Angle-Dependent Magnetoresistance in the Weakly Incoherent Interlayer Transport Regime in a Layered Organic Conductor

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We present comparative studies of the orientation effect of a strong magnetic field on the interlayer resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ samples characterized by different crystal quality. We find striking differences in their behavior, which is attributed to the breakdown of the coherent charge transport across the layers in the lower quality sample. In the latter case, the nonoscillating magnetoresistance background is essentially a function of only the out-of-plane field component, in contradiction to the existing Fermi-liquid theories.

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The mechanism of the interlayer charge transfer is one of the central questions in understanding the nature of various ground states and electronic properties of many exotic layered conductors of current interest, such as, for example, organic [1–3] or metal oxide superconductors [3–5]. In particular, the problem of discriminating between coherent and incoherent interlayer transport has received much attention (see, e.g., [6–12]).

If the coupling is strong enough, so that the interlayer hopping time, $\tau_h \sim \hbar/t_\perp$, where $t_\perp$ is the interlayer transfer integral, is considerably shorter than the transport scattering time τ, the electron transport is fully coherent and can be adequately described within the anisotropic three-dimensional (3D) Fermi-liquid (FL) model. In the other, incoherent limit, $\hbar/t_\perp \gg \tau$, the successive interlayer hopping events are uncorrelated; thus the electron momentum and the Fermi surface (FS) can be defined only in the plane of the layers. In the *strongly* incoherent regime there is no interference between the electron wave functions on adjacent layers, and the interlayer hopping is entirely caused by scattering processes. Consequently, the temperature-dependent resistivity across the layers, $\rho_\perp(T)$, is nonmetallic. On the other hand, one can consider the case of a weak overlap of the wave functions on adjacent layers, so that the interlayer transport is mostly determined by one particle tunneling. This *weakly* incoherent transport was studied in a number of theoretical works [7,8,13] assuming that the intralayer momentum is conserved during a single tunneling but successive tunneling events are uncorrelated due to scattering within the layers. The transverse resistivity $\rho_\perp$ has been shown to be almost identical to that in the coherent case, sharing with the latter the metallic temperature dependence [13] and most of the high-field magnetotransport phenomena [7,8]. Thus the following question arises: Is there a substantial physical difference between the coherent and weakly incoherent interlayer transport regimes?

Moses and McKenzie [7] proposed to use the angle-dependent magnetoresistance (MR) to distinguish between the two cases: When the field is turned in a plane normal to the layers, a narrow peak is often observed at the orientations nearly parallel to the layers [14]. This so-called *coherence peak* is associated with a topological change of electron cyclotron orbits on a 3D FS slightly warped in the direction perpendicular to the layers [14–16] and can only exist in the coherent regime. Its absence in the weakly incoherent transport model [7] is a natural consequence of the assumed strictly 2D FS.

The observation of the coherence peak has been used as an argument for the coherent interlayer coupling in a number of layered conductors [9–12]. However, no systematic experimental study of the weakly incoherent regime has been done thus far. Here we present comparative studies of the orientation effect of a high magnetic field on the interlayer MR of different samples of the layered organic conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$ [where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene]. We argue that, depending on the crystal quality, either the coherent or the weakly incoherent transport regime can be realized in this material. In agreement with the theoretical predictions, the coherence peak is observed only in the highest quality samples. However, by contrast to the coherent case, an important new feature, which cannot be explained by existing theories, has been found in the weakly incoherent regime: the MR in a field strongly tilted towards the layers turns out to be insensitive to the in-plane field component.

α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is one of the most anisotropic organic conductors [14]. Its electronic system comprises a quasi-1D and a quasi-2D conduction band. This compound exhibits a complex ‘‘magnetic field-pressure-temperature’’ phase diagram which can be consistently explained by a charge-density-wave (CDW) formation at a very low temperature, $T_{\mathrm{CDW}} \approx 8$ K [17–19]. In the CDW state the quasi-1D carriers are gapped, whereas the quasi-2D band remains metallic. Since we are presently focusing on the metallic magnetotransport, numerous anomalies associated with field-induced CDW transitions should be avoided. We will, therefore, consider the zero-pressure CDW state only at relatively low fields, up to 10 T, at which no significant change of the electronic system occurs. In addition, we present data taken at a high pressure, $P = 6.2$ kbar, which suppresses the CDW, restoring the fully normal metallic state. The measurements have been done at $T = 1.4$ K.

Figure 1 illustrates the dependence of the interlayer resistance of two different samples on the magnetic field orientation, measured at zero pressure. The latter is defined by the polar angle θ between the field direction and the normal to the plane of the layers, and by the azimuthal angle φ between the projection of the field on the plane and the crystallographic a axis. Both samples exhibit prominent angular MR oscillations (AMRO) periodic in tanθ: the

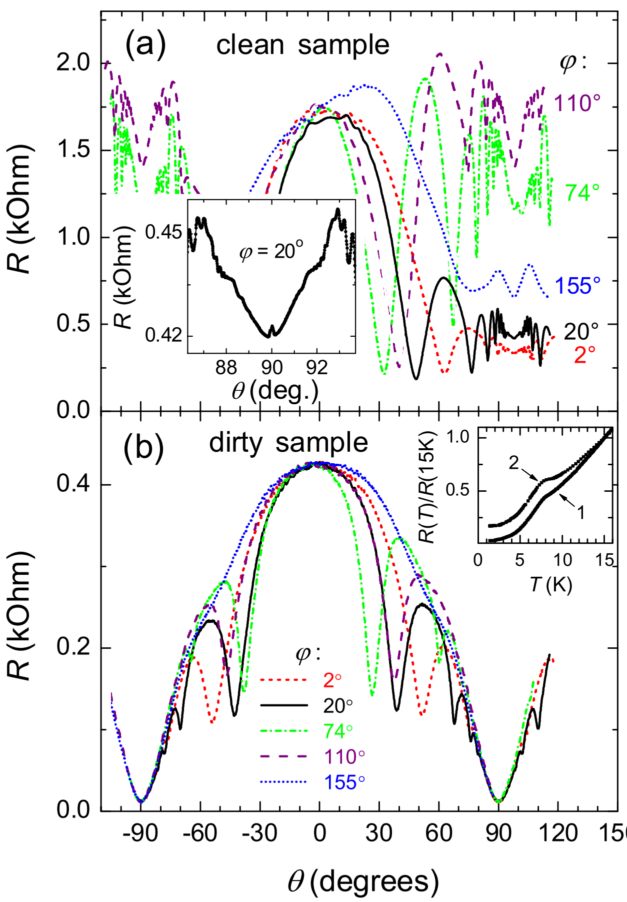

FIG. 1 (color online). Interlayer resistance of two crystals of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ as a function of the polar angle θ recorded at different azimuthal angles φ, $P = 0$ kbar, $B = 10$ T. Insets: (upper panel) details of a θ sweep for the clean sample, showing the small coherence peak; (lower panel) temperature dependence of the zero-field interlayer resistance of the clean (curve 1) and dirty (curve 2) samples; the shoulder at $\approx$8 K is due to the CDW transition.

resistance sharply drops at the Lebed magic angles [20]. This behavior is well known for the present material and is associated with the open-orbit motion of the metallic quasi-2D carriers in the presence of a CDW potential [21]. What we want to focus on now is the nonoscillating background which turns out to be drastically different in the two samples shown in Fig. 1.

In the highest quality sample [see Fig. 1(a)] the nonoscillating MR component displays a rather complex behavior strongly depending on the azimuthal angle φ. In particular, the φ dependence of the resistance at the field aligned exactly parallel to the layers, i.e., at $\theta = 90°$, is directly related to the in-plane curvature of the FS [14,22]. Further, a detailed inspection of the θ dependence around $\theta = 90°$ reveals a very narrow peak as shown in the inset of Fig. 1(a). It is, to our knowledge, the first observation of the coherence peak in the present compound.

The peak has been found at the azimuthal orientations, $0° \leq \varphi \leq 50°$, its width $\Delta\theta$ varying between 0.12° and 0.35°. One can, therefore, evaluate the FS corrugation in the interlayer direction [14]: $\Delta k_{\|}/k_F \approx \Delta\theta/k_F d \simeq 1.5 \times 10^{-3}$, where we have taken the mean value $\Delta\theta = 0.23°$, the intralayer Fermi wave number $k_F \simeq 0.14$ Å^{-1}, and the interlayer spacing $d \approx 20$ Å [2]. Further, estimating roughly the Fermi energy from the de Haas–van Alphen data [2], $\varepsilon_F \sim 40$ meV, we arrive at an extremely low value of the interlayer transfer integral: $t_{\perp} \approx (\Delta k_{\|}/2k_F)\varepsilon_F \sim 0.03$ meV. The corresponding hopping time, $\hbar/t_{\perp} \sim 20$ ps, is comparable to the scattering time estimated from damping of the AMRO at increasing θ [14], $\tau \simeq 15$ ps. Given the approximate character of the above estimations, we cannot judge about the exact ratio between the two quantities. It appears, however, that the interlayer coherence condition, $\hbar/t_{\perp} \ll \tau$, is not fulfilled. Nevertheless, the observed coherence peak indicates that a significant part of the carriers is in the coherent regime and can be ascribed to a 3D FS.

Another kind of the angular dependence is observed on the second sample as illustrated in Fig. 1(b). The amplitude of the AMRO is considerably weaker here and the oscillations are damped, with tilting the field towards $\pm 90°$, much faster than in the previous case, yielding the factor of 3 shorter scattering time, $\tau \simeq 5$ ps. We, therefore, will refer to this sample as the "dirty" one, by contrast to the "clean" sample considered above. Note, however, that both samples are clean enough in the sense that the strong field criterion, $\omega_c\tau \gg 1$, is always fulfilled in fields of a few Tesla.

No coherence peak has been found for the dirty sample at any φ, suggesting a breakdown of the interlayer coherence [7,8]. On the other hand, the presence of AMRO and the metallic temperature dependence $R(T)$ [see inset of Fig. 1(b)] indicate the weakly rather than strongly incoherent regime to be realized in the present case.

The most obvious distinction of the dirty sample is the behavior of the nonoscillating MR background: the latter decreases steadily as the field is tilted towards the layers, producing a broad dip around $\theta = \pm 90°$. Remarkably, as seen from Fig. 1(b), this behavior is practically independent of the azimuthal orientation of the field rotation plane.

To verify that the drastic difference in the behavior of the clean and dirty samples is related to the metallic magnetotransport and not to some specific features of the CDW state, we have performed measurements under high pressure at which the whole material is entirely normal metallic. Examples of the θ sweeps recorded for clean and dirty samples at 6.2 kbar are shown in Fig. 2. The FS and, therefore, the electron orbit topology are different from those at zero pressure. This is, in particular, reflected in the AMRO behavior [23,24]: now the oscillations are mostly determined by closed orbits on the cylindrical FS. Despite the radical modification of the MR behavior upon applying pressure, the major differences between the clean and dirty samples remain the same as in the zero-pressure state. The clean sample exhibits a small narrow peak around $\theta = 90°$ [see the inset of Fig. 2(a)] and a strong dependence on the azimuthal orientation φ. By contrast, the dirty sample shows no coherence peak and is insensitive to φ at sufficiently high tilt angles θ.

