Violation of Kohler's rule by the magnetoresistance of a quasi-two-dimensional organic metal

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The interlayer magnetoresistance of the quasi-two-dimensional metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is considered. In the temperature range from 0.5 to 10 K and for fields up to 10 T the magnetoresistance has a stronger temperature dependence than the zero-field resistance. Consequently Kohler's rule is not obeyed for any range of temperatures or fields. This means that the magnetoresistance cannot be described in terms of semiclassical transport on a single Fermi surface with a single scattering time. Possible explanations for the violations of Kohler's rule are considered, both within the framework of semiclassical transport theory and involving incoherent interlayer transport. The issues considered are similar to those raised by the magnetotransport of the cuprate superconductors. [S0163-1829(98)13219-8]

Currently a great deal of attention is being paid to the large magnetoresistance of layered materials such as magnetic multilayers[1] and manganese perovskites.[2] This is motivated by potential applications in magnetic recording and by the challenge of understanding the physical origin of the magnetoresistance, which is very different from that in conventional metals.[3] The magnetotransport of the metallic phase of the cuprate superconductors also differs significantly from conventional metals.[4–6] In this paper we show that the magnetoresistance of a particular organic metal may also be unconventional.

Layered organic molecular crystals based on the bis-(ethylenedithia-tetrathiafulvalene) (BEDT-TTF) molecule are model low-dimensional electronic systems.[7,8] The family α-(BEDT-TTF)$_2M$Hg(SCN)$_4$[M=K,Rb,Tl] have a rich phase diagram depending on temperature, pressure, uniaxial stress, and magnetic field: metallic, superconducting, and density-wave phases are possible.[9–11] Band-structure calculations predict coexisting quasi-one-dimensional (open) and quasi-two-dimensional (closed) Fermi surfaces.[12] At ambient pressure these materials undergo a transition at a temperature T_{DW} (8 K in the M=K salt) into a low-temperature metallic phase that has been argued to be a density wave (DW). This phase is destroyed in high magnetic fields. There is currently controversy as to whether this phase is a spin-density wave, a charge-density wave, or a mixture of both.[9,13–16]

The following picture of the low-temperature phase has been proposed.[17,18] The nesting of the quasi-one-dimensional Fermi surface leads to a density-wave instability at T_{DW}. Below T_{DW} a gap opens on the quasi-one-dimensional Fermi surface and the associated carriers no longer contribute to the transport properties. The density wave introduces a new periodic potential into the system resulting in reconstruction of the quasi-two-dimensional Fermi surface. One of the proposed Fermi surface reconstructions involves large open sheets.[17] Semiclassical transport theory can then explain the large magnetoresistance and its angular dependence in the low-temperature phase.[18] The complete field dependence of the resistance can also be explained if magnetic breakdown is taken into account.[19] However, in this paper we show that the temperature dependence of the magnetoresistance is inconsistent with the above picture. In particular, the magnetoresistance is shown to violate Kohler's rule,[20] raising issues similar to those considered for the cuprate superconductors.[4–6]

The temperature and field dependence of the magnetoresistance of many metals can be analyzed in terms of Kohler's rule.[3] Semiclassical transport theory based on the Boltzmann equation predicts Kohler's rule to hold if there is a single species of charge carrier and the scattering time τ is the same at all points on the Fermi surface. The dependence of the resistance on the field is then contained in the quantity $\omega_c\tau$, where ω_c is the frequency at which the magnetic field B causes the charge carriers to sweep across the Fermi surface. Since the resistance in zero field is proportional to the scattering rate, the field dependence of the magnetoresistance of samples with different scattering times (either due to different purity or temperature T) can be related by rescaling the field by the zero-field resistance $R(0,T)$:

$$\frac{R(B,T)}{R(0,T)} = F(\omega_c\tau) = f\left(\frac{B}{R(0,T)}\right). \tag{1}$$

This is Kohler's rule and the corresponding plots are known as Kohler plots. It holds regardless of the topology and geometry of the Fermi surface.

Resistance measurements were performed on a single crystal of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ using a standard four-wire ac technique with a 10-μA current along the **b** axis (the least conducting axis). Sample contacts were made on the faces of the **a-c** planes with 12.5-Mm gold wire attached via carbon paint. The magnetic field was applied parallel to the **b** axis. Measurements were performed in a ^{3}He cryostat using a 33-T Bitter magnet at the National High Magnetic Field Laboratory in Tallahassee. Figure 1 shows the field dependence of the interlayer resistance at several different temperatures. The magnetic field is parallel to the current

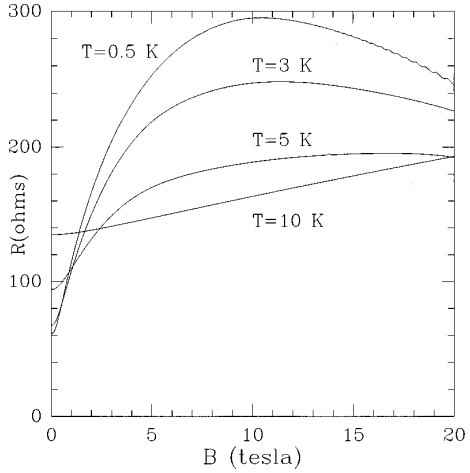

FIG. 1. Magnetic field dependence of the interlayer resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at several temperatures. The magnetic field and the current direction were perpendicular to the layers, i.e., parallel to the least-conducting direction.

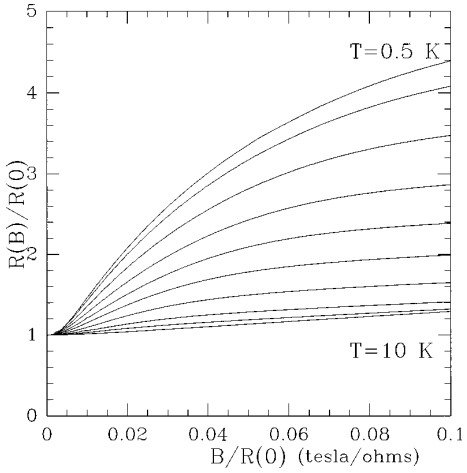

FIG. 2. Kohler plot of the magnetoresistance. The temperatures of the curves shown are (from top to bottom) 0.5, 1.5, 3.0, 3.5, 4.2, 5.0, 6.0, 7.0, 8.0, and 10.0 K. If Kohler's rule held then all the curves would lie on top of on one another.

and perpendicular to the layers. The data are consistent with previously published data on this class of materials.[16,19,21–23]

Given that the current direction and magnetic field are parallel one expects no Lorentz force on the electrons. This raises the question of the origin of such a large longitudinal magnetoresistance. Semiclassical theories explain this by assuming that the interlayer hopping also involves a substantial simultaneous hopping parallel to the layers.[18] Hill has shown how such hopping, and the associated warping of the Fermi surface in the interlayer direction, can be used to explain cyclotron resonance experiments.[24] The microscopic justification for assuming this type of interlayer hopping is not clear.

The strong angular dependence of the interlayer magnetoresistance[10,17,22,25] implies that it is predominantly orbital in origin. When the field is parallel to the layers or at certain magic angles the magnetoresistance is several times smaller than when the field is perpendicular to the layers. If the magnetoresistance was predominantly due to the field coupling to the spins it should be almost isotropic.

Figure 2 shows a Kohler plot of the data in Fig. 1 as well as data at additional temperatures. It covers fields up to about 10 T. If Kohler's rule held all of the curves would collapse onto a single curve. They do not because the magnetoresistance varies strongly with temperature but the zero-field resistance is only weakly temperature dependent (Fig. 1). Note that there is *no* field range over which Kohler's rule holds. This rules out explaining the deviation in terms of quantum effects or magnetic breakdown.

We now consider five possible explanations for the violation of Kohler's rule, within the framework of semiclassical transport theory. (i) The electronic structure varies with temperature due to the formation of the density wave. This can explain the temperature dependence between 4 and 10 K. However, in density-wave systems the electronic energy gap varies very little at temperatures less than half the transition temperature.[26] In this system, below 4 K, there is little change in the zero-field resistance (see Fig. 1), Hall resistance,[23] Knight shift, and nuclear magnetic relaxation rate, $1/(T_1T)$.[27] This suggests that the electronic structure and density of states does not vary significantly below 4 K and so cannot explain the large temperature dependence of the magnetoresistance.[28]

(ii) There is more than one type of carrier and their mobilities have different temperature dependences. The existence of more than one type of carrier in the low-temperature phase is suggested by the observation of more than one magneto-oscillation frequency[29] and more than one cyclotron resonance frequency.[30] To illustrate how this can lead to violations of Kohler's rule we consider the case of two carriers.[31] Let n_1 and n_2 denote the densities and μ_1 and μ_2 denote the mobilities of the carriers. The zero-field resistance is $\rho_0=(n_1\mu_1+n_2\mu_2)^{-1}$. At low fields the transverse[32] magnetoresistance is[3]

$$\frac{\Delta\rho_{xx}}{\rho_0}=\frac{n_1n_2\mu_1\mu_2(\mu_1-\mu_2)^2B^2}{(n_1\mu_1+n_2\mu_2)^2}. \tag{2}$$

Hence, if μ_1 and μ_2 have a different temperature dependence so will the resistance and magnetoresistance. To see this clearly consider the particular case where $n_1\sim n_2$ and $\mu_1\gg\mu_2$ then $\rho_0\simeq(n_1\mu_1)^{-1}$ and $\Delta\rho_{xx}/\rho_0\simeq(n_2\mu_1\mu_2/n_1)B^2$. Hence, if μ_2 has a much stronger temperature dependence than μ_1 then the desired behavior is obtained.[33] However, in this limit the Hall resistance is $R_H\simeq\mu_2/(n_1\mu_1)$ and so should be strongly temperature dependent. However, this is inconsistent with observations (albeit on a different sample).[23,34]

(iii) The temperature dependence of the scattering rate varies significantly at different points on the Fermi surface.[35] Similar ideas about ''hot spots'' have been proposed to explain the magnetotransport in the cuprates[36] and quasi-one-dimensional organic metals.[37,38] A different temperature dependence for the resistance and magnetoresistance arises because the former is related to the inverse of the average of the scattering time over the Fermi surface and the latter (at high fields) is related to the average of the scattering rate over the Fermi surface.[38] Alternatively, the magnetoresistance can be shown to be the variance of the Hall angle over

the Fermi surface.[4] The nonuniform scattering rate also leads to a temperature dependence of the Hall resistance R_H since it is given by[39]

$$R_H = \frac{1}{ne}\frac{\langle \tau^2 \rangle}{\langle \tau \rangle^2}, \tag{3}$$

where $\langle \cdots \rangle$ denotes an average over the Fermi surface. However, again this explanation requires the Hall resistance to vary significantly below 4 K. However, this is not observed.

(iv) The scattering times associated with the magnetoresistance and the zero-field resistance are distinct and have different temperature dependences. This hypothesis[6] has been proposed to explain the unusual temperature dependence of the magnetotransport (including the violation of Kohler's rule) in the metallic phase of the cuprate superconductors.[4,5] Distinct scattering times are associated with the decay of electric and Hall currents and denoted by τ_0 and τ_H, respectively. The zero-field conductivity $\sigma_{xx}(0) \sim \tau_0$, the magnetoconductivity $\sigma_{xx}(B) - \sigma_{xx}(0) \sim B^2 \tau_0 \tau_H^2$,[32] and the Hall conductivity $\sigma_{xy} \sim B \tau_0 \tau_H$. Consequently, this explanation also requires the Hall resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ to vary significantly below 4 K. Measurements suggest that it does not.[23,34]

(v) The scattering time τ is field dependent in a way that $\tau(B,T)/\tau(0,T)$ is temperature dependent. Several calculations have considered the electron-electron scattering rate in the quasi-one-dimensional Bechgaard salts (TMTSF)$_2X$ and suggested that it is field dependent.[40] Alternatively, if the scattering is due to local magnetic moments, possibly associated with a spin-density wave, then that will vary with field.[2] Although these explanations for the violation of Kohler's rule are possible it should be stressed that if they are correct then the origin of the magnetoresistance in these materials is quite different from what has been proposed.[17,18]

The possible failure of semiclassical transport theory to describe the interlayer magnetoresistance raises the question: Is the interlayer transport incoherent, i.e., does the concept of Bloch states (on which the Boltzmann equation depends) have meaning?

For this class of materials Yoshioka[41] has proposed an explanation for the magnetoresistance and its angular dependence that does not involve coherent interlayer transport. Yoshioka's model assumes that there is a periodic potential due to a density wave in each layer. A magnetic field then produces a periodic potential whose period along the **b** axis, i.e, perpendicular to the layers, is *incommensurate* with the interlayer spacing. If the magnitude of this potential is larger than the interlayer hopping rate then all the states along the **b** axis will be localized.[42] The strength of the incommensurate potential increases with field and makes the states more localized. Hence, the interlayer resistance increases with increasing field. The incommensurability of the potential varies as the field is tilted. At certain angles the potential will become commensurate, the states will no longer be localized and the magnetoresistance will vanish. The model correctly predicts these angles.[41] Although all the states are localized the conductivity should be nonzero at finite temperature due to variable range hopping. As the temperature is lowered variable range hopping becomes harder and the resistance increases.[43] Hence, in this model the temperature dependence of the magnetoresistance is unrelated to that of the zero-field resistance and Kohler's rule would not be expected to hold. However, this model would predict that, contrary to what is observed, the magnetoresistance does not saturate as the temperature is lowered.[44]

The issue of incoherent interlayer transport has recently been considered for the cuprate superconductors[45,46] and for the layered organic crystal (TMTSF)$_2$PF$_6$, which under pressure is a quasi-one-dimensional metal. Its magnetoresistance strongly violates Kohler's rule and only depends on the component of magnetic field perpendicular to the layers.[47] Although there are some similarities there are also differences to the material studied here. For example, in (TMTSF)$_2$PF$_6$, the angular dependence of the magnetoresistance has a minimum when the field is perpendicular to the layers whereas for α-(BEDT-TTF)$_2$KHg(SCN)$_4$ it is a maximum.[10] Although it would be interesting to apply the ideas in Ref. 46 to the data presented here it is not clear how to do so.

In conclusion, the temperature dependence of the interlayer magnetoresistance of the quasi-two-dimensional metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$ cannot be explained in terms of existing theoretical models including, (i) semiclassical transport on a single Fermi surface with a single scattering time[18] and (ii) Yoshioka's model[41] involving incoherent interlayer transport. We suggest several directions for future work. Experimentally, Kohler's rule should be tested outside the low-temperature phase and in other metals based on the BEDT-TTF molecule. Hall resistance and magnetoresistance measurements should be done on the same sample to completely rule out the ''hot spot'' and two scattering time hypotheses for these systems. Theoretically, we need calculations of the magnetoresistance for models[45,46] involving incoherent interlayer transport.

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