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Coherent versus incoherent interlayer transport in layered metals

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The magnetic-field, temperature, and angular dependence of the interlayer magnetoresistance of two different quasi-two-dimensional (2D) organic superconductors is reported. For κ-(BEDT-TTF)$_2$I$_3$, where BEDT-TTF is bisethylenedithio-tetrathiafulvalene, we find a well-resolved peak in the angle-dependent magnetoresistance at $\Theta = 90°$ (field parallel to the layers). This clear-cut proof for the coherent nature of the interlayer transport is absent for β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. This and the nonmetallic behavior of the magnetoresistance suggest an incoherent quasiparticle motion for the latter 2D metal.

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The usual fundamental concept describing the electronic transport in metallic crystals is based on the coherent motion of electrons in band or Bloch states. For a number of cases, however, the simple semiclassical Boltzmann transport theory fails and a more complex transport mechanism has to be invoked.[1] Renowned examples are, besides the cuprate superconductors, some quasi-one-dimensional[2–4] (1D) and 2D organic metals[5] revealing non-Fermi-liquid properties. Certain signatures in their interlayer transport suggest an incoherent motion of the charge carriers between the layers. Incoherent interlayer transport is expected when the intralayer scattering rate τ^{-1} is much larger than the interlayer hopping integral t_c ($\hbar/\tau \gg t_c$). In that case the interlayer conductivity is proportional to the tunneling rate between two adjacent layers and a Fermi surface is only defined within the layers.[1] Nevertheless, in case the intralayer momentum is conserved during the tunneling process certain metallic properties persist even without a 3D Fermi surface.

Some potential candidates that might fit into the above scenario are the 2D organic metals and superconductors of the type (BEDT-TTF)$_2X$, where BEDT-TTF is bisethylenedithio-tetrathiafulvalene and X stands for a monovalent anion. Although the observation of magnetic quantum oscillations provides definitive evidence for a well-developed 2D Fermi surface,[1,6] the interlayer transport in some of these 2D conductors might be incoherent. There exists no unequivocal proof for an *incoherent* transport mechanism as proposed in Ref. 1. There are, however, unambiguous tests for *coherent* interlayer transport: (i) beats in magnetic quantum oscillations, (ii) a peak in the angular-dependent magnetoresistance when the magnetic field is parallel to the layers, and (iii) a crossover from a linear to quadratic field dependence of the interlayer magnetoresistance.[1] These features, therefore, can experimentally be utilized to preclude incoherent interlayer transport. Further on, the quantitative analysis of the features (i) and (ii) can be used to ‘‘measure’’ the degree of two dimensionality, i.e., the value of t_c, in layered metals.

Indeed, in a number of 2D organic conductors the occurrence of feature (i) and/or (ii) proved the coherent nature of interlayer transport.[7] For the organic metals investigated here, κ-(BEDT-TTF)$_2$I$_3$ and β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, feature (i) is absent, i.e., no beats were detected in magnetic quantum oscillations down to very low fields,[8–10] which render them possible candidates for metals with incoherently coupled layers. Although some further aspects of their transport properties could not be explained by the usual Fermi-liquid theory,[5,8] results of the other tests (ii) and (iii) have not been reported so far. Here we show that a well-defined 3D Fermi surface exists in κ-(BEDT-TTF)$_2$I$_3$ whereas no indication for a coherent interlayer transport can be detected in β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. Indeed, for the latter material the experimental results clearly reflect properties that are not explicable by conventional Fermi-liquid theory.

Since both metals investigated here are superconductors with $T_c = 3.5$ K [κ-(BEDT-TTF)$_2$I$_3$] and $T_c = 4.4$ K [β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$],[11] sufficiently large magnetic fields have to be applied to attain the normal state for all field orientations. The band-structure parameters of both metals have been measured comprehensively by use of de Haas–van Alphen (dHvA) and Shubnikov–de Haas (SdH) measurements.[8–10] The wave form and the field dependence of the magnetic quantum oscillations could not be described by 3D theories that proved both materials as highly 2D metals.[12,13] The in-plane Fermi surfaces have been mapped out in detail utilizing angular-dependent magnetoresistance oscillations (AMRO).[14,15] The origins of these oscillations were first explained by Yamaji[16] assuming a corrugated 3D Fermi-surface cylinder. If this corrugation ($\propto t_c$) indeed exists and if it is large enough, beats of the magnetic quantum oscillations are expected. The absence of these beats sets an upper limit for t_c (see below). However, since a 3D Fermi surface is not a necessary ingredient to explain AMRO,[1] the specification of a corrugation by t_c might be meaningless; incoherent interlayer transport might be present instead.

The single crystals investigated in this study have been prepared electrochemically as described earlier.[17,18] Thin current leads (15 μm gold wire) were glued with graphite paste

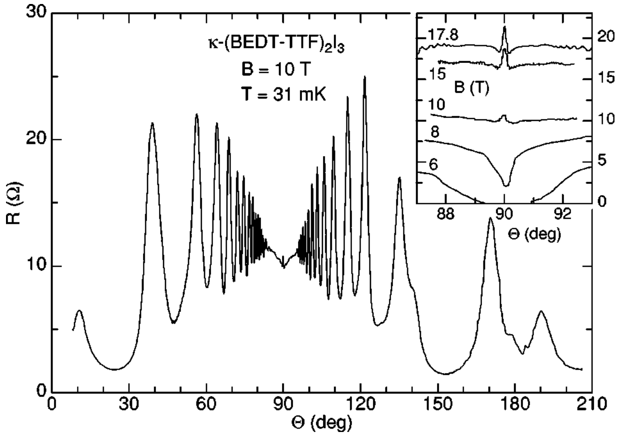

FIG. 1. Angular dependence of the interlayer resistance of κ-(BEDT-TTF)$_2$I$_3$ at $B = 10$ T and $T = 31$ mK. The inset shows the region close to 90° for different magnetic fields. At $B \geqslant 10$ T a clear resistance peak evolves at 90°.

to the samples. The interplane resistance was measured by a four-point method with a current of a few microampere either by use of a low-frequency ac-resistance bridge or a lock-in amplifier. The measurements were performed at the High Magnetic Field Laboratory in Tallahassee in a dilution refrigerator equipped with a superconducting 20 T magnet and in a ^{3}He cryostat in fields up to 33 T. Thereby, the samples could be rotated *in situ* around one axis.

Figure 1 shows the angular dependence of the resistance of κ-(BEDT-TTF)$_2$I$_3$ measured at $B = 10$ T and $T = 31$ mK. The huge oscillations—R changes by more than a factor of 10—are found to be equidistant in $\tan\Theta$, where Θ is the angle between the applied magnetic field B and the normal to the conducting plane. Similar AMRO data with smaller amplitude at $T = 1.6$ K have been reported earlier.[14] The maxima of the oscillations are given by

$$\tan\Theta = \frac{\pi(n \pm 1/4)}{k_B^{max} c'}, \tag{1}$$

where n counts the maxima, c' is the spacing between adjacent layers, and k_B^{max} is the maximum projection of the in-plane Fermi wave vector $k_F(\varphi)$ onto the field-rotation plane. φ is an azimuthal angle. The minus (plus) sign corresponds to positive (negative) angles. This simplified formula is valid when no in-plane component of the hopping vector exists.[19] Here, the linear regression of the peak number n versus $\tan\Theta$ yields $k_B^{max} = 3.36(5) \times 10^9$ m^{-1} with $c' = 1.64$ nm for κ-(BEDT-TTF)$_2$I$_3$. This agrees with the result of Ref. 14 and fits the assumption of an almost circular in-plane Fermi surface with $k_F = k_B^{max} = \text{const}$. In that case k_F is given by $k_F = (2eF/\hbar)^{1/2} = 3.43 \times 10^9$ m^{-1}, with the well-known dHvA frequency of the so-called β orbit of $F = 3870$ T.[6,8,9,12,14]

