Magnetic field induced coherence-incoherence crossover in the interlayer conductivity of a layered organic metal

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The angle-dependent interlayer magnetoresistance of the pressurized (to the normal metallic state) layered organic metal α -(BEDT-TTF)₂KHg(SCN)₄ is found to change from the conventional behavior at low magnetic fields to an anomalous one at high fields. The dependence of this field-induced crossover on the sample purity and temperature reveals parallel contribution of the classical Boltzmann and incoherent channels in the interlayer conductivity. The latter channel, having a metallic temperature dependence but being insensitive to an in-plane magnetic field, may be responsible for magnetoresistance anomalies observed in a number of layered metals. We propose a possible mechanism for the incoherent channel combining interlayer tunneling via local hopping centers and intralayer diffusion.

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I. INTRODUCTION

Dimensional crossovers and their influence on transport properties and electronic states is a long standing and still controversial issue in the field of highly anisotropic correlated conductors, such as superconducting cuprates, cobaltates, organics, intercalated compounds, etc. One of the most frequently discussed mechanisms of breaking the interlayer band transport in a layered metal is due to scattering. If the scattering rate τ^{-1} is larger than the interlayer hopping rate, $\tau_h^{-1} \sim t_\perp / \hbar$, the quasiparticle momentum and Fermi surface are only defined within conducting layers, i.e., become strictly two dimensional (2D). Nevertheless, as long as the charge transfer between two adjacent layers is determined by direct one electron tunneling ("weakly incoherent" regime¹), the interlayer resistivity $\rho_{\perp}(T)$ is predicted to be identical to that in the fully coherent three-dimensional (3D) case^{2,3} and thus proportional to the resistivity along the layers $\rho_{\parallel}(T)$. At increasing temperature, the conductivity due to direct tunneling decreases and other conduction mechanisms associated, e.g., with small polarons^{4,5} or resonant impurity tunneling^{3,6} may come into play. This may lead to a crossover from a low-temperature metallic to a high-temperature, seemingly, nonmetallic temperature dependence of ρ_{\perp} which was reported for various layered materials.^{7–11} However, it does not explain the fact that the resistivity anisotropy in many of these compounds grows continuously upon cooling deep into the metalliclike regime of $\rho_{\perp}(T)$.^{7–9,12}

In addition to the latter apparent inconsistency, recent magnetotransport experiments have revealed a low-temperature behavior strongly violating theoretical predictions. The interlayer resistance R_{\perp} of a, presumably, weakly incoherent sample of the organic metal α -(BEDT-TTF)₂KHg(SCN)₄ has been found to be insensitive to a strong magnetic field applied parallel to layers.¹³ This is, in particular, reflected in a broad dip in the angular dependence of magnetoresistance which is centered at θ =90° and scales with B_{\perp} =B cos θ , where θ is the angle between the field and the normal to layers. While a similar dip

in the angle-dependent magnetoresistance (AMR) has been observed on a number of other layered materials with different in-plane Fermi-surface topologies,^{14–17} its origin remains unexplained.

For the organic conductor (TMTSF)₂PF₆ characterized by a flat weakly warped Fermi surface the anomalous dip structure was reported for a field rotation in the plane of the Fermi sheets.^{14,18} It was, however, noticed¹⁸ that the dip develops only at a high enough magnetic field B > 1 T; at low fields the curves $R_{\perp}(\theta)$ display a conventional shape with a maximum at the field parallel and a minimum at the field perpendicular to layers. The dramatic change in the AMR behavior was interpreted as a result of a field-induced confinement of conducting electrons. Semiclassically, the excursion of a charge carrier across the layers is restricted by a strong inplane magnetic field B_{\parallel} and limited to within one layer when $B_{\parallel} \ge B_c = 4t_{\perp} / edv_F$, where e is the elementary charge, d is the interlayer period, and v_F is the Fermi velocity. This was suggested to lead to a dimensional crossover and a consequent breakdown of the Fermi-liquid behavior.¹⁹ While the fieldinduced confinement scenario¹⁹ describes qualitatively a number of features of the magnetoresistance in (TMTSF)₂PF₆, it still does not provide a consistent explanation for the dip around $\theta = 90^{\circ}$. It remains also unclear why the crossover field increased with temperature in the experiment.¹⁸ Further, as it will be shown below, the crossover between the low-field, conventional, and high-field, anomalous AMR can also be observed on a system possessing a cylindrical Fermi surface. It is unclear, to what extent the field-induced confinement can be effective in this case.

In the present paper we report on the crossover in the shape of the angle-dependent interlayer magnetoresistance of α -(BEDT-TTF)₂KHg(SCN)₄. All the measurements were done under a pressure of ≈ 6 kbar in order to suppress the density-wave formation and stabilize the normal metallic state²⁰ with a well-defined Fermi surface consisting of a pair of open sheets and a cylinder.²¹ We show that the field-induced confinement model¹⁹ is inconsistent with the evolution of the crossover with temperature and sample purity. On

the other hand, the observed behavior is strongly suggestive of two parallel contributions to the interlayer conductivity: a classical Boltzmann channel, σ_c , and an anomalous incoherent channel, σ_i . We propose a possible explanation of the field and temperature dependence of σ_i , without invoking non-Fermi-liquid effects.

II. EXPERIMENT

The samples used in this study were single crystals of α -(BEDT-TTF)KHg(SCN)₄ in the form of platelets with typical dimensions of $\sim 0.5 \times 0.7 \text{ mm}^2$ in the plane of highly conducting layers (crystallographic *ac* plane) and a thickness of 0.07-0.2 mm. To measure the interlayer resistance, two pairs of annealed platinum wires of 20 μ m diameter were glued to both *ac* faces of a sample by a conducting graphite paste, yielding contact resistances of $\simeq 10 \ \Omega$. The contacted samples were placed into a 10 mm diameter piston-cylinder pressure cell made of beryllium copper. A pressure of up to 8 kbar was applied using GKZh silicon oil as pressure medium and locked at room temperature. A pressure drop of ~1.5 kbar during cooling down to liquid-helium temperatures was measured using a calibrated manganin resistive gauge placed in the sample space close to the samples. Two samples could be mounted in the cell for simultaneous measurements at identical conditions. The loaded cell was fixed on a two-axis rotation stage allowing an *in situ* variation in the sample orientation with respect to the applied magnetic field. The orientation was defined as shown in the inset in Fig. 1: by the polar angle θ between the field direction and the normal to the *ac* plane and by the azimuthal angle φ between the field projection on the *ac* plane and the *a* axis. The absolute values of both angles could be determined to an accuracy better than 0.5°, the angular resolution was better than 0.1° . The measurements were performed in a ⁴He flow cryostat with a base temperature of 1.4 K. Magnetic fields up to 15 T were generated by a superconducting coil. The experimental results presented below for two samples, studied in the same measurement run, have been reproduced on several other samples measured separately under similar conditions.

III. RESULTS AND DISCUSSIONS

A. Crossover in the AMR shape

Figure 1 shows AMR patterns from two samples of α -(BEDT-TTF)₂KHg(SCN)₄ under a pressure of 6 kbar, recorded at T=1.4 K, at different field intensities, B=0.12, 0.5, 3, and 15 T. For both samples, the field is rotated in the plane perpendicular to the highly conducting *ac* plane and forming an angle $\varphi \approx 20^{\circ}$ with the *a* axis, the latter being perpendicular to the open Fermi sheets. The oscillatory behavior, particularly pronounced at high fields, is due to the semiclassical angular magnetoresistance oscillations and Shubnikov–de Haas effects, as described in detail elsewhere.²² It reveals a high crystal quality of both samples, providing an estimate for the transport scattering time, $\tau_1 \approx \tau_2/3 \sim 5$ ps for samples 1 and 2, respectively. In the rest



FIG. 1. (Color online) (a) Angle-dependent interlayer magnetoresistance of a relatively dirty sample, 1, of α -(BEDT-TTF)₂KHg(SCN)₄ in the high-pressure metallic state recorded at T=1.4 K at magnetic fields (bottom to top): 0.12, 0.5, 3, and 15 T; $\varphi \approx 20^{\circ}$. (b) Same for a very clean sample, 2. The upper inset illustrates the definition of angles θ and φ ; the lower inset: enlarged fragment of the 3 T curve showing a small "coherence peak."

of the paper we focus on the nonoscillating magnetoresistance component.

The low-field curves shown in Fig. 1 have a conventional form for both samples: the AMR is maximum (minimum) at the field directed nearly parallel (perpendicular) to layers. At fields above ~1 T, a broad dip around $\theta = \pm 90^{\circ}$ develops in the AMR of sample 1. Already at B=3 T the magnetoresistance displays an absolute minimum at the field aligned parallel to layers (and perpendicular to the current). This crossover in the AMR shape is very similar to that reported for (TMTSF)₂PF₆.¹⁸ Note, however, that in the present case the Fermi surface contains, besides open sheets, a cylindrical part and $\varphi \approx 20^{\circ}$ corresponds to the field rotating close to the plane perpendicular to the open sheets. Such a geometry is clearly unfavorable for the field-induced confinement scenario.¹⁹

A comparison between the AMR of two samples shown in Fig. 1 reveals yet another disagreement with the field-induced confinement model. While the confinement field B_c is formally independent of the scattering time, the model implies a sufficiently high τ so that the strong-field criterion, $\omega_c \tau > 1$ (where ω_c is the characteristic frequency of orbital



FIG. 2. (Color online). Kohler plot of the normalized interlayer conductivity of sample 1 for the field aligned parallel to conducting layers obtained from field sweeps at different temperatures. Inset: temperature dependence of the zero-field resistance R_0 (thick line) and the resistances of the coherent, $R_c \propto 1/\sigma_c$, (circles) and incoherent, $R_i \propto 1/\sigma_i$, (triangles) channels, see text.

motion in a magnetic field), is fulfilled. Therefore, the effect should be seen, first of all, in clean samples. By contrast, in our case the crossover is observed in the relatively dirty sample 1, whereas the cleaner sample 2 preserves the normal anisotropy up to the highest field applied.

B. Two channels in the interlayer conductivity

For understanding the observed behavior, it is instructive to study the magnetoresistance of sample 1 as a function of magnetic field, aligned parallel to layers, and its evolution with temperature. The relevant data are shown in Fig. 2 in the form of a Kohler plot. Here, $R_0(T)$ is the zero-field resistance (see the inset) and the normalized field-dependent interlayer conductivity $\sigma(B,T)/\sigma(0,T)$ is obtained from R(B)measurements, taking into account that $\sigma(B) \propto 1/R(B)$ in our quasi-two-dimensional material. According to Kohler's rule, the magnetoresistance or, in our representation, magnetoconductivity at different fields and temperatures should be just a function of $B/R_0(T)$. This rule is strongly violated in Fig. 2: the curves corresponding to different temperatures rapidly diverge from each other. On the other hand, all the curves (at least, those from 1.4 to 10 K) tend to saturation²³ at B/R_0 ≥ 0.5 .

The described behavior suggests two parallel contributions in the conductivity:

$$\sigma(\boldsymbol{B},\tau) = \sigma_c(\boldsymbol{B},\tau) + \sigma_i(\boldsymbol{B}_{\perp},\tau). \tag{1}$$

Here, the first term on the right-hand side is the coherent Boltzmann conductivity depending on both the strength and orientation of a magnetic field.^{24,25} In a field parallel to layers it decreases proportional to $(\omega_c \tau)^{\alpha}$ with $1 \le \alpha \le 2$ and, at a high enough field, the second term in Eq. (1) becomes dominant. We associate the latter with incoherent interlayer



FIG. 3. (Color online). Kohler plot of the coherent part of interlayer conductivity of sample 1 in the field parallel to the layers at temperatures 1.4 to 10 K. The coherent conductivity has been determined from the data in Fig. 2: $\frac{\sigma_c(B,T)}{\sigma_c(0,T)} = \frac{\sigma(B,T)/\sigma(0,T) - \sigma_i(T)/\sigma(0,T)}{1 - \sigma_i(T)/\sigma(0,T)}$. The resistance $R_c(T)$ corresponding to the coherent channel at zero magnetic field is taken from the inset in Fig. 2.

charge transfer. In agreement with previous observations,¹³ the incoherent conductivity is insensitive to the in-plane magnetic field; however, it does depend on the field component B_{\perp} perpendicular to layers. This is why the resistance increases, as the field is tilted from the direction parallel to layers [3 and 15 T curves in Fig. 1(a)]. At high fields, the total conductivity is dominated by σ_i in a large angular interval around $\theta = \pm 90^\circ$, which leads to a scaling behavior of magnetoresistance: $R(B, \theta) = R(B \cos \theta)$.¹³

Evaluating the relative contribution of $\sigma_i(T)$ to the total zero-field conductivity $\sigma(0,T)$ of sample 1 from the level, at which the curves in Fig. 2 come to saturation, and using the $R_0(T)$ data plotted in the inset of Fig. 2, one can extract separately the temperature dependences of the classical Boltzmann (circles in the inset) and incoherent (triangles) channels. Note that even the anomalous, incoherent channel shows a metallic behavior.

