Novel Interplay of Fermi-Surface Behavior and Magnetism in a Low-Dimensional Organic Conductor

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(Received 6 April 1992)

We have used the magneto-oscillatory behavior of the quasi-two-dimensional orbits in the charge-transfer organic salt (BEDT-TTF)$_2$KHg(SCN)$_4$ to probe its underlying magnetic ground states, which arise from the coexisting quasi-one-dimensional nature of the material. The complex nature of the oscillations results from the field dependence of these states. Our results allow comparison between several competing models which have attempted to describe the unusual properties of this material.

PACS numbers: 74.70.Kn, 71.25.Hc, 75.30.Kz

The chemistry and symmetry of anisotropic organic conducting single crystals permit a variety of open and closed orbit configurations in a quasi-two-dimensional Fermi-surface representation [1]. In this Letter we present key results from a comprehensive investigation of the material (BEDT-TTF)$_2$KHg(SCN)$_4$, which is known to have both closed hole and open electron orbits [2]. We have used the magneto-oscillatory behavior [Shubnikov-de Haas and de Haas–van Alphen (SdH and dHvA) effects] of the closed orbits to probe the nature and phase boundaries of the underlying magnetic phases, which apparently arise from the coexisting quasi-one-dimensional character of the material. We have studied pressure and angular dependence over a large range of temperature and magnetic field. Our results support the existence of a SDW phase below 8 K and a new magnetic phase at very high fields below 1 K. We observe that pressure removes the SDW phase. This investigation was carried out at the Francis Bitter National Magnet Laboratory (FBNML).

The title compound is one of the class of highly two-dimensional organic conductors based on the charge transfer system (BEDT-TTF)$_2X$, where the choice of X and the details of the crystal structure provide a wide range of quasi-two-dimensional normal-state (Fermi surface) and superconducting properties [3]. Although the isostructural compound α-(BEDT-TTF)$_2$(NH$_4$)Hg(SCN)$_4$ is superconducting [4] below 1.15 K, (BEDT-TTF)$_2$KHg(SCN)$_4$ is metallic at least down to 0.1 K. The calculated band structure and Fermi surface [5] in the most conducting a-c plane has both two-dimensional hole closed orbits and one-dimensional electron open orbits. Our results suggest finite dispersion of the hole orbits in the least conducting (b) direction.

There has been considerable experimental attention paid to this material since its synthesis and the initial discovery of unusual magnetoresistance (MR) behavior and SdH oscillations at low temperatures including a "kink" above about 22.5 T [6]. Our initial work established the existence of double SdH and dHvA oscillations which were first interpreted in terms of spin splitting of the Landau levels [7]. In Fig. 1 many of the important features of the magnetoresistance and magnetization are shown for the case of the lowest temperatures of our investigation. Further work confirmed the splitting behavior [8] and an interpretation in terms of Landau levels split by the $g\mu_B H$ term with a further shift due to antiferromagnetic exchange was presented. This model (model A) is appealing because it allows for the change in the relative positions of the spin-split levels with magnetic field, and for different scattering rates (and therefore different wave forms) for the two spin directions (see also Higgins and

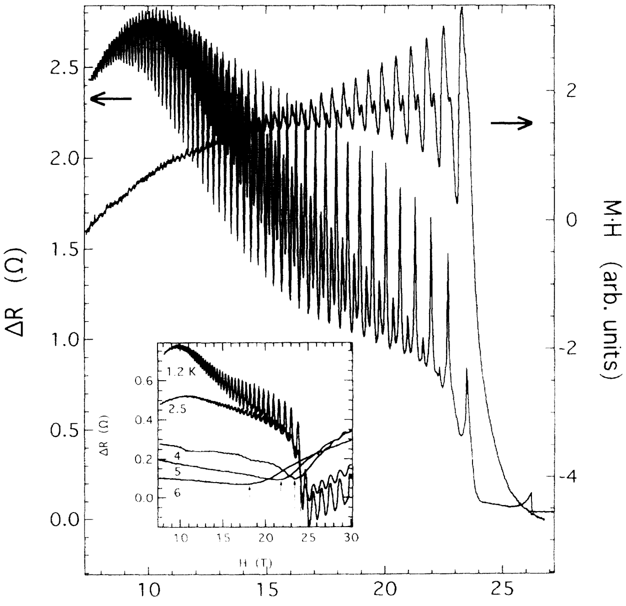

FIG. 1. Magnetoresistance and magnetization measurements taken in the Hybrid magnet at FBNML with a background field of 7.5 T, at 0.10 K. Inset: Temperature dependence of the kink field H_K.

Lowndes [9]). An alternative explanation (model B) for the split level behavior has been provided by Kang [10], who suggests that for a two-dimensional Fermi surface the splitting will result when the cyclotron energy $\hbar\omega_c$ is comparable to the bandwidth $4t_b$ in the least conducting direction. Recently, Pratt *et al.* [11] suggested (model C) that there is a SDW nesting along the *b* direction, which doubles the unit cell thereby creating *two* closed orbits in the new reduced zone. They further suggest that the kink feature in the magnetoresistance is associated with a transition between antiferromagnetic and ferromagnetic order. Cyclotron resonance work by the same group [12] indeed shows two resonant features for the material. Finally, application of the Lifshitz-Kosevich (LK) formalism (model D) is also relevant since it successfully predicts the magneto-oscillatory behavior of at least two related quasi-two-dimensional (Q2D) materials [13].

The onset of magnetic order and unusual Fermi-surface character in (BEDT-TTF)$_2$KHg(SCN)$_4$ below 8 K has been established by several works. The susceptibility below 8 K is highly anisotropic [8]: $\chi_{B\parallel a\text{-}c}$ drops rapidly towards the core diamagnetic limit, indicating in-plane antiferromagnetic order; $\chi_{B\parallel b}$ shows a gradual paramagnetic rise. Further studies of the temperature-dependent resistivity [14] show a shoulder at 8 K. In the present work we studied the shoulder at several pressures. By 8 kbar, the shoulder disappears. Such behavior is characteristic for organic materials where SDW phases are suppressed in favor of metallic phases at high pressure. Additional evidence for a substantially different character of the material at low temperatures comes from observation of the reversal of the resistivity extrema (minima instead of maxima) in the effect described by Yamaji [15], below 8 K. The origin of the magnetic order may arise from nesting of the open-orbit part of the Fermi surface along the *a* axis, as is observed in the quasi-one-dimensional Bechgaard salts [16]. Here the resulting SDW phases are highly dependent on temperature, magnetic field, and pressure. Or as Pratt *et al.* describe [11], the nesting could be in the *c* direction.

We have made extensive resistance and magnetization measurements on (BEDT-TTF)$_2$KHg(SCN)$_4$ over a broad range of temperature, magnetic field (including angular dependence), and pressure. The magnetic phase diagram which has evolved from our measurements and those of other workers is shown in Fig. 2. Zero-field resistivity and low-field susceptibility measurements establish the point near 8 K. Systematic magnetoresistance measurements by our group in steady state fields and also by the Institute of Solid State Physics (ISSP) group in pulsed fields [17] establish the temperature dependence of the kink field $H_K(T)$. Lower-temperature measurements indicate the onset of a hysteretic region below about 1 K. The phase enclosed is most likely an antiferromagnetic SDW state. Above the hysteretic region at low temperatures there appears to be a new magnetic phase with a character distinctly different from the SDW phase.