As mentioned, the bare observation of an AMRO signal is no proof for a 3D Fermi surface. Indeed, for κ-(BEDT-TTF)$_2$I$_3$ no nodes in the dHvA and SdH signals are visible with oscillations of the β orbit starting at about $B_{min} = 2.8$ T.[9] This means that the maximum dHvA-frequency difference is $\Delta F = (3/4) B_{min} = 2.1$ T.[20] Consequently, the estimated corrugation amplitude should be less than $t_c \approx 16$ μeV, since $\Delta F / F = 4 t_c / \epsilon_F$ with the Fermi energy $\epsilon_F = \hbar^2 k_F^2 / 2m^*$ and the effective mass m^* $[= 3.9$ m_e for κ-(BEDT-TTF)$_2$I$_3]$.[6,8] This maximum t_c is indeed much smaller than $\hbar/\tau \approx 0.14$ meV estimated from a Dingle temperature of about 0.25 K corresponding to a scattering time $\tau \approx 4.9 \times 10^{-12}$ s.[8,14] Therefore, according to the so-called Mott-Ioffe-Regel incoherent interlayer transport might be expected. However, looking carefully at the resistance data around 90° (Fig. 1) a small peak can be seen in R. This becomes much clearer in the inset of Fig. 1 where data taken with high angular resolution are shown for different magnetic fields at angles close to 90°. As soon as the superconductivity is quenched completely, the peak at 90° evolves and becomes larger in amplitude at higher fields. This peak definitely proves that the interlayer transport in κ-(BEDT-TTF)$_2$I$_3$ is *coherent*. This and previous results[7] indicate that the τ obtained from a Dingle analysis seems to have no relation to the relevant scattering time in the Mott-Ioffe-Regel. Therefore, this *Regel* should only be used as an order-of-magnitude estimate for possible incoherent transport.

As observed previously for other 2D materials small local minima to the left and right of the 90° peak evolve.[19,21,22] The peak itself is very narrow with a full width between the minima of only about 0.34°, independent of the field strength. Although this is much narrower than reported for any other 2D metal so far, it is broader than expected from the maximum t_c estimated above. Although there is a dispute on whether the physical origin of the 90° peak is due to self-crossing orbits[23] or due to small closed orbits,[22] there is no controversy that the peak occurs only for a 3D warped Fermi surface. Assuming a symmetric cylindrical Fermi-surface topology, Hanasaki *et al.* have derived a relation between the Fermi-surface parameters and the half width of the peak.[22] By use of their equation $\Theta_{peak/2} = t_c c' k_F / \epsilon_F$ with $\Theta_{peak/2} = 0.17°$ in our case, we obtain $t_c \approx 61$ μeV which is about a factor of 4 larger than the maximum t_c estimated from the absence of beating nodes. This difference cannot be explained by an azimuthal, i.e., φ dependence of t_c as observed for Sr$_2$RuO$_4$.[21,24,25] Careful AMRO measurements of another κ-(BEDT-TTF)$_2$I$_3$ sample for different φ resulted—consistently within error bars—in $\Theta_{peak/2} = 0.20(2)°$ independent of the azimuthal angle. Our results indicate that the theories need to be refined for a quantitatively better estimate of t_c.

A *qualitatively* different picture occurs for the 2D organic metal β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. From the absence of beating nodes in dHvA and SdH oscillations that start at about $B_{min} = 1.7$ T with a frequency of $F = 199(1)$ T and a cyclotron effective mass of $m^* = 2.0(1) m_e$,[10] we estimate a maximum $t_c = 18.5$ μeV. Previous AMRO measurements showed that the small Fermi surface—occupying only 5% of the in-plane Brillouin zone—consists of a strongly elongated ellipsoid with an axis ratio of about 1:9 (0.26×10^9 m^{-1} $< k_F < 2.4 \times 10^9$ m^{-1}).[15] In this experiment the applied field of 10 T was not sufficient to suppress superconductivity at

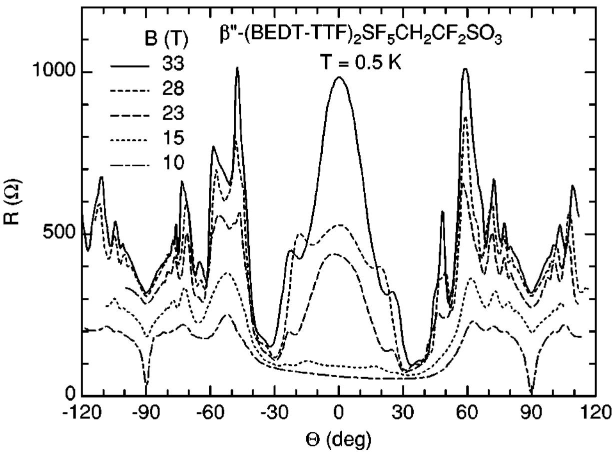

FIG. 2. Angular dependence of the interlayer resistance of β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ at $T = 0.5$ K for different magnetic fields up to 33 T.

90°. At $T = 0.5$ K, fields above about 15 T are necessary to reach the normal state. However, as Fig. 2 shows, even for fields up to 33 T no indication of a peak at 90° appears. With increasing field only AMRO peaks and SdH oscillations become dominant.[26] From a linear regression of the AMRO peak number versus $\tan\Theta$ we obtain $k_B^{max} \approx 1.1 \times 10^9$ m^{-1} with $c' = 1.74$ nm.

The data shown in Fig. 2 were taken at fixed azimuthal angle $\varphi \approx 80°$, where $\varphi = 0$ corresponds to a field rotation through the k_a axis.[15] Since t_c may vary largely with φ, additional AMRO data were collected at a number of different φ. Figure 3 shows the resistance of a second sample for four different φ at $T = 1.3$ K for $B = 23$ T close to $\Theta = 90°$. For all investigated azimuthal angles φ, all magnetic fields, and all samples a peak at 90° never occured. With an approximate angular resolution of 0.01° for the polar angle Θ (R was continuously monitored when the samples were rotated manually), t_c must be smaller than about 10^{-6} eV estimated conservatively by use of above formula for $\Theta_{peak/2}$. This almost two orders of magnitude smaller t_c than observed so far, strongly suggests an *incoherent* interlayer-transport mechanism for β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$.

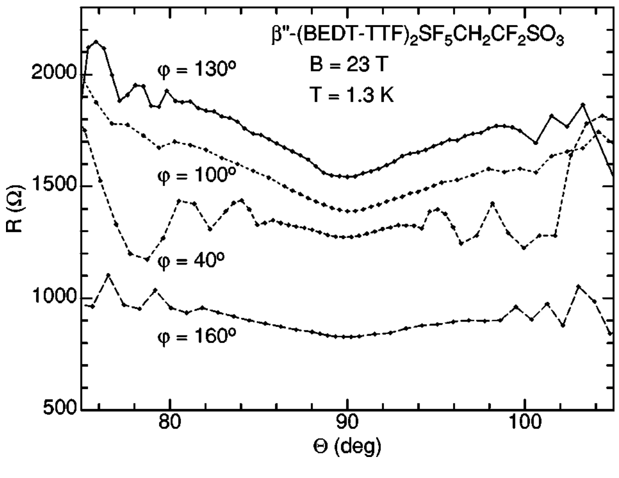

FIG. 3. Interlayer resistance close to 90° of another β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ sample for different azimuthal angles φ.

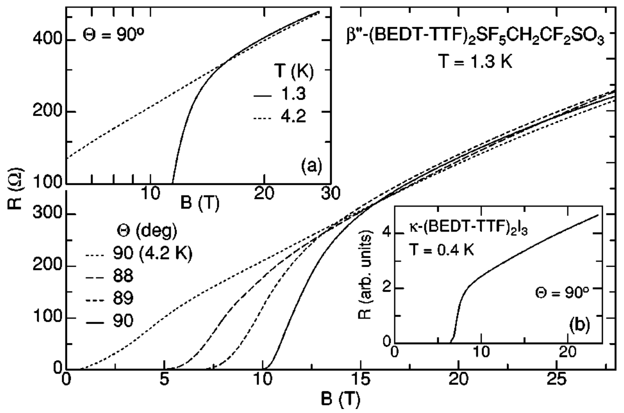

FIG. 4. Field dependence of the interlayer resistance of a third β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ sample close to 90°. The inset (a) shows the data for $\Theta = 90°$ at $T = 1.3$ K and $T = 4.2$ K in a double-logarithmic scale. The inset (b) shows R versus B for κ-(BEDT-TTF)$_2$I$_3$ at 90°.

As a final test for coherent transport [see point (iii) above], we measured carefully the field dependence of the interlayer resistance for fields aligned within the highly conducting planes. Perfect alignment of the samples was easily achieved in fields low enough to retain superconducting traces at $\Theta = 90°$. The resulting data (Fig. 4) show clearly that R at $\Theta = 90°$ grows less than linear with B. For intentionally misaligned field orientations a somewhat steeper, but still less than linear field dependence is observed (see the examples at 88° and 89° in Fig. 4). Thus, there is definitely *no* indication for a crossover to quadratic behavior in B as expected for coherent transport at large fields.[1,27] There is, however, a drawback regarding the relevance of this test: for κ-(BEDT-TTF)$_2$I$_3$, we find almost the same field dependence of R at 90° [inset (b) of Fig. 4] although the peak at 90° proves coherent transport. Equally, for the layered metal Sr$_2$RuO$_4$ not a B^2 behavior but a superlinear B dependence ($\propto B^{1.5}$) was observed.[24] Although $eB\tau/m^* \gg 1$ seems to be fulfilled, larger fields might be necessary to verify the B^2 behavior.[27] A double-logarithmic plot of R versus B [inset (a) of Fig. 4] reveals that in the present case R grows approximately with $B^{0.9}$ at $T = 4.2$ K. However, both in the linear as well as in the double-logarithmic plot clear curvatures of the data are apparent. Above about 20 T, $R \propto \ln B$ fits the data reasonably well (not shown). However, higher fields are necessary to determine the limiting field dependence.

All the above-discussed results give *no* experimental evidence for coherent interlayer transport in the 2D organic metal β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. Along these lines, previous results corroborate the existence of only very weakly coupled perfectly two-dimensional metallic sheets with non-Boltzmann-like interlayer transport. Accordingly, the pronounced two dimensionality of the Fermi surface is evidenced by inverse-sawtooth-like dHvA oscillations which perfectly fit the theoretical prediction for a 2D metal with fixed chemical potential.[13] Further on, deviations from the conventional Bloch-Boltzmann transport theory were observed in the interlayer magnetoresistance for fields close to $\Theta=0$.[5] A field-induced metal-insulator transition and a violation of Kohler's rule was found.[28] All these peculiarities reflect that β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is a highly unusual metal. On the one hand, the interlayer resistance at $B=0$ is metallic from lowest T up to room temperature for all samples we investigated and a 2D in-plane Fermi surface can clearly be resolved. On the other hand, the electronic transport perpendicular to the layers is most probably incoherent and cannot be described by conventional theories.

In conclusion, we proved that the highly 2D organic metal κ-(BEDT-TTF)$_2$I$_3$ has a well-developed 3D Fermi surface and the electronic transport can be described by the coherent motion of electrons in Bloch states. The interlayer overlap integral $t_c \approx 61$ μeV is only slightly smaller than the scattering rate $\hbar/\tau \approx 0.14$ meV setting this material just at the borderline to incoherent electronic transport. The latter seems to occur in the organic metal β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ for which $t_c < 1$ μeV and where all experimental tests to observe signatures for coherent interlayer transport failed.

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