduction), we can obtain k_F from the Shubnikov–de Haas fundamental field using the following relationship:¹⁷

$$k_F = (4\pi e B_F / h)^{1/2}. \quad (3)$$

Using the experimental value $B_F = 656$ T, and the gradient obtained from Fig. 6(c), we find $b = 2.1 \pm 0.1$ nm, very close to the value of 2.0 nm obtained from x-ray-crystallography measurements.^{5,10}

III. DISCUSSION

The experimental data reported above show that $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ undergoes a field-induced alteration in the band structure at ~ 22 T, observable as a drop in resistance that has been labeled the “kink” transition. Above this field, the behavior of the magnetoresistance is rather conventional for a BEDT-TTF salt, and indicates a Fermi surface very similar to that calculated^{11,12} (see Fig. 1) from the room-temperature lattice parameters. The Fermi surface area ($\sim 16\%$ of the room-temperature Brillouin zone) given by the Shubnikov–de Haas oscillation frequency ($B_F = 656 \pm 10$ T) is similar to the calculated area of the hole pocket (19% of the room-temperature Brillouin zone). Furthermore, the oscillations in the magnetoresistance as a function of angle are accurately predicted by the Yamaji formula [Eq. (2)] and the known unit cell, indicating that the section of Fermi surface dominating the transport is rather isotropic (see Fig. 1). Finally, the angular dependence of the harmonic content of the Shubnikov–de Haas oscillations may be fitted using $g^*m^*(0^\circ) = 4.02$.

In contrast, the behavior below the “kink” feature is significantly different. Hysteresis occurs in the magnetoresistance, and there are two series of Shubnikov–de Haas oscillations, one at $B_F = 670 \pm 5$ T (B_{F1}), and the other (B_{F2}), which is sample dependent. Harmonic analysis of the B_{F1} series, which is close in frequency to the high-field Shubnikov–de Haas oscillations, yields $g^*m^*(0^\circ) = 3.07$, suggesting that the effective mass changes at the “kink.” Another difference compared to the high-field data is apparent from the low-field angle-dependent oscillations in the background magnetoresistance (Fig. 5). The tangents of the angles at which the peaks in magnetoresistance occur (40° and 70°) do not lie on a straight line through the origin as a function of peak index minus one quarter [cf. Fig. 6(c)], indicating that the Yamaji formula [Eq. (2)] is probably not applicable. If one tries to force a straight-line fit through the points and the origin, the gradient indicates an interplane spacing of $b \sim 3.3$ nm when k_F is derived from the fundamental field

of the B_{F1} series of oscillations [670 T; see Eqs. (2) and (3)]; this is ~ 1.6 times the structural interplane spacing. The value is only slightly improved by using the fundamental field of the B_{F2} series to estimate k_F . Others have suggested that this anomaly is evidence that the 2D hole pocket in the Fermi surface is anisotropic;¹⁰ however, the fact that magnetoresistance maxima are seen at the same angles for the field in both the ab and bc planes indicates that the 2D section of the Fermi surface is, in fact, rather isotropic. Instead, the anomalous positions of the magnetoresistance oscillations are probably due to the presence of two interacting sections of a two-dimensional Fermi surface suggested by the two different Shubnikov–de Haas frequencies and their mixed harmonics [Figs. 2(a) and 2(b)].

Osada *et al.*¹⁰ have postulated that the “kink” transition represents the field at which the cyclotron energy is equal to the bandwidth $4t$ in the b direction. At fields below this condition, the density of states at the Fermi energy will involve many overlapping Landau levels, whereas above it, Osada *et al.* maintain that the energy spectrum becomes “2D-like,” due to the removal of the overlap between neighboring Landau levels.¹⁰ In this way, the prominent increase of the Shubnikov–de Haas oscillation amplitude and fall in resistance at the “kink” seen in the results of Osada *et al.* was explained.¹⁰ There are, however, several reasons to doubt this mechanism. First, other BEDT-TTF salts with similar band structure, such as $(\text{BEDT-TTF})_2\text{NH}_4\text{Hg}(\text{SCN})_4$, should also exhibit “kinks” at a similar field; none has yet been observed. Secondly, one would expect the “kink” feature to show the same angular dependence as the cyclotron energy (i.e., to depend on the component of magnetic field perpendicular to the 2D plane) and hence move to higher total fields as the field is tilted away from b^* . Figure 6 shows that the converse is, in fact, true. Thirdly, under certain conditions, the amplitude of the Shubnikov–de Haas oscillations has been observed to *decrease* at the “kink,” rather than increase.²²

$(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ is thought to be in an antiferromagnetic ground state below ~ 10 K, perhaps due to an incommensurate spin-density wave associated with the quasi-one-dimensional part of the band structure.⁸ A spin-density wave would open up an energy gap along this part of the band, preventing the quasi-one-dimensional electrons taking part in the low-temperature electrical conductivity; this is supported by the rather isotropic magnetoresistance (Fig. 5). The remaining conduction electrons (in the 2D pockets; Fig. 1) and the spin-density wave will form an interacting system; this is suggested by the changes in the resistance and magnetoresistance occurring at the same temperature as the reported onset of antiferromagnetism.⁷

