Field-induced metal-insulator transition in a two-dimensional organic superconductor

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The quasi-two-dimensional organic superconductor $\beta''$-(BEDT-TTF)$_2$CF$_2$SO$_4$$_{(CH_2)_{4}}$ shows very strong Shubnikov–de Haas (SdH) oscillations which are superimposed on a highly anomalous steady background magnetoresistance, $R_0$. Comparison with de Haas–van Alphen oscillations allow a reliable estimate of $R_0$ which is crucial for the correct extraction of the SdH signal. At low temperatures and high magnetic fields insulating behavior evolves. The magnetoresistance data violate Kohler’s rule, i.e., cannot be described within the framework of semiclassical transport theory, but converge onto a universal curve appropriate for dynamical scaling at a metal-insulator transition.

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The electrical transport in metals can usually be described by the coherent motion of electrons in Bloch states with well-defined wave vectors. A common approach to this problem is the Boltzmann transport theory which works well for most metals and semiconductors. There are, however, a number of cases where a more complex transport mechanism is involved and where the simple approach fails. Prominent examples are the cuprate superconductors and organic metals which reveal unusual normal-state properties. A central issue for these layered materials is whether the electronic conduction can be described by the coherent motion of Bloch electrons with well-defined wave vectors or whether the interlayer transport is caused by an incoherent diffusive motion of the electrons between the layers.

With the assumption of a constant scattering time $\tau_s$ for all charge carriers the semiclassical transport theory predicts a universal temperature and field dependence of the magnetoresistance which can be described as $R(B,T)/R(0,T) = f(B/R(0,T))$, where $f(x)$ is a universal function. This is known as Kohler’s rule which holds for many metals regardless of the Fermi-surface topology. Furthermore, for $B$ parallel to the current, no magnetoresistance is expected semiclassically. Deviations from this behavior are known to occur for the interlayer transport in some organic conductors. This was taken as an indication for incoherent transport. Other evidence for the failure of conventional transport theory is the very large low-temperature normal-state resistivity (a few $\Omega$cm) which would correspond to mean-free paths much shorter than interatomic distances.

For the quasi-two-dimensional (2D) organic metals one can assume that the interlayer transport is caused by uncorrelated tunneling events between the layers. Thereby the transport is incoherent because the electrons are scattered many times within the layer before a tunneling event takes place. This may occur when the time it takes for an electron to hop between the layers is much larger than $\tau_s$, i.e., $h/t_c \gg \tau_s$, where $t_c$ is the interlayer hopping integral. In case the intralayer momentum is conserved during the tunneling process and an interference between the wave functions on adjacent layers is possible, McKenzie and Moses showed that certain metallic properties persist even though no three-dimensional (3D) Fermi surface would exist.

A potential candidate which might fit into the above scenario is the 2D organic superconductor $\beta''$-(BEDT-TTF)$_2$CF$_2$SO$_4$$_{(CH_2)_{4}}$, where BEDT-TTF stands for bisethylenedithio-tetrathiafulvalene. The Fermi surface has been mapped out by de Haas–van Alphen (dHvA) experiments. Shubnikov–de Haas (SdH) and angular-dependent magnetoresistance oscillations (AMROs) were observed. High-field dHvA measurements proved the existence of an ideal 2D Fermi surface. In line with an incoherent transport mechanism, neither beats in the magnetic quantum oscillations nor a peak in the AMROs for field parallel to the layers was observed. Another not-explained phenomenon is the peak in $R(B)$ at low fields and low temperatures. At zero field, the material seems to be close to an insulating phase, since replacing CH$_2$CF$_2$ in the anion with CH$_2$ yields an insulator.

Although the measured dHvA oscillations of $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_4$ could quantitatively be understood by a 2D theory, the SdH oscillations seemed to show strong deviations in the field and temperature dependence from the usually expected behavior. Similar observations have been reported for the SdH oscillations in other organic metals.

A principal problem inherent to transport data is the correct extraction of the SdH signal, which is given by...
the relative conductance oscillations $\Delta \sigma/\sigma = \sigma/\sigma_b - 1$, with $\sigma_b$ the steady part of the relevant conductance of the band which is responsible for the oscillations \[20\]. Only for $\Delta \sigma/\sigma$ the framework of the Lifshitz–Kosevich (LK) theory can be applied \[13\]. In general, a tensor inversion from the measured longitudinal and transverse resistances is necessary to obtain $\sigma_b$. In the present case, however, the Hall component of the resistivity tensor is negligible and for the used field configuration ($B$ is applied parallel to the interlayer-transport current) it should be even exactly zero. Therefore, the SdH signal is given by $\Delta \sigma/\sigma = R_b/R - 1$, where the main task is reduced to the reliable determination of $R_b = 1/\sigma_b$ out of the measured resistance $R$. In the following we specify a method how this can be reasonably performed. The extracted $R_b$ reveals a field-induced insulating behavior, which cannot be described by the semiclassical transport theory but can be scaled onto a universal curve. The observed dynamical scaling suggests a scenario of this metal-insulator transition as a quantum phase transition.

The $\beta^\prime$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ single crystals (labelled $A$ to $C$ hereafter) were grown by electro-crystallization \[21\]. For the transport measurements 15 µm gold-wire current leads were glued with graphite paste to the samples. $R$ was measured either with a lock-in amplifier or by use of a four-point low-frequency ac-resistance bridge with currents of a few µA. The anisotropic magnetization, $M$, was measured by means of a capacitance cantilever torque magnetometer. Thereby, the contacted crystal $A$ was placed on top of the cantilever plate allowing the simultaneous measurement of both SdH and dHvA signals. The measurements were performed at the High Magnetic Field Laboratories in Grenoble in fields up to 28 T and in Tallahassee up to 30 T.

In first high-field SdH measurements, $R_b$ was estimated by a polynomial fit to the measured $R$ data \[14\]. Thereby the oscillation amplitude of $\Delta \sigma/\sigma$ reduced towards lower temperatures for fields above about 20 T (see Fig. 3 in \[9\]). This is contrary to the standard theories for magnetic quantum oscillations \[22\]. In order to get a better estimate for $R_b$, we simultaneously measured the SdH and dHvA effect of a new and better-quality sample ($A$).

Figure 1 shows the torque signal, $\tau$, (dotted line) and the magnetoresistance (solid line) measured during falling field which was tilted by about $0.4^\circ$ in order to resolve a non-zero dHvA signal. In line with previous results \[10\] the dHvA signal shows an “inverse sawtooth” wave form - after subtraction of a quadratic background magnetoresistance - which can be explained quantitatively by a 2D theory with fixed chemical potential \[10,14\].

The highly asymmetric wave form becomes more apparent in the strong anharmonicities of $B^2dM/dB$, i.e., in the derivative of the dHvA signal times $B^2$ (inset of Fig. 1) which is expected to be directly proportional to the SdH signal $\Delta \sigma/\sigma$ \[13,23\]. With the assumption that the SdH signal should be consistent with the dHvA effect and that the theory for SdH oscillations \[20\] holds even for magnetoresistance oscillations as large as in the present case, the background magnetoresistance $R_b$ can be estimated. The SdH signal $\Delta \sigma/\sigma$ is proportional to the oscillating part of the density of states at the Fermi level $\Delta N(\epsