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 layered organic metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is found to undergo a dramatic change from the classical conventional behavior at low magnetic fields to an anomalous one at high fields. This field-induced crossover and its dependence on the sample purity and temperature imply the existence of two parallel channels in the interlayer transport: a classical Boltzmann conductivity $\sigma_c$ and an incoherent channel $\sigma_i$. We propose a simple model for $\sigma_i$, explaining its metallic temperature dependence and low sensitivity to the inplane field component.

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 $\tau^{-1}$ is larger than the interlayer hopping rate, $\tau_i^{-1} \sim t_i/h$, 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” regime 1), the interlayer resistivity $\rho_\perp(T)$ is predicted to be identical to that in the fully coherent three-dimensional (3D) case 2 3. At increasing temperature, the conductivity due to direct tunneling decreases and other conduction mechanisms associated, e.g., with small polaronz 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 $\rho_\perp$. Such a scenario is qualitatively consistent with a nonmonotonic dependence $\rho_\perp(T)$ with a maximum at $T_m \sim 100$ K reported for various layered materials 7 8 9 10 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 8 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)$_2$KHg(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 $\theta = 90^\circ$ and scales with $B_\perp = B \cos \theta$, where $\theta$ 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 inplane Fermi surface topologies 14 15 16 17, its origin remains unexplained.

For the organic conductor (TMTSF)$_2$PF$_6$ 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, noticed 18 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 of 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 $B_\parallel \geq B_c = 4t_\perp/edv_F$, 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 field-induced confinement scenario 19 describes qualitatively a number of features of the magnetoresistance in (TMTSF)$_2$PF$_6$, 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.

Here, we report on the crossover in the shape of the angle-dependent interlayer magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. All the measurements were done under a pressure of $\approx 6$ kbar in order to suppress the density-wave formation and stabilize the normal metallic state 20 with a well defined Fermi surface consisting of
a pair of open sheets and a cylinder \[21\]. We show that the field-induced confinement model \[19\] 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, \(\sigma_c\), and an anomalous, incoherent channel, \(\sigma_i\). We propose a possible explanation of the field and temperature dependence of \(\sigma_i\), without invoking non-Fermi liquid effects.

Figure 1 shows AMR patterns from two samples of \(\alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4\) recorded at \(T = 1.4\) K, at different field intensities, \(B = 0.12, 0.5, 3,\) and \(15\) T. The transport current is applied perpendicular to the ac plane, that is the plane of conducting layers. Azimuthal angle \(\varphi\) measured between the field projection on the ac plane and the \(a\) axis, the latter being perpendicular to the open Fermi sheets, is \(\approx 90^\circ\) for both samples. The oscillatory behavior, particularly pronounced at high fields is due to angular magnetoresistance oscillation 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, \(\tau\), the confinement field \(B_{\text{c}}\) is formally independent of the scattering time, the strong field criterion, \(\omega_c \tau > 1\) (\(\omega_c\) is the characteristic frequency of orbital 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.

To explain 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 is shown in Fig. 2 in the form of a Kohler plot. Here, \(R_0(T)\) is the zero-field resistance shown in the inset and the normalized field-dependent interlayer conductivity \(\sigma(B,T)/\sigma(0,T)\) has been obtained from \(R(B)\) measurements, taking into account that \(\sigma(B) \propto 1/R(B)\) in our quasi-2D 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, saturating at different levels \[23\]. On the other hand, the saturation occurs at approximately the same \(B/R_0\), independent of \(T\).

The described behavior suggests two parallel contributions in the conductivity:

\[
\sigma(B,\tau) = \sigma_c(B,\tau) + \sigma_i(B,\tau).
\]  

FIG. 1: (color online). Angle-dependent interlayer magnetoresistance of a relatively dirty sample, \# 1, of \(\alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4\) 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 \(\theta\) and \(\varphi\); the lower inset: enlarged fragment of the 3 T curve showing a small “coherence peak”.
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 \[ \tau_1 \leq \omega \tau_1 \] in a field parallel to layers it decreases proportional to \((\omega \tau_1)^n\) with \(1 \leq n \leq 2\) and, at a high enough field, the second term in Eq. (11) becomes dominant. We associate the latter with incoherent interlayer charge transfer. In agreement with previous observations [13], the incoherent conductivity is insensitive to the inplane 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 T and 15 T curves in Fig. 1(a)]. At high fields, the total conductivity is dominated by \(\sigma_i\) 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 \(\sigma_i\) 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 (empty circles in the inset) and incoherent (filled triangles) channels. Note that even the anomalous, incoherent channel shows a metallic behavior.

The two-channel model provides a natural explanation for the anomalous dip in the AMR found, at certain conditions, on very clean samples of \(\alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4\). In such samples, the conductivity is dominated by \(\sigma_i\), as long as the inplane field component parallel to the open Fermi sheets is small. This is, in particular, reflected in the shape of the small-\(\varphi\) AMR of sample \# 2 shown in Fig. 1(b): the angular dependence in the vicinity of \(\theta = \pm 90^\circ\) is rather flat and shows a narrow peak, revealing a coherent 3D Fermi surface [24]. When the inplane 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 \(\sigma_i\) 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 \((\text{TMTSF})_2\text{X}\) with \(\text{X}=\text{PF}_6\) [20] and ReO\(_4\) [13] or \(\beta''-(\text{BEDT-TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3\) [10].