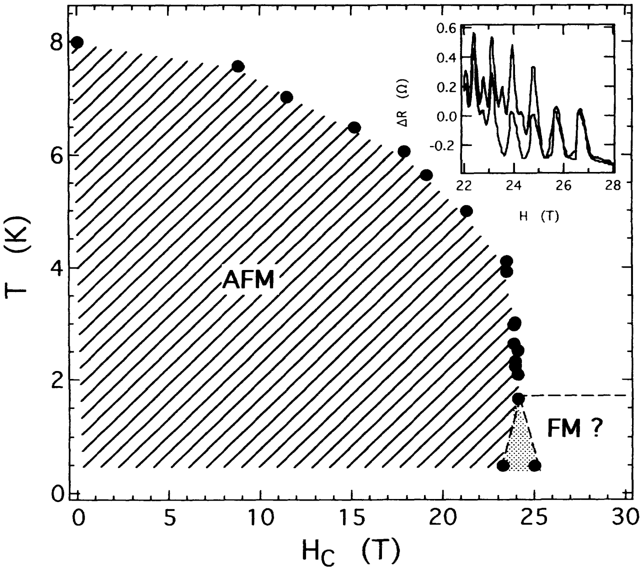

FIG. 2. Proposed *T-H* phase diagram of (BEDT-TTF)$_2$KHg(SCN)$_4$. The dot near zero field denotes the resistance shoulder; dots above 5 T denote this work. Hatched region, SDW phase; shaded region, hysteretic behavior. The region where we expect the high-field anomalous phase to be present is also shown. The inset details the hysteretic MR and SdH behavior near H_K.

We now consider the behavior of the resistance and magnetization.

Magnetoresistance.—The low-field magnetoresistance increases most likely because of the closed orbits. Above 10 T the magnetoresistance decreases and at low temperatures drops precipitously at H_K. If the quasi-one-dimensional (Q1D) FS nesting is reduced with increasing field, and even removed at H_K, this would account for the decrease.

SdH effect.—Oscillations start at about 8 T and the splitting of the fundamental is evident above 10 T. The relative position of the secondary peaks changes smoothly from being just on the high-field side of the main peaks, to being evenly spaced between the main peaks at H_K. Above the kink the spacing is observed to shift abruptly, with the secondary peaks forming a "shoulder" on the high-field side of the main peaks. At low temperatures the oscillations above the kink are dominated by the main peaks, and at the lowest temperatures all oscillatory behavior above the kink is highly suppressed. The magnetoresistance and oscillatory behavior both show considerable hysteresis near and above the kink field. If we apply model B to any pair of oscillatory peaks just below H_K (see for instance Fig. 1) we can estimate the separation of the extremal orbits in energy. Here the Landau level spacing corresponds to 1.99 meV for an effective mass $m^* = 1.4m_0$; m_0 is the bare electron mass. For an SdH oscillation frequency of 670 T, the Fermi energy is 55.4 meV. The ratio of the two energies is 0.018 in this case, which compares favorably with the value 0.015 from Ref. [7] based on the beating of the second harmonic (here interpreted as the warping of the FS).

Magnetization and dHvA effect.—The magnetization in our case is a dc torque measurement [18]. Here we find evidence for split levels as well, and at low temperatures there is a shift in the character of the oscillations at 22.5 T. Above 24 T there is a pronounced change in the background moment which is accompanied by a very strong suppression of oscillatory behavior.

We next turn to the angular dependence [6]. The period of *both* the main and secondary oscillations follows the expected 2D Fermi surface $1/\cos(\theta)$ dependence, where θ is the angle between the magnetic field and the normal to the conducting planes. Examples of the behavior are shown in Fig. 3. However, an analysis of the main and secondary peak heights and wave forms indicates that the linewidth (scattering) is enhanced at *lower* magnetic fields for increasing angle, and that the secondary (split) peak linewidth increases much more than the main peak with increasing angle. In the magnetization, the angular dependence is quite remarkable. The anomalous magnetic signal develops a very wide range with angle, and it is clear that the oscillations exist above and below the onset of this magnetic transition. For angles near 90°, we have observed what appears to be a spin-flop transition near 16 T, with very little hysteresis.

Finally, we discuss the pressure dependence. The pressure was applied with a standard pressure clamp at the values 1, 4, and 8 kbar. The magnetoresistance at 0.5 K for each case is shown in Fig. 4. At 1 kbar the zero-field resistance shoulder and the general form of the magnetoresistance and split level oscillation behavior were present. However, at 4 kbar the zero-field shoulder was highly suppressed, the kink behavior appeared at lower fields, and there was no evidence for splitting of the levels, only a beat behavior near 10 T. At 8 kbar, the shoulder was gone, the magnetoresistance was completely monotonic (no kink), and at the highest fields there was evidence for a slight increase in the magnetoresistance and clear evidence for the onset of level splitting above 21 T, which may be field induced. We note a dependence of the Fermi surface area on pressure, $d\ln(A)/dP = 0.019$ kbar^{-1}, which exceeds the value in copper [19] by more than an order of magnitude.

The measurements described above allow us to draw several conclusions about the low-temperature, field-dependent phase diagram of this material within the main region of the phase diagram of Fig. 2 below H_K. It is evident that below 8 K a magnetic phase is stabilized, which is most likely of an antiferromagnetic SDW form. The nesting which produces the SDW phase may be in the open orbits in the $\mathbf{k}_a$ direction, or due to the 1D periodicity in the least conducting direction $\mathbf{k}_b$. For an applied pressure of 8 kbar, the SDW state is removed. At ambient pressure, in the SDW state, there is a splitting of the Landau levels which may be the result of one or more of the four models mentioned above. The angular-dependent data would appear to rule out model A, since we find that the split levels both obey a $1/\cos(\theta)$ dependence with field, which contradicts the isotropic form expected for the Zeeman and exchange terms [8]. The angular dependence is consistent with model B, but the fact that the pressure removes the splitting for a relatively low change in the closed orbit area, and removes the ambient pressure SDW phase, makes it less likely to explain the

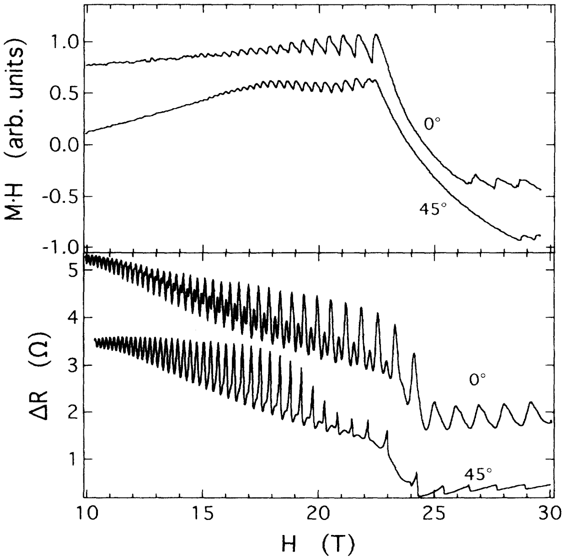

FIG. 3. Angular-dependent magnetization (top panel) (torque signal) and magnetoresistance (bottom panel) for several angles at 0.5 K.

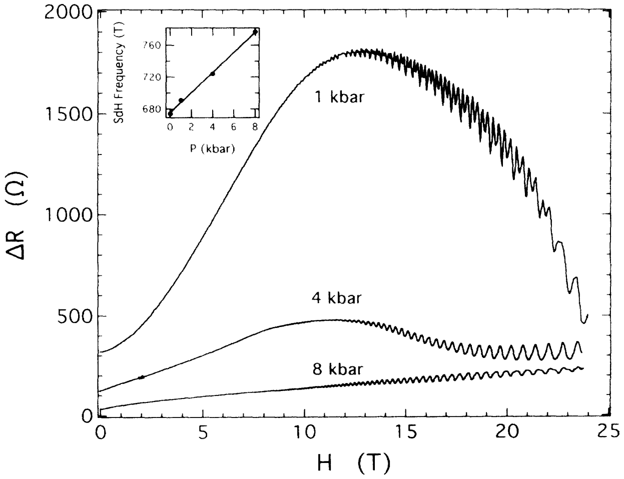

FIG. 4. SdH oscillations and magnetoresistance for different pressures at 0.5 K. Note that only the magnetoresistance change is presented, and that the traces are offset for clarity. Inset: Variation of SdH frequency with pressure.

experimental details. Similarly the anomalous Yamaji effect cannot be explained by this model alone. As noted below, model B should, however, be applicable near the quantum limit. In contrast, model C does support the experimental facts most closely. It satisfies the $1/\cos(\theta)$ dependence of the split levels and allows for a systematic shift in the relative positions of the peaks (i.e., two close but independent orbits); the more complex nature of the double FS in the SDW state may explain the negative Yamaji effect below 8 K; and when the nesting is removed with pressure it restores the single warped FS with beating of the SdH signal far from the quantum limit. Finally, we have considered model D, and have done LK simulations on our data. Below H_K we obtain a fair representation of the oscillatory behavior including the splitting, harmonic ratio, and node near H_K for values of the parameters $m_c^* = 1.4m_0$ (Ref. [6]), $m_b^* = 0.4m_0$, $g = 2.014$ (Ref. [20]), $T_D = 1.5$ K, where m_c^* is the cyclotron mass, m_b^* is the band mass, g is the electron g factor, and T_D is the Dingle temperature. Inclusion of two frequencies, and a field- and spin-dependent Dingle temperature, brings the simulation closer to the data. This analysis will be described more extensively in a future paper [21]. However, the simulation fails to provide for the field dependence of the relative positions of the split peaks. We conclude that the LK simulation is relevant, but that some modification of symmetry of periodicity (*which does not affect the fundamental closed orbit area*) arising from the Q1D character dramatically affects the parameters of the LK model and the background magnetoresistance.

We believe that there is a new magnetic state at low temperatures above the kink, distinct from the SDW state. Investigation of the relative position of the split levels below and above the peak shows a change, which in light of model C, would indicate a change in the nesting. Furthermore, we observe strong hysteresis effects in both SdH and dHvA signals, both of which show anomalous behavior above H_K, and both of which show a strong alteration of oscillatory behavior. In the SdH signal, we see highly distorted, hysteretic wave forms with a strong angular dependence. In the dHvA signal, there is a dramatic change in the background signal. It is quite possible that the kink field represents a transition between an antiferromagnetic phase to a spin-flop or ferromagnetic phase. The very strong field and angular dependence of the secondary oscillation in the SdH signal would indicate that it is the one stabilized by the nesting, and the first to be affected when the nesting either changes or is destroyed as H_K is approached. The appearance of a splitting of the levels above the kink field, but with a distinctly different relative position and amplitude ratio (nearly the same) may indeed arise from the mechanisms in model B if the b-axis nesting has been removed.

In conclusion, we believe that the Q1D nesting (SDW) is field and temperature dependent, and that it accounts for the phase diagram. Furthermore, the magneto-oscillatory behavior associated with the Q2D orbits can be described by the LK formalism with the inclusion of LK parameters which depend on the details of the underlying SDW behavior. In particular, we expect that there are two extremal orbits, and that the Dingle temperature is magnetic field dependent.

This work was supported under NSF Grant No. DMR-88-18510. The authors would like to thank Russ Datars and Paul Chaikin for very helpful discussions. The high-field work was carried out at the Francis Bitter National Magnet Laboratory (supported by the NSF DMR).

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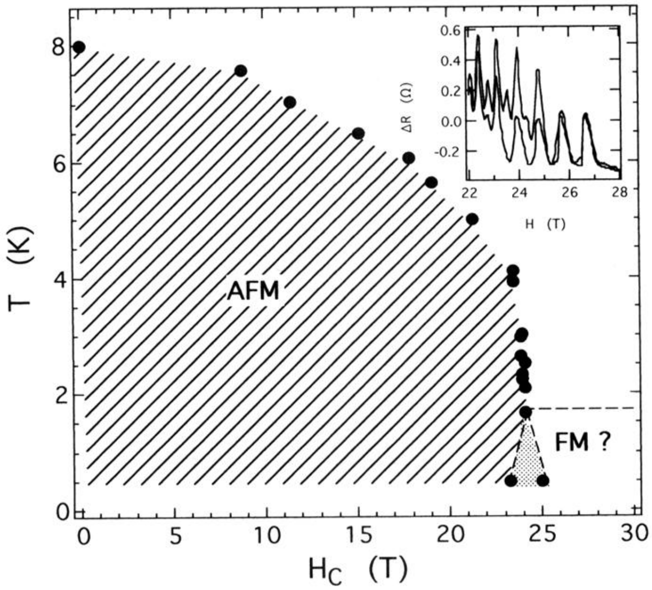

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