High-field magnetoresistance oscillations in α -[bis(ethylenedithio)tetrathiafulvalene] $_2$ KHg(SCN) $_4$: The effects of magnetic breakdown, exchange interactions, and Fermi-surface reordering

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High-field magnetoresistance measurements have been performed on single-crystal samples of the charge-transfer salt α -ET $_2$ KHg(SCN) $_4$ [where ET is bis(ethylenedithio)tetrathiafulvalene] using fields of up to 30 T. We examine the character of the Shubnikov-de Haas oscillations for a range of samples, temperatures, and field regimes to understand the role played by spin splitting and magnetic breakdown effects. Angle-dependent magnetoresistance oscillations (AMRO) in magnetic fields below the kink transition (22 T) indicate that the low-magnetic field band structure is dominated by a one-dimensional (1D) open section of Fermi surface (FS) inclined at $\sim 21^\circ$ to the crystallographic b^*c plane; above the kink, the AMRO change in character, indicating that the FS now possesses a 2D closed section in the form of a distorted cylinder. The data are discussed in terms of a recent model of the low-field band structure and give definitive proof that the kink is the signature of a magnetic-field-induced change in the FS.

I. INTRODUCTION

Charge-transfer salts of the form ET $_2X$ [where ET is bis(ethylenedithio)tetrathiafulvalene and X is a monovalent anion] have been the subject of intense experimental study since high-quality single crystals became available.^{1,2} The ET molecules are linked to each other by overlap of their molecular π orbitals and stack along side one another, separated by sheets of the anion X , to form a two-dimensional (2D) conductive network. Within this family of materials α -ET $_2MHg(SCN)_4$ [$M=K$ (Ref. 3), Tl (Ref. 4), Rb (Ref. 5), or NH $_4$ (Ref. 6)] were synthesized as modifications of κ -ET $_2Cu(NCS)_2$ in an attempt to obtain a higher superconducting transition temperature (T_c). The salts are isostructural (the so-called α phase) and as a consequence have almost identical predicted Fermi surfaces (FS's) consisting of a 2D closed hole pocket and a pair of 1D planar FS sheets⁶ [Fig. 1(a)].

The salt α -ET $_2NH_4Hg(SCN)_4$ is a superconductor,

with $T_c \sim 1$ K. In contrast, the salts ET $_2MHg(SCN)_4$ (where $M=K$, Tl, and Rb) remain metallic down to < 100 mK (Ref. 7) and all show the onset of antiferromagnetic order at temperatures $T_N \sim 8$ –10 K with the easy axis in the highly conducting ac plane.^{8,9} Electron-spin-resonance (ESR) (Ref. 10) and NMR (Ref. 11) measurements also show anomalous changes below T_N in the intensity of the ESR signal and in the proton-relaxation time, respectively. Hall resistance measurements¹² have demonstrated that the carrier density steeply decreases below T_N . These phenomena have been linked to the possible presence of a spin-density-wave (SDW) ground state as evidenced by a recent muon-spin-rotation study of the $M=K$ salt.¹³ No such behavior has been observed in α -ET $_2NH_4Hg(SCN)_4$.

Another transition has been discovered in the low-temperature resistance of the salts with $M=K$, Tl, and Rb,¹⁴ but this time as a function of *field* (see Fig. 2). This so called "kink transition"¹⁵ is not observed in

FIG. 1. (a) Calculated Fermi surface for $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ (after Ref. 3). (b) Below T_N , the Fermi surface is modified to form new open sheets and small cylinders: Thus a magnetic breakdown network arises (after Ref. 9).

$\alpha\text{-ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$ [Fig. 2(b)] below 50 T; instead, a series of Shubnikov-de Haas oscillations (SdHO's) of the conventional form are observed with a periodicity within a few percent of that expected from the predicted 2D hole pocket and superimposed upon a monotonically increasing magnetoresistance (MR). However, in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ the MR rapidly increases up to ~ 10 T (at 0.5 K), but above this field, the MR decreases until at ~ 23 T a kink in the MR appears, above which the MR increases with increasing field [Fig. 2(a)]. Below the kink and above ~ 7 T the MR exhibits significant hysteresis,^{8,9,16,17} particularly when the *ac* plane of the sample is tilted with respect to the magnetic field.¹⁸ It

FIG. 2. Magnetoresistance measured in a pulsed magnetic field for $\alpha\text{-ET}_2\text{MHg}(\text{SCN})_4$ with (a) $M = \text{K}$ and (b) $M = \text{NH}_4$. The magnetic field is applied along the b^* direction (after Ref. 23).

has been suggested that this kink transition is the point at which the external field destroys the low-temperature antiferromagnetic state,¹⁶ but the low-temperature band structure and the field-induced changes giving rise to the kink have remained the subjects of speculation.

Recently, a SDW ground state with a nesting vector $\mathbf{Q} = \zeta \mathbf{A}/6 + \mathbf{C}/6 + (2\eta - 1)\mathbf{B}/6$ has been proposed for $\alpha\text{-ET}_2\text{TiHg}(\text{SCN})_4$ (Ref. 9, 19) [where $\mathbf{A}$, $\mathbf{B}$, and $\mathbf{C}$ are reciprocal lattice vectors of the room temperature crystal structure and η and ζ are ± 1 (Refs. 9, 19)]. The action of the proposed SDW state on the calculated band structure results in a warped quasi-1D FS tilted by 26° with respect to the b^*c plane, plus small 2D pockets [Fig. 1(b)]. We believe that a similar effect takes place in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$.

In this paper, we present the results of a detailed study of the MR as a function of magnetic field and angle in single-crystal samples of $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$. We examine the character of the SdHO's for a range of samples, temperatures, and field regimes and measure the extent to which exchange-enhanced spin splitting and magnetic breakdown involving the various sections of the reorganized FS are important in determining the harmonic structure of the observed SdHO's. Angle-dependent MR oscillations (AMRO's) in magnetic fields up to 30 T in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ have been used to investigate the FS topology both below and above the kink transition. Below the kink, we show that the low-magnetic-field band structure is dominated by a 1D open section of FS inclined at $\sim 21^\circ$ to the crystallographic b^*c plane, agreeing well with the model discussed in Ref. 9; magnetic breakdown between the small 2D pockets and the 1D sheets [Fig. 1(b)] explains the various SdHO frequencies and their anomalous field dependences. Above the kink, the AMRO's change in character, indicating that the FS now possesses a 2D closed section in the form of a distorted cylinder. The data therefore give definitive proof that the kink is the signature of a magnetic-field-induced change in the FS.

This paper is organized as follows: In Sec. II, we describe the experimental techniques. Section III describes the field-dependent resistance of $\text{ET}_2\text{KHg}(\text{SCN})_4$ and discusses the importance of spin splitting. Section IV contains a full description of a series of AMRO measurements below and above the kink transition. The main results of these experiments are discussed in Sec. V and the conclusions are listed in Sec. VI.

II. EXPERIMENT

Single crystals of $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ were grown by electrocrystallization of BEDT-TTF, prepared by the method of Larsen and Lenoir, in a mixture of 90% 1,1,2-trichloroethane and 10% ethanol using KSCN, $\text{Hg}(\text{SCN})_2$, and 18Crown6 as the source of the anion.²⁰ Typical dimensions for the resulting black distorted-diamond-shaped platelets are $1 \times 0.5 \times 0.05 \text{ mm}^3$, with the plane of the plate corresponding to the highly conducting 2D layers. Gold wires were attached to both *ac* platelet faces using platinum paint, resulting in contact

resistance values of less than $\sim 60 \Omega$.

MR measurements were carried out over a wide range of orientations of the crystals in the field using ^3He and ^4He cryostats which allowed the sample to be rotated about two perpendicular axes *in situ* with a precision of $\pm 1^\circ$. The sample orientation was determined to $\pm 1^\circ$ by x-ray techniques and by measuring the polarized infrared reflectivity at room temperature.²¹ Magnetic fields up to 20 T were provided by the Nijmegen Bitter magnets, and the Nijmegen Hybrid II magnet was used to study the MR of $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ up to 30.4 T.²²

Standard four-wire ac techniques (5–150 Hz) were used for all measurements; the current was directed in the interplane b^* direction. Great care was taken to avoid sample heating: The currents used depended on the geometry of the sample and were generally in the range 0.2–20 μA .

III. MAGNETORESISTANCE

A. General features

The magnetoresistance (MR) of $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ shows a number of striking features which may be summarized as follows:

(1) There is a region of strong negative MR (the kink) around 22 T when the field is parallel to b^* (Refs. 8, 9, 15–17, 23, 24) (Fig. 2).

(2) At magnetic fields around and below the kink, there is strong hysteresis in the MR (Refs. 8, 9, 16, 17).

(3) Above the kink, there is one series of conventional SdHO's of frequency, $B_F = 656 \pm 10 \text{ T}$, which are similar in character to those observed in $\alpha\text{-ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$. Below the kink, the oscillations are of a similar frequency, $B_{F1} = 670 \pm 5 \text{ T}$, but more complex, exhibiting a strong second harmonic component at certain angles.^{8,9,15–17,24,25} The frequency of the oscillations is within a few percent of that expected from band structure calculations. In addition, depending on sample quality (Dingle temperature), a second series of SdHO's can be clearly observed below the kink transition in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ and $\alpha\text{-ET}_2\text{TiHg}(\text{SCN})_4$.^{17,26}

(4) The background MR oscillates in size as the sample is tilted in the field.^{9,16}

Observations (1) and (3) will be examined in the context of the proposed FS [Fig. 1(b)] in detail in this section; the fourth, concerning the angular dependence of the MR, is reserved for the following section.

B. Effective mass and scattering

The temperature dependence of the MR in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ is shown in Fig. 3. SdHO's ("quantum" MR) are seen as superimposed upon a ("classical" MR) background; both effects are strongly temperature dependent. Since the amplitudes of the quantum oscillations are scaled by the classical MR (a sample with larger resistance has larger-amplitude quantum oscillations),²⁷ great care must be taken when analyzing the temperature dependence of the oscillations in order to find the effective mass.

FIG. 3. Magnetoresistance of a single crystal of $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ at several temperatures. The temperature in degrees kelvin is marked by each trace. Shubnikov-de Haas oscillations and the field-induced resistive kink at $\sim 22 \text{ T}$ can be clearly seen.

A simple "background subtraction" is insufficient in this case since the background is strongly field and temperature dependent,²⁸ and so the measured data were divided by a fitted field- and temperature-dependent background. An effective mass of $(1.9 \pm 0.1)m_e$ is obtained by a direct fit to the Lifshitz-Kosevich formula and is field independent, as is the Dingle temperature. This is in disagreement with previous reports which estimate a somewhat lower value of $1.4m_e$;^{17,25} this may be due to an incorrect analysis by simple background subtraction rather than background division. Our measured effective masses of the higher harmonics up to third order behave as predicted by the semiclassical theory,²⁹ indicating the validity of the approach. Above the kink transition, the effective mass is measured to be $(2.1 \pm 0.2)m_e$ which is of the same value as below the kink transition within experimental error.

Dingle plots are shown in Fig. 4 for applied fields below and above the kink. At the kink transition, a discontinuous change in slope (parametrized by the Dingle temperature T_D) and, hence, scattering time τ_D (since the effective mass is the same below and above the kink) are observed, corresponding to a change in Dingle temperature which is typically in the range 1.5–2.3 K ($\tau_D \sim 0.5$ – 0.8 ps) below the kink to $\sim 6.5 \text{ K}$ ($\tau_D \sim 0.2 \text{ ps}$) above the kink. This observation is in good qualitative agreement with an unpublished de Haas-van Alphen study.³⁰

C. Spin splitting

In a magnetic field, the Landau levels (themselves separated in energy by $\hbar\omega_c$) can be further split into a set of spin-up and spin-down levels (Zeeman levels), each sepa-

FIG. 4. Dingle plot for oscillations below and above the kink transition. The measured Dingle temperatures are shown in the figure.

rated by $g\mu_B B$. Each set of splittings leads to quantum oscillations of the same frequency²⁹ but a phase difference equal to the field-independent ratio $g\mu_B B/\hbar\omega_c$.

The g factor for α -ET₂KHg(SCN)₄ has been estimated by the harmonic ratio method¹⁶ which utilizes the fact that the Zeeman splitting is angle independent whereas the cyclotron splitting depends on the perpendicular component of the magnetic field. Spin splitting gives rise to a $\cos[p g \pi m^*(\theta)/2m_e]$ reduction factor in the Lifshitz-Kosevich formula²⁹ (where p is the harmonic number). Tilt angle experiments¹⁶ yielded $gm^*(0)/m_e \sim 3.1$ (in the range 16–19 T) so that together with our value $m^*(0) \sim 1.9 m_e$ we deduce a g factor of 1.6.³¹

In the lower half of Fig. 5, we show the oscillatory component of the lowest-temperature MR data in Fig. 3 obtained by dividing by the classical background. The

FIG. 5. The data shown in Fig. 3 at 0.5 K are shown with the background divided out, together with a fit with the Lifshitz-Kosevich formula with the field-dependent g factor $g = 1.55 + 0.3/B$.

Fourier transform of these data [Fig. 6(a), showing data for temperatures between 0.5 K and 1.92 K] shows very high harmonic content: The effective mass of the peak of the second harmonic is twice as large as that of the fundamental, in accordance with the Lifshitz-Kosevich formula.²⁹ Strong spin splitting at such low magnetic fields is not inconceivable if the Zeeman splitting were to be strongly enhanced by a magnetic exchange interaction. The best fit to the Lifshitz-Kosevich formula (summed up to the $p = 2$ term) was obtained by using a magnetic-field dependent g factor²⁵ $g = g_{\infty} + B_{ex}/B$ using the experimentally determined FS parameters, where the exchange field is defined by $B_{ex} \equiv \Delta E/\mu_B$. A good fit is obtained using $gm^* = 1.9(1.55 + 0.3/B)$ [Figs. 5 (upper half) and 6(b)]. These results are in agreement with those of Sasaki and Toyota²⁵ except that their value of B_{ex} is substantially larger. The g factor agrees well with the above results of the harmonic ratio method.

D. Magnetic breakdown

In addition to the fundamental frequency ($B_{F1} = 670$ T) and its second harmonic (both discussed above), we observe other frequency components which we denote $B_{F2} = 150$ –200 T and $B_{F1} + B_{F2} = 820$ –870 T, whose amplitudes and frequencies are slightly sample or cooling method dependent. The ranges quoted for these additional oscillations reflect the variation in B_{F2} from sample to sample. We can relate these additional frequencies to the model of the proposed low-field Fermi surface discussed in the Introduction: B_{F2} corresponds to the small 2D pocket (lens orbit) in Fig. 1(b); B_{F1} corresponds to the larger orbit produced by breakdown and includes the 1D Fermi sheets. All of the frequencies observed have a magnetic field dependence characteristic of a magnetic breakdown network.³² The amplitude of the small FS pocket frequency B_{F2} is large at low magnetic fields; its amplitude decreases steadily above ~ 12 T, while in the same region the amplitudes of B_{F1} and $B_{F1} + B_{F2}$ steadily increase.

In some samples, the amplitudes of the frequencies B_{F2} and $B_{F1} + B_{F2}$ are small relative to that of B_{F1} (e.g., see Fig. 6). In other samples, the amplitudes of all three frequencies are comparable, and strong beating is found in the oscillatory part of the MR (Fig. 7); in these samples, the magnetic field dependence of the amplitude of B_{F1} differs from that predicted by the standard Lifshitz-Kosevich formula.