The decrease of the MR of the dirty sample, as the field direction approaches the plane of the layers, and its independence of the azimuthal angle φ suggests that it does not feel the magnetic field component parallel to the layers. To check this, we have made θ sweeps at different field values and replotted the resistance as a function of the field projection on the normal to the layers. The result for zero pressure is shown in Fig. 3. Except the vicinities of the magic angles, all the curves, recorded at fields from 2 to 10 T, collapse on a single line. A similar behavior is observed at higher fields in the high-pressure state. Moreover, the curves shown in Fig. 3 nicely coincide with the field dependence $R(B_\perp)$ taken at the field perpendicular to the layers (dashed gray line in Fig. 3). Thus, we conclude that the MR of the dirty sample at high tilt angles is essentially a function of only the field component perpendicular to the layers.

This is a surprising and somehow counterintuitive result. Normally, an in-plane magnetic field acts to confine charge carriers to the layers, thus increasing the interlayer resistivity. The theory predicts a strong linear or superlinear MR in strong fields parallel to the layers, both in the coherent [22,25] and weakly incoherent [7,8] interlayer transport regimes. The exact field dependence is determined by the FS geometry. Since the latter is generally anisotropic in the plane of the layers, the MR strongly depends on the azimuthal orientation of the field [22,26,27]. For the coherent regime, the theoretical predictions are in a good agreement with our results on the clean sample as well as with

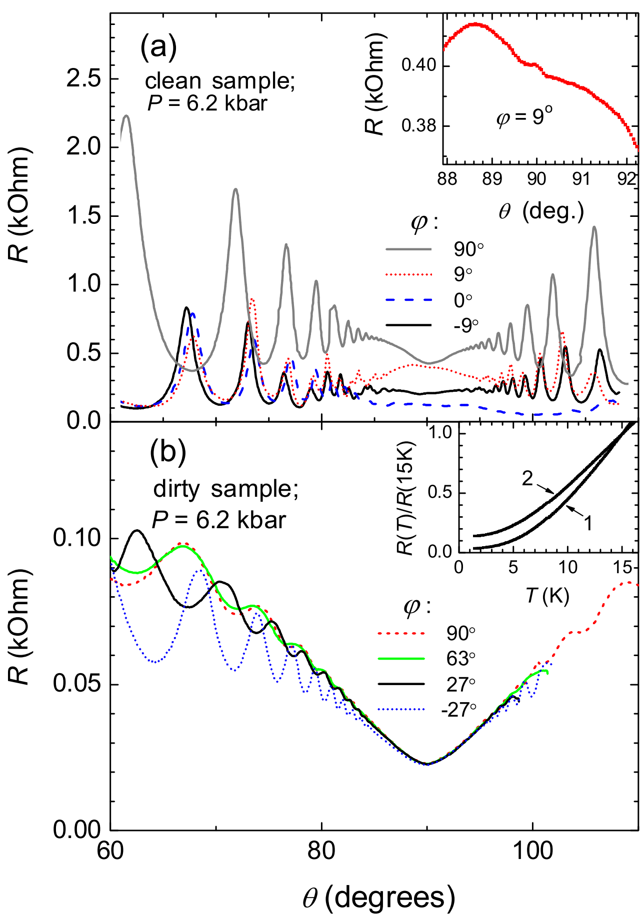

FIG. 2 (color online). Angle-dependent magnetoresistance of (a) the clean sample and (b) the dirty sample at $P = 6.2$ kbar, $B = 20$ T. Insets: (upper panel) fragment of the $\varphi = 9°$ curve for the clean sample with the coherence peak; (lower panel) temperature-dependent zero-field resistance of the clean (curve 1) and dirty (curve 2) samples at the same pressure.

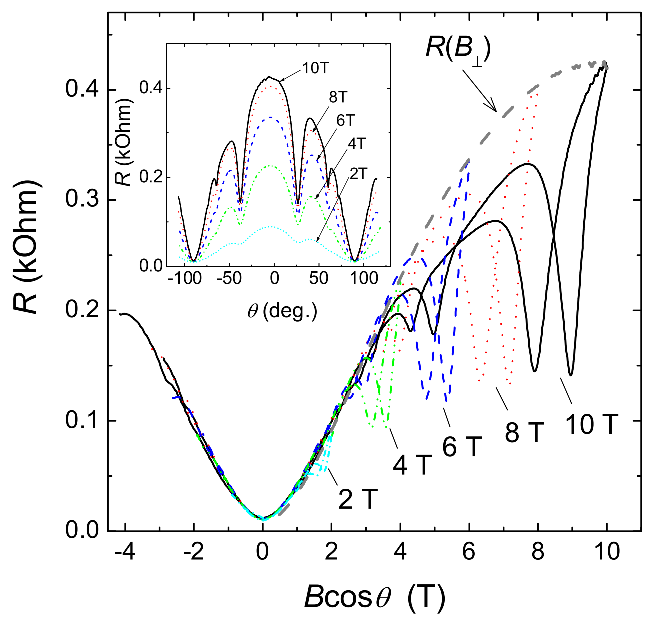

FIG. 3 (color online). Magnetoresistance of the dirty sample at $P = 0$ as a function of the out-of-plane field component. The raw θ sweeps recorded at different field strengths are shown in the inset.

numerous other experiments [14]. This is, however, not the case for the weakly incoherent regime, as follows from the data on the dirty sample. The fact that its resistance is insensitive to the in-plane field is clearly in conflict with the mentioned FL theories.

A tempting possibility would be to invoke the non-Fermi-liquid (NFL) model proposed by Clarke and coworkers [3] to explain a similar anomalous MR in the quasi-1D conductor $(TMTSF)_2PF_6$ [28]. However, the theory [3] suggests incoherence to be a property of *clean* weakly coupled low-dimensional NFLs arising due to strong electron correlations rather than due to disorder. By contrast, we observe the coherence-incoherence transition (or crossover) at increasing disorder in the system. Moreover, in the coherent regime both the AMRO behavior (especially the 2D AMRO in the pressurized state) and the background MR appear to be fully consistent with the FL model.

An important point is that the anomalous behavior of the dirty sample is observed in both the zero- and high-pressure states of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$, characterized by different FS geometries. Moreover, a similar broad dip centered at $\theta = 90°$ was found in the angle-dependent MR of other highly anisotropic materials: the purely quasi-1D compound $(TMTSF)_2PF_6$ [28,29], purely quasi-2D artificial GaAs/AlGaAs superlattice [11], and layered superconductor β''-$(BEDT\text{-}TTF)_2SF_5CH_2CF_2SO_3$, combining open and cylindrical FSs [9]. A similar anisotropy of the interlayer MR, $\rho_\perp(B \parallel I) > \rho_\perp(B \perp I)$, although in the low-field limit, has been reported for the cuprate superconductor $La_{2-x}Sr_xCuO_4$ in the incoherent regime [30]. Kuraguchi *et al.* [11] already noted that a change in the interlayer transfer integral leads to a radical change in the MR anisotropy although their data were not sufficient to establish the independence of the in-plane field component.

In conclusion, our data on the angle-dependent interlayer MR of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$ reveal a dramatic sample dependence which is most likely caused by the crossover between the coherent and weakly incoherent interlayer transport regimes. In the coherent regime the MR is highly sensitive to both the polar and azimuthal orientations of the applied magnetic field that is consistent with the conventional anisotropic 3D FL theory. By contrast, in the weakly incoherent case the nonoscillating MR background does not depend on the azimuthal orientation, in fields strongly inclined towards the layers, and can be scaled by a function of only the out-of-plane field component. This anomalous behavior appears to be a general feature of the weakly incoherent magnetotransport, regardless of the in-plane FS geometry. However, the mechanism responsible for it remains unclear, indicating that a considerable modification of the existing theory is necessary.

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