Evidence that the “kink” transition is associated with a change in this magnetic order comes from recent magnetization data in which a large increase in the magnitude of the magnetization is seen at ~ 22 T.²² In order to explain the presence of the “kink” transition and the different behavior above and below it, we suggest that, at low fields, the system is not only antiferromagnetically ordered within the plane, but that there is also a magnet-

ic superlattice in the b^* direction, causing a doubling of the unit cell along this axis (and a consequent halving of the Brillouin zone in this direction). A possible mechanism for this is canted antiferromagnetic order within each plane, with the direction of the canted moment alternating from one layer to another [Fig. 7(a)]. The transition at ~ 8 K might represent the onset of (commensurate) interplane order among incommensurate intraplane spin-density waves.

If the spin distribution acts as a static background from which the conduction electrons scatter, the band structure will be divided into two magnetic subbands separated by a magnetic band gap with a size of the order of the exchange energy between conduction electrons and the spin-density wave. The formation of a magnetic band gap will double the number of Shubnikov–de Haas frequencies; a schematic representation of the possible Fermi surface is shown in Fig. 7(b). The size of the postulated magnetic band gap may be estimated by assuming that the difference in frequency between B_{F1} and B_{F2} results from differences in area of the two concentric 2D Fermi-surface sections at the zone corners [Fig. 7(b)]. Using the experimental value of the effective mass to determine the curvature of the bands close to the Fermi energy, and typical values of B_{F1} and B_{F2} , we obtain a magnetic energy gap between 5 and 10 meV. This is larger than the interplane interaction energy suggested by the ordering temperature of ~ 8 K, and may be evidence of a stronger

FIG. 7. Schematic of effects of magnetic superlattice. (a) represents the magnetic moments in an incommensurate spin-density-wave antiferromagnetic state, including a $2b$ magnetic superlattice due to a canted arrangement of dipoles. A possible form of the two-dimensional sections of the Fermi surface is shown in (b); there are now two concentric hole pockets at the zone corner (cf. Fig. 1). A magnetic energy gap ~ 10 meV has been used to calculate this figure (see text). (c) represents the suggested arrangement of dipoles at fields above the “kink” transition.

interaction between the spin-density wave and the conduction electrons.

The non-Dingle field dependence of the SdH amplitude of B_{F2} could signal magnetic depopulation of one of the new sections of Fermi surface [see Fig. 7(b)]. This will affect only the phase, and not the frequency, of the Shubnikov-de Haas oscillations, provided that the change in areal carrier density per pocket is approximately linear in B ;²³ this is the natural dependence in a two-dimensional system when the energy shifts are linear in B .

High external magnetic fields may change the magnetization in such a way that all planes become equivalent and hence the magnetic band gap is removed; a possible mechanism, suggested by the experimentally observed increase in magnetization,²² is for the canting direction to become uniform in all planes. The number of states available for small- k scattering at the Fermi surface will be lowered [cf. Figs. 7(b) and 1], thus reducing the resistance. We therefore identify this change in magnetic order with the "kink" transition occurring at ~ 22 T. We also note that typical magnetic energies (cyclotron or spin splittings) at 22 T are of the same order of magnitude as the interplane energy suggested by the transition temperature of ~ 8 K, giving further support to the connection between the antiferromagnetic order and the occurrence of the "kink." However, more work remains to be done on the magnetic properties of $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$ before a detailed account of the "kink" transition can be given.

At lower magnetic fields, the varying degree of internal magnetic order between samples, caused perhaps by different cooling rates, will lead to different splittings between B_{F1} and B_{F2} . The hysteresis in resistance will occur because on the downsweep, the spin system will be more ordered, leading to less spin-disorder scattering (see, e.g., Ref. 24); it is noticeable that the lower resis-

tance is almost invariably seen on the downsweep, supporting this mechanism.

IV. SUMMARY

We have used high-field magnetoresistance measurements at a variety of temperatures and orientations to study the band structure of $(\text{BEDT-TTF})_2\text{KHg}(\text{SCN})_4$, which is thought to be antiferromagnetic below ~ 8 K. We have observed at low fields two dominant series of Shubnikov-de Haas oscillations, one of which is sample dependent and has an anomalous field-dependent amplitude. Above a resistive "kink" transition at ~ 22 T the only detectable oscillation for B perpendicular to the highly conducting layers has a fundamental field $B_F = 656 \pm 10$ T, and the band structure is similar to that calculated using the room-temperature crystal structure. We have proposed that the low-field effects result from a magnetic superlattice in the b^* direction; the interactions responsible for this are overcome by the external magnetic field at ~ 22 T, resulting in the "kink" and the simpler high-field behavior.

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