We believe that the difference between these samples is related to the degree of Landau level broadening induced by sample quality and/or electron heating. We demonstrate this by considering in detail the data presented in Fig. 7 which are shown for the same sample but under different experimental conditions. When the applied current is 4 μ A, the electron heating is small and the Landau level broadening is low. Consequently, the MR is dominated by spin-splitting effects and can be fitted using the same parameters and analysis as that presented above [Fig. 8(a)]. With a larger current of 10 μ A, the spin-splitting features are suppressed, but a strong beating

FIG. 6. (a) The Fourier transform of the data in Fig. 3, the transform being performed after division by the background MR. The temperature is as indicated in degrees kelvin. The two most significant peaks are the fundamental (B_{F1}) and its second harmonic ($2B_{F1}$) which is much larger. (b) The Fourier transform of the fit in Fig. 5 for data at 0.5 K.

pattern is evident. The Fourier transforms of the two cases are shown in Fig. 7(b) and illustrate that the effect of the electron heating is to suppress the first breakdown orbit (B_{F1}) and especially its harmonics (reflecting their relatively large effective mass) while only mildly reducing the amplitude of orbits involving one or more circuits of the small pocket, thereby increasing their relative importance.

The magnetic breakdown orbits which dominate the 10 μ A oscillations can be fitted using a coupled network breakdown model.³³ The proposed FS has a small quasi-two-dimensional section ($B_{F2} = 150$ –200 T) which is separated by a very small gap from a quasi-one-dimensional section. As the magnetic field is increased, electrons may tunnel between adjacent sections of the FS, thus describing a larger k -space orbit which is observable as a higher-frequency series of oscillations ($B_{F1} = 670$ T). Other closed semiclassical orbit combinations are observable such as $2B_{F1} - B_{F2}$ (1140–1190 T) and $B_{F1} + B_{F2}$ (820–870 T). A reasonable fit to the experimental data [Fig. 8(b)] can be thus obtained using the experimentally measured FS parameters and a magnetic breakdown field of $B_0 = 0.15$ T. The effective mass of the B_{F2} orbit could not be measured as its SdHO amplitude is so low.

As a first approximation, the effective mass of an orbit is assumed to scale with its area (so that $m_2^* = 0.6 m_e$). The data imply that the small FS pocket corresponding to B_{F2} is separated by a small energy gap of 9 K from the quasi-one-dimensional sections. The tunneling of electrons across this gap allows the observation of the oscillations of frequency B_{F1} . The gap is so small that breakdown occurs at very low fields, but, as discussed above, is most clearly observable when the Landau level broadening induced by high electron heating or large Din-

FIG. 7. (a) The oscillatory component of the MR at 0.75 K is shown for sample currents of 4 μ A and 10 μ A. (b) The Fourier transform of the data in (a).

gle temperatures reduces the usually dominant effect of the spin splitting. The observed variation from sample to sample in the area B_{F2} of the small 2D pocket may be due to the formation of slightly different SDW ground states, each with a different nesting vector, thus giving rise to a slightly different low-temperature FS, depending on the precise cooling method and/or sample quality.

E. Summary

Below the kink, the SdHO's are of a similar frequency ($B_{F1} = 670 \pm 10$ T) to those observed in α - $\text{ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$ but are more complex, exhibiting a prominent second harmonic at low tilt angles which we attribute to spin splitting. In agreement with Sasaki and Toyota,²⁵ we find that a field-dependent exchange enhancement of the spin splitting due to the antiferromagnetic order can explain the change in oscillation phase with magnetic field. The g factor deduced is in excellent agreement with harmonic ratio data. Depending on sample quality and the value of the current used, other SdHO frequencies are present which have a magnetic field de-

pendence reminiscent of strong magnetic breakdown.³² This is in qualitative agreement with the model of the low-field Fermi surface.

Above the kink, a single series of oscillations ($B_f = 656 \pm 10$ T) grow strongly in amplitude; at very high field (~ 40 T), the data for α - $\text{ET}_2\text{KHg}(\text{SCN})_4$ and α - $\text{ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$ become very similar. The difference in the behavior between the two nominally identical materials at fields below the kink can be attributed to the onset of a spin-density wave (SDW) at temperature below T_N , associated with a pair of 1D sheets nesting together in α - $\text{ET}_2\text{KHg}(\text{SCN})_4$ [Fig. 1(b)]. The high-field data are in qualitative agreement with the Fermi surface in Fig. 1(a), with the MR primarily resulting from the 2D sections of the FS. The kink represents the field-induced transition between the two regimes.

The presence of a SDW which alters the FS at the kink can be most firmly established by angle-dependent measurements; these provide proof of the geometry of the model for the low-field band structure and allow a qualitative test of the proposed changes in the FS at the kink transition; such measurements are the subject of the following section.

IV. ANGLE-DEPENDENT MAGNETORESISTANCE OSCILLATIONS

A. Introduction

MR data have been recorded while single crystals of α - $\text{ET}_2\text{KHg}(\text{SCN})_4$ were rotated in different magnetic fields up to 30 T. In Figs. 9–12 the horizontal axis represents the angle θ between the magnetic field and the b^* axis. Each trace can be labeled with an azimuthal angle ϕ describing the plane of rotation; $\phi = 0^\circ$ represents rotation about the a axis and increasing values of ϕ represent angular displacement of the rotation axis in the ac plane towards the c axis; rotation about the c axis corresponds to $\phi \sim 90^\circ$. The experiment was performed by first setting ϕ at a particular magnetic field; the sample was then rotated through the angle θ in that magnetic field.

There exist two types of angle-dependent MR oscillations (AMRO's) depending on whether the Fermi surface is quasi-1D or quasi-2D.

1. 1D oscillations

Recently, Osada *et al.*³⁴ have proposed a model to explain the dips observed in the resistivity of the organic conductors $(\text{TMTSF})_2\text{X}$ ($\text{X} = \text{ClO}_4, \text{PF}_6$) as a function of the direction of the applied magnetic field. They have calculated the conductivity for a band structure consisting of weakly corrugated quasi-1D sheets across which electrons travel in open orbits. They find that at particular directions of the applied magnetic field there are large resonances in the conductivity when the condition for the existence of periodic orbits of sufficiently small period is satisfied.

This model has recently been applied to α -

FIG. 8. (a) The $4 \mu\text{A}$ data in Fig. 7(a) are plotted against B and can be compared with a fit using a field-dependent g factor using the same parameters used to fit another sample in Fig. 3. (b) The $10 \mu\text{A}$ data in Fig. 7(a) are plotted against B and are shown with a fit to a coupled network model using measured band parameters and a breakdown field of 0.15 T.

$\text{ET}_2\text{KHg}(\text{SCN})_4$.³⁵ If the 1D sections of the Fermi surface are inclined at an angle ϕ_0 to the b^*c plane, then resonances are expected to be observed when³⁴

$$\tan \theta \cos(\phi - \phi_0) = \frac{m}{n} \frac{b}{c}, \quad (1)$$

where m and n are integers. This theory predicts that resistivity dips at fractional values of m/n should be observed, as well as at integer values ($n = 1$), that there should be a missing dip at $m = 0$, and that the oscillations should be symmetric in $\tan \theta$. As shown below, it is found experimentally that this is not the case: Fractional effects are only weakly observed under certain conditions (if at all), there is no missing dip at $m = 0$, and the symmetry in $\tan \theta$ is broken by an offset. These points are inadequately treated in Ref. 35.

To accurately model the data, it is necessary to include the fact that the Fourier components of the corrugations in the 1D Fermi sheets (which correspond to transfer integrals along particular lattice vectors) should be properly defined on an *oblique* (rather than rectangular) lattice since $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ is triclinic, and as will be shown below, the quasi-1D planes cut the Brillouin zone at an angle. The obliquity of this 2D lattice on which the transfer integrals are defined will be parametrized by the quantity d . We therefore consider a weakly corrugated quasi-1D Fermi surface with dispersion relation given by

$$E(\mathbf{k}) = \hbar v_F(|k_x| - k_F) - \sum_{m,n} t_{mn} \cos[(mb + nd)k_y + nck_z], \quad (2)$$

which consists of a pair of sheets at $k_x = \pm k_F$ (for simplicity of description, we have taken $\phi_0 = 0$ so that the 1D planes are perpendicular to the x axis). The t_{mn} are the Fourier components of the sheet corrugation associated with the lattice vector $\mathbf{R}_{mn} = (0, mb + nd, nc)$ and b and c are the size of the unit cell (assumed orthorhombic). We are free to set $t_{00} = 0$ since this term only produces a shift in the Fermi energy. The velocity $\mathbf{v}$ of each electron as a function of momentum $\mathbf{k}$ can then be calculated using $\mathbf{v}(\mathbf{k}) = \hbar^{-1}[dE(\mathbf{k})/d\mathbf{k}]$. This quantity will be time dependent since the electron's momentum $\mathbf{k}$ varies with time according to the equation of motion $\hbar \dot{\mathbf{k}} = -e\mathbf{v} \times \mathbf{B}$ with the magnetic field $\mathbf{B}$ given by $\mathbf{B} = (0, B \sin \theta, B \cos \theta)$. In this paper, we will only consider the effect of the magnetic field in the quasi-one-dimensional plane $\phi = 0$. If there is an out-of-plane component, then one can approximately treat the problem as if the effective (in-plane) magnetic field is $B \cos \phi$. This ignores the fact that the electron motion in the Fermi sheets is no longer along straight lines (this feature will be treated elsewhere³⁶).

The conductivity can be calculated with the Boltzmann transport equation and we find

$$\sigma_{xx} = e^2 g(E_F) v_F^2 \tau \quad (3)$$

and $\sigma_{xy} = \sigma_{yx} = \sigma_{xz} = \sigma_{zx} = 0$, but

$$\begin{pmatrix} \sigma_{yy} & \sigma_{yz} \\ \sigma_{yz} & \sigma_{zz} \end{pmatrix} = \frac{g(E_F) e^2}{\hbar^2} \sum_{m,n} \begin{pmatrix} (mb + nd)^2 & (mb + nd)nc \\ (mb + nd)nc & (nc)^2 \end{pmatrix} t_{mn}^2 \frac{\tau}{1 + (G_{mn} \tau v_F)^2}, \quad (4)$$

where $G_{mn} = \frac{eB}{\hbar} [(mb + nd) \cos \theta - nc \sin \theta]$. A resonance in the conductivity occurs when $G_{mn} = 0$, i.e., when