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” \(\sigma_c\) and making the incoherent channel dominant in the field and angular dependence of magnetoresistance.

Turning to a possible origin of the incoherent conduction channel, its metallic behavior apparently comes into conflict with the existing theories of incoherent interlayer charge transfer (see [3, 4, 5, 6] and references therein) predicting an insulating temperature dependence. In addition, those theories do not account for the significant dependence of \(\sigma_i\) 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 inplane mean free path \(l_\tau = v_F \tau\): \(l_i = (n_i d)^{-1/2} \gg l_\tau\). 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:

\[ R_{\perp} = R_{\text{hc}} + R_0 \]

The first part, \(R_{\text{hc}}\), is the hopping-center resistance itself, which is almost independent of magnetic field and can have a weak nonmetallic temperature dependence.
\( R_{\text{hc}}(T) \). The second part, \( R_{\parallel} \), is the intralayer resistance, which comes out because the electrons must travel along the conducting layer over a distance \( \sim l_{\tau} \). In the limit \( l_{i} \gg l_{\tau}, \) the 2D intralayer current density \( j(r) \) at each point is proportional to the electric field \( E(r) \) at this point: \( j_{\alpha}(r) = \sigma_{\alpha\beta}E_{\beta}(r) \), where \( \sigma_{\alpha\beta} \) is just the macroscopic 3D inplane conductivity. For simplicity, we consider an isotropic inplane conductivity: \( \sigma_{\alpha\beta} = \sigma_{\parallel} \delta_{\alpha\beta} \).

Since the charge density does not change with time, the inplane current must satisfy div\( \mathbf{j}(r) = 0 \) 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

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

where \( I_{0} \) is the current through the hopping center located at point \( r_{i} \). \( R_{\parallel} \) is determined by the inplane mean voltage drop between two successive hopping centers:

\[
I_{0}R_{\parallel}(T) \approx 2 \int_{r_{\tau}}^{l_{i}} E(r) \, dr = \frac{I_{0} \ln(l_{i}/l_{\tau})}{\pi \sigma_{\parallel}d}. \tag{4}
\]

As the lower cutoff in the integral \((4)\) we take the mean free path \( l_{\tau} \), which neglects the resistance of the ballistic region \( |r - r_{i}| < l_{\tau} \) around each impurity. Since the ballistic conductivity is much higher than the diffusive one, this approximation should work well, at least when \( \ln(l_{i}/l_{\tau}) \gtrsim \ln(l_{\tau}/d) \).

The mean voltage drop between two adjacent conducting layers is \( E_{0d} = I_{0}(R_{\text{hc}} + R_{\parallel}) \), where \( E_{0} \) is the external electric field perpendicular to the layers. The total current density in the interlayer direction is \( j_{\parallel} = I_{0}n_{d} = \sigma_{\parallel}E_{0} \), yielding the interlayer conductivity:

\[
\sigma_{\parallel} = \frac{\pi \sigma_{\parallel}n_{d}d^{3}}{\pi d \sigma_{\parallel}R_{\text{hc}} + \ln(l_{i}/l_{\tau})}. \tag{5}
\]

The present simple model can be generalized by including the distribution of the hopping centers \( n[R_{\text{hc}}(T)] \) and performing integration over \( R_{\text{hc}}(T) \). 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), \( R_{\text{hc}} \approx 1/\sigma_{\parallel} \) and \( \sigma_{\parallel} \) should be just proportional to the intralayer conductivity. The fact that the temperature dependence of the incoherent channel is considerably weaker than that of the coherent one (inset in Fig. 2), implies that \( R_{\text{hc}} \) is larger than \( 1/\sigma_{\parallel} \) and only slightly varies with (or is independent of) temperature. Such conditions can be fulfilled if the hopping occurs via resonance impurities [3]. Further, both \( R_{\text{hc}} \) and \( 1/\sigma_{\parallel} \) are largely insensitive to the inplane magnetic field in the incoherent regime whereas the significant dependence on the out-of-plane field component obviously comes from the intralayer conductivity. Thus, the proposed mechanism is consistent with the main features of the incoherent channel observed in the experiment.

In conclusion, we have shown that the anomalous behavior of the angle-dependent interlayer magnetoresistance in the highly anisotropic layered metal \( \alpha-(\text{BEDT-TTF})_{2}\text{KHg(SCN)_{4}} \) can be described by parallel contribution of two conduction channels, \( \sigma_{c} \) and \( \sigma_{i} \), providing, respectively, coherent and incoherent interlayer charge transfer. A sufficiently high inplane component of magnetic field changes the proportion of \( \sigma_{c} \) and \( \sigma_{i} \) 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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