Taking into account that the incoherent channel is insensitive to an in-plane field, it is natural to replot the data in Fig. 2 after excluding its contribution to the total conductivity. The resulting Kohler plot for the coherent channel, $\sigma_c(B,T)/\sigma_c(0,T) = [\sigma(B,T) - \sigma_i(T)]/[\sigma(0,T) - \sigma_i(T)]$, is shown in Fig. 3. Note that the total zero-field resistance $R_0(T)$ is replaced by the resistance $R_c(T)$ corresponding to the coherent channel (circles in the inset of Fig. 2) in the argument of the Kohler plot. By contrast to the total conductivity, the coherent channel demonstrates a very good scaling according to Kohler's rule: the curves in Fig. 3 are almost indistinguishable from each other, thus providing a substantial support to our two-channel model.

The presence of two channels in the interlayer conductivity provides a natural explanation for the anomalous dip in the AMR found, at certain conditions, on very clean samples of α -(BEDT-TTF)₂KHg(SCN)₄.¹³ In such samples, the conductivity is dominated by σ_c as long as the in-plane field component parallel to the open Fermi sheets is small. This is, in particular, reflected in the shape of the small- φ AMR of sample 2 shown in Fig. 1(b): the angular dependence on the vicinity of $\theta = \pm 90^{\circ}$ is rather flat and shows a narrow peak, revealing a coherent 3D Fermi surface.²⁴ When the in-plane field component is turned from the a axis to the c axis, which is parallel to the open Fermi sheets, the coherent conductivity rapidly drops down.²⁴ Under these conditions, the incoherent channel σ_i may become important, which leads to the anomalous dip structure in the b^*c -rotation patterns of the AMR even for relatively clean samples.¹³ In the same way can be interpreted the 90° dips observed in the AMR of clean samples of other highly anisotropic compounds, like $(TMTSF)_2X$ with $X = PF_6$ (Ref. 26) and ReO₄ (Ref. 15) or β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃.¹⁶

The proposed model also explains the temperature dependence of the crossover field B_c observed in the experiment on $(\text{TMTSF})_2\text{PF}_6$.¹⁸ Indeed, while the contribution of the coherent channel decreases with increasing temperature, as seen from Fig. 2, it still remains significant at $T \sim 10$ K. At the same time, the scattering rate, which is proportional to the resistance of the coherent channel, grows by about an order of magnitude between 1.4 and 10 K (see inset in Fig. 2). Therefore, a much higher field is necessary at 10 K for "freezing out" σ_c and making the incoherent channel dominant in the field and angular dependence of magnetoresistance.

C. Possible mechanism of the incoherent channel

The metallic behavior of the incoherent conduction channel, see inset in Fig. 2, apparently comes into conflict with the existing theories of incoherent interlayer charge transfer,^{3–6} predicting an insulating temperature dependence. In addition, those theories do not account for the significant dependence of σ_i on magnetic field normal to layers. To comply with the experimental observations, we propose to consider elementary events of incoherent interlayer hopping via local centers, such as resonance impurities,^{3,6} in combination with diffusive intralayer transfer from one hopping center to another. The essential requirement of our model is that the volume concentration of hopping centers n_i be small so that the average distance l_i between them along the 2D layers is much larger than the in-plane mean free path l_{τ} $=v_F \tau$: $l_i = (n_i d)^{-1/2} \gg l_{\tau}$. This condition, being opposite to the model,³ looks reasonable since the concentration of resonant impurities (i.e., those impurities which form an electron level with energy close to the Fermi energy) is definitely much lower than the concentration of all kinds of impurities. The current through each hopping center is limited by the resistance R_{\perp} , which contains two in-series elements:

$$R_{\perp} = R_{\rm hc} + R_{\parallel}.\tag{2}$$

The first part, R_{hc} , is the hopping-center resistance itself, which is almost independent of magnetic field and can have a weak nonmetallic temperature dependence $R_{hc}(T)$. The second part, R_{\parallel} , is the intralayer resistance, which comes up because the electrons must travel along the conducting layer over a distance $\sim l_i$. In the limit $l_i \geq l_{\tau}$, the 2D intralayer current density j(r) at each point is proportional to the electric field E(r) at this point: $j = \sigma_{\parallel} dE$, where an isotropic inplane conductivity is assumed for simplicity.

A stationary in-plane current obeys: $\operatorname{div} j(r)=0$ everywhere except the hopping-center spots. In the vicinity of each hopping center, the current and electric field are roughly axially symmetric and given by

$$E(r-r_{i}) = \frac{j(r-r_{i})}{\sigma_{\parallel}d} = \frac{I_{0}}{2\pi\sigma_{\parallel}d} \frac{(r-r_{i})}{|r-r_{i}|^{2}},$$
(3)

where I_0 is the current through the hopping center located at point r_i . R_{\parallel} is determined by the in-plane mean voltage drop between two successive hopping centers:

$$I_0 R_{\parallel}(T) \simeq 2 \int_{l_{\tau}}^{l_i} E(r) dr = \frac{I_0 \ln(l_i/l_{\tau})}{\pi \sigma_{\parallel} d}.$$
 (4)

As the lower cutoff in the integral [Eq. (4)] we take the mean free path $l_{\tau\tau}$ which neglects the resistance of the ballistic region $|\mathbf{r}-\mathbf{r}_i| < l_{\tau\tau}$. Since the ballistic conductivity is much higher than the diffusive one, this approximation should work well, at least when $\ln(l_i/l_{\tau\tau}) \ge \ln(l_{\tau\tau}/d)$.

The mean voltage drop between two adjacent conducting layers is $E_0 d = I_0 (R_{hc} + R_{\parallel})$, where E_0 is the external electric field perpendicular to the layers. The total current density in the interlayer direction is $j_t = I_0 n_i d = \sigma_i E_0$, yielding the interlayer conductivity:

$$\sigma_i = \frac{\pi \sigma_{\parallel} n_i d^3}{\pi d \sigma_{\parallel} R_{\rm hc} + \ln(l_i / l_{\tau})}.$$
(5)

The present simple model can be generalized by including the distribution of the hopping centers $n[R_{hc}(T)]$ and performing integration over $R_{hc}(T)$. The exact result will depend on the particular physical model of the hopping centers. In the trivial case of short circuiting the layers (e.g., by dislocations) $R_{\rm hc} \approx 1/d\sigma_{\parallel}$ and σ_i should be just proportional to the intralayer conductivity. The fact that the temperature dependence of the incoherent channel is metalliclike, however, considerably weaker than that of the coherent one (inset in Fig. 2), implies that $R_{\rm hc}$ is larger than $1/d\sigma_{\parallel}$ and only slightly varies with (or is independent of) temperature. Such conditions can be fulfilled if the hopping occurs via resonance impurities.³ Further, assuming hopping centers are nonmagnetic, $R_{\rm hc}$ is insensitive to magnetic field and the dependence of $\sigma_i(\mathbf{B})$ is determined by $\sigma_{\parallel}(\mathbf{B})$. The latter is subject to the usual galvanomagnetic effect in a field normal to conducting layers, which leads to a decrease in σ_i at increasing B_{\perp} . However, an in-plane magnetic field leaves σ_{\parallel} and, hence, σ_i unchanged due to the absence of coherent orbital motion across the layers. Therefore, the magnetoresistance associated with the incoherent channel is expected to be a function of the out-of-plane field component only: $R(B) = R(B \cos \theta)$. Thus, the proposed mechanism is consistent, at least, qualitatively, with the main features of the incoherent channel observed in the experiment.

IV. SUMMARY

In conclusion, we have shown that the anomalous behavior of the angle-dependent interlayer magnetoresistance in the highly anisotropic layered metal α -(BEDT-TTF)₂KHg(SCN)₄ can be described by parallel contribution of two conduction channels, σ_c and σ_i , providing, respectively, coherent and incoherent interlayer charge transfers. A sufficiently high in-plane component of magnetic field changes the proportion of σ_c and σ_i in favor of the latter, thus causing an apparent dimensional crossover. However, by contrast to the field-induced confinement scenario,¹⁹ this crossover does not imply a change in the dynamic properties of charge carriers. The proposed model is able to explain not only the observed crossover but also anomalous features found in other layered metals situated in the transient region between the fully coherent and incoherent transport regimes.

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