$$\tan \theta = \frac{mb}{nc} + \frac{d}{c}. \quad (5)$$

2. 2D oscillations

Peaks in the AMRO's of quasi-2D materials have been treated elsewhere^{37,38} and are connected with the vanishing of the electronic group velocity perpendicular to the 2D layers. The angles θ_i at which the maxima occur are given by $b' k_{\parallel} \tan(\theta_i) = \pi(i \pm 1/4) + A(\phi)$, where the signs $-$ and $+$ correspond to positive and negative θ_i respectively, b' is the effective interplane spacing, $k_{\parallel}$ is the maximum Fermi wave vector projection on the plane of rotation of the field, and $i = \pm 1, \pm 2, \dots$.³⁷ Here positive i correspond to $\theta_i > 0$ and negative i to $\theta_i < 0$.³⁷ The gradient of a plot of $\tan \theta_i$ against i may thus be used to find one of the dimensions of the FS and, if the process is repeated for several planes of rotation of the field, the complete FS may be mapped out. $A(\phi)$ is determined by the inclination of the plane of warping; hence this may also be found.³⁷

B. Experimental data

In Fig. 9, we show data from a low-field experiment on $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ at 1.5 K. Strong AMRO dips are seen which become more pronounced as the field increases.

FIG. 9. Angle-dependent MR in $\alpha\text{-ET}_2\text{KHg}(\text{SCN})_4$ at 1.5 K for magnetic fields in the range 1–15 T. The azimuthal angle ϕ is set to 0° .

Although the dips become sharper and the peaks flatter with increasing field, the positions of the dips do not change. Figure 10 shows data from an experiment on a different sample in fields up to 19 T as θ is varied in the full range between -90° and 90° . Notice that the traces are asymmetric in θ ; there is no perfect mirror reflection symmetry about $\theta = 0$ as one might expect from a straightforward application of the theory in (Ref. 34); as described above, the asymmetry in the AMRO traces is due to the obliquity of the 2D lattice whose points correspond to Fourier components of the corrugation of the Fermi sheets.

In Fig. 11(a), AMRO data at 15 T are shown as θ is varied for a range of values of ϕ . Notice that when $\theta = 0^\circ$, the magnetic field is parallel to b^* independently of the value of ϕ , so that these points show identical resistance; each trace has been offset for clarity. $\phi = 0^\circ$ corresponds to rotation in the b^*c plane; $\phi = 90^\circ$ corresponds to rotation in the b^*a plane. The minima in the MR are very sharp and are periodic in $\tan \theta$ at each azimuthal angle ϕ , although the period is a function of ϕ .

The periodicity of the minima (deduced from a plot of minimum index versus $\tan \theta$) can be plotted against the azimuthal angle ϕ [Fig. 12(a)] and is seen to vary as $(1.25 \pm 0.05) / \cos(\phi - \phi_0)$, where we find $\phi_0 = -21^\circ \pm 3^\circ$. Thus the maximum amplitude and minimum periodicity in $\tan \theta$ occur when the field rotation plane forms an angle $21^\circ \pm 3^\circ$ with the b^*c plane. A polar plot of the positions of the resistance minima shows a characteristic family of lines inclined at this angle to the vertical [Fig. 12(a), inset]. This corresponds to a 1D sheet tilted with respect to the b^*c plane by this angle, in reasonable qualitative agreement with the model for the low-temperature band structure discussed in the Introduction, which predicts a one-dimensional plane tilted by 26° .⁹ Similar behavior is

FIG. 10. Angle-dependent MR in $\alpha\text{-ET}_2\text{KHg(SCN)}_4$ at 1.5 K for magnetic fields in the range 7–19 T for θ in the full range $-90^\circ < \theta < 90^\circ$. The azimuthal angle ϕ is set to 0° . Note the asymmetry in the traces.

observed in all of the samples studied in this work and in a majority of samples examined by other workers.³⁹

Before turning to higher-field data, two further points should be noted. In this field region, below the kink, the only effect of increasing the magnetic field is to make the AMRO dips more pronounced (Figs. 9 and 10); there is no evidence for a FS transition resulting from a phase boundary in the low-field region (below 10 T) of the type recently suggested by some authors.⁴⁰ Furthermore, hysteresis is observed in both swept-field and swept-angle (AMRO) experiments below the kink, but appears to vanish at AMRO minima. This interesting effect will be the subject of a future paper.

As the magnetic field rises through the kink, a dra-

FIG. 11. AMRO's in $\alpha\text{-ET}_2\text{KHg(SCN)}_4$ at 1.5 K for several values of the angle ϕ at an applied field of (a) 15 T and (b) 24 T.

matic change in the AMRO's is observed. This can be seen (for a fixed value of ϕ) for magnetic fields in the range 17–30 T in Fig. 13.^{41,42} The AMRO's change from a series of sharp dips to a series of sharp peaks. A measurement of the ϕ dependence of these peaks at 24 T [Fig. 11(b)] shows that they are observable with similar amplitudes at all azimuthal angles and their position in θ shows much weaker dependence on ϕ than the dips observed below the kink,¹⁸ strongly suggesting that the mechanism responsible for the peaks is quasi-2D in nature. Note that the kink transition is angle dependent (for a detailed discussion see Ref. 18) so that, for example, at 24 T it is still possible to find 1D oscillation minima near to $\theta = 90^\circ$ [Fig. 11(b)].

The 2D peaks in the AMRO data at 24 T can be used to determine the 2D FS at high fields: The gradient of a plot of $\tan \theta_i$ against i can be used to find the dimension

FIG. 12. Analysis of the ϕ and θ dependence of the AMRO's (a) minima at 15 T and (b) maxima at 24 T. The inset in (a) shows a polar plot of the θ and ϕ values of the resistance minima. The main figure shows a fit of $1.25/\cos(\phi - \phi_0)$ to the observed ϕ dependence of the period in $\tan \theta$ of the minima. This fit was used to determine the dotted straight lines in the inset and corresponds to a 1D Fermi sheet inclined at $\sim 21^\circ$ to the b^*c plane. A similar result can be obtained for the 1D minima in the 24 T data but, in the range $-60 \lesssim \theta \lesssim 60$, the 1D minima vanish and 2D maxima appear. For each ϕ , the positions of the minima can be fitted to $b^*k_{\parallel} \tan(\theta_i) = \pi(i \pm 1/4) + A(\phi)$ [an example fit at $\phi = -95^\circ$ is shown on the left side of (b)] so that the shape of the 2D Fermi surface can be mapped out [(b), right].

FIG. 13. Evolution of the peaks in the angle-dependent MR of α -ET₂KHg(SCN)₄ at 1.5 K with increasing magnetic field in the range 17–30 T. The sharp MR minima characteristic of quasi-1D AMRO's give way to pronounced maxima characteristic of quasi-2D AMRO's. This reflects the proposed magnetic-field-induced change in the FS.

of the FS in one direction; if the process is repeated for several planes of rotation, the complete FS shape may be mapped out. For the 24 T raw data,¹⁸ a FS shape [Fig. 12(b)] is derived which is of roughly the correct area to account for the frequency of the SdHO's above the kink. A 2D hole pocket of approximately this size is also predicted by band-structure calculations,⁶ although our derived FS is more elongated.

V. DISCUSSION

The sharp change in AMRO behavior near the kink can be attributed to the transition between a FS dominated by quasi-1D sections to one dominantly quasi-2D. Thus we have observed a direct 1D $\rightarrow$ 2D transition in the FS induced by a magnetic field. The same transition in FS's occurs with heating through T_N : A recent experiment on α -ET₂KHg(SCN)₄ at 14 T showed that as the temperature was raised through 8 K the AMRO's changed in character, indicating that the FS has changed from predominantly 1D to predominantly 2D in form.⁴³ Our preliminary measurements of AMRO's on α -ET₂KHg(SCN)₄ as a function of temperature support this observation.⁴⁴

The AMRO and SdHO results lead us to conclude that for magnetic fields above the kink, or for temperatures $T > T_N$, the FS is similar to that in Fig. 1(a), and by implication similar to that in α -ET₂NH₄Hg(SCN)₄, whereas the rearranged low-temperature FS below the kink is in qualitative agreement with that in Fig. 1(b), consisting of a tilted 1D section and a small closed pocket (corresponding to the SdHO frequency B_{F2}). However, it should be noted that the AMRO data for α -ET₂KHg(SCN)₄ at fields below the kink imply a 1D FS sheet tilted by 21° with respect to the b^*c plane, whereas the model shown in Fig. 1(b), derived for α -ET₂TiHg(SCN)₄, predicts a one-dimensional plane tilted by 26° .⁹ This suggests that the SDW nesting vectors in α -ET₂KHg(SCN)₄ and α -ET₂TiHg(SCN)₄ are differ-

ent. It has been pointed out that the operation of the SDW nesting vector implied by the 21° FS sheet angle in α -ET₂KHg(SCN)₄ on the calculated band structure would not result in any small pockets such as those in Fig. 1(b).^{45–47} However, the presence or absence of small pockets in the rearranged band structure depends on the exact shape of the two-dimensional section in the original FS; as observed in the experiments reported here and in other works,^{12,43} this seems more elongated than the almost circular cross sections suggested by the band-structure calculations [Fig. 1(a)].

No AMRO's due to open sections of the FS are seen at fields above the kink in α -ET₂KHg(SCN)₄ [and also in α -ET₂NH₄Hg(SCN)₄]. We suggest that the reason for this is due to the fact that these open sheets are comparatively flat and have low harmonic content. In contrast, for α -ET₂KHg(SCN)₄ below the kink, and at temperatures below T_N , due to the folding of the 2D pocket caused by the action of the SDW, the resulting 1D sheets are strongly warped with high harmonic content. This large corrugation of the 1D sheets permits the observation of large AMRO's.

In a recent paper, Sasaki and Toyota have measured AMRO's and the current-direction dependence of the MR in α -ET₂KHg(SCN)₄; they have also attempted to provide a structural motivation for the nesting vector which they assume for this material by relating the symmetry of the real-space crystal structure to the proposed low-temperature band structure.⁴⁸ This explanation accords well with their measured 1D FS tilted by 30° , but this tilt angle is substantially higher than that measured in this work and also than that reported by Kartsovnik *et al.*³⁹

Because the nesting vectors in α -ET₂KHg(SCN)₄ and α -ET₂TiHg(SCN)₄ are almost incommensurate, the SDW may possibly be poorly locked in to a particular orientation, so that slight changes in cooling or differences in sample quality might slightly alter the angle of the 1D FS observed by AMRO's or give rise to the variation in the area of the closed pocket and hence the observed spread in the values of B_{F2} .

Additional low-frequency SdHO's have been observed in experiments carried out at very low temperatures;⁴⁰ these oscillations were attributed to a further rearrangement of the FS at low fields.⁴⁰ As discussed above, the field independence of the positions of the AMRO minima at fields below the kink does not support this conclusion. The low-frequency SdHO's observed are instead likely to be due to vestigial imperfectly nested parts of the 1D FS.⁴⁹ An analogous situation is known to occur in β'' -ET₂AuBr₂, where SdHO frequencies due to small pockets left by imperfect nesting of a SDW are clearly seen.⁵⁰ The FS in Fig. 1(b) is made up of a rearrangement of the 2D pocket in Fig. 1(a) only (the 1D section is assumed to nest), as evidenced by the B_{F1} mass being the same on either side of the kink transition; imperfect nesting would lead to additional very small FS pockets.

Recent experiments have used a field modulation technique to give enhanced detection of high-frequency SdHO's in α -ET₂KHg(SCN)₄; a frequency was observed which appeared to correspond to a breakdown orbit with

the same area as the whole, unrearranged, Brillouin zone [i.e., Fig. 1(a) of Ref. 51]. Uji *et al.*⁵¹ took this to imply that the FS in their samples was not rearranged, but identical to that at high temperatures [Fig. 1(a)]; a breakdown orbit involving tunneling between the two- and one-dimensional sections would have the same area as the Brillouin zone. A similar frequency (at about 4300 T) is observed in Fourier spectra of the MR of samples studied in this work (see Fig. 14); as the other data discussed above seem to unambiguously indicate that the low-temperature FS is radically rearranged from its high-temperature form, two possible reasons for the presence of the “whole Brillouin zone” frequency are suggested.

(1) It is possible that the peak in the Fourier spectrum corresponds to a mixed harmonic of the other SdHO frequencies ($6B_{F1} + B_{F2}$); several other mixed harmonics of this kind are present in Fig. 14 and in the data of Uji *et al.*⁵¹ Recent work on β'' -ET₂AuBr₂, which also possesses a SDW ground state at low temperatures, has shown that such mixed harmonics are enhanced by the Shoenberg magnetic interaction.⁵⁰

(2) The low-temperature ground states of α -ET₂MHg(SCN)₄ (M =K, Rb) are very sensitive to hydrostatic pressure or uniaxial stress.^{14,47} It has therefore been suggested that small strained regions of samples (due to contacts, adhesive, etc.) could retain a Fermi surface characteristic of the high-temperature phase;⁴⁵ these small regions could be responsible for the “whole Brillouin zone” frequency.

It has also been suggested⁵¹ that magnetic breakdown is responsible for the whole form of the magnetoresistance. However, magnetic breakdown is a very tiny effect in the classical MR, and the enormous MR effects such as AMRO's that are observed in α -ET₂KHg(SCN)₄ must be due to the gross features of the FS, like the presence of the 1D sheets. The model we have described of the FS rearrangement at the kink transition adequately accounts for the observed properties without having to invoke further exotic effects or appeal to sample inhomogeneities.

FIG. 14. The Fourier transform of the magnetoresistance of α -ET₂KHg(SCN)₄ below the kink at 0.5 K plotted on a logarithmic scale to show high-order harmonics.

VI. CONCLUSIONS

We have performed MR measurements as a function of temperature, field strength, and field direction on single-crystal specimens of the charge-transfer salt α - $\text{ET}_2\text{KHg}(\text{SCN})_4$. Below the kink transition in field, the SdHO's are of a similar frequency to those observed in α - $\text{ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$ but are more complex, exhibiting a prominent second harmonic at low tilt angles due to spin splitting. We find evidence of a field-dependent exchange enhancement of the Landau level splitting due to the antiferromagnetic order which can explain the change in oscillation phase with magnetic field. Depending on sample quality and the value of the current used, other SdHO frequencies are present which have a magnetic field dependence which can be fitted using a coupled network model to describe magnetic breakdown. The AMRO's are 1D in character and indicate the presence of a quasi-1D section of the FS inclined at an angle of $21^\circ \pm 3^\circ$ to the b^*c plane. Above the kink, a single series of oscillations grows strongly in amplitude and at high field (~ 40 T) is very similar to those observed in α - $\text{ET}_2\text{NH}_4\text{Hg}(\text{SCN})_4$; the AMRO dips observed below the kink give way to sharp peaks which are fairly independent of azimuthal angle and indicate a FS with predominantly quasi-2D character.

The difference in the behavior between the two very similar materials at low fields can be attributed to the

onset of a SDW in α - $\text{ET}_2\text{KHg}(\text{SCN})_4$ at temperatures below T_N , associated with the nesting of a pair of 1D sheets. This difference in behavior is perhaps related to the volume of the unit cell, and hence to the warping of the 1D sheets. The size of the unit cell is related to that of the Brillouin zone and hence the 2D hole pocket area, which is $\sim 14\%$ of the Brillouin zone when $M=\text{NH}_4$ (Refs. 52, 53) and 16% for $M=\text{K}$ (Refs. 15, 17, 16). This trend continues for $M=\text{Rb}$ which has a slightly larger 2D hole pocket than the K salt, a transition to antiferromagnetic order at ~ 10 K, and a kink at ~ 35 T;⁵⁴ in other words the SDW ground state appears to be more stable. This can be related to the increasing flatness of the 1D sheets which accompanies the increase in the size of the hole pocket; this favors nesting and hence the SDW ground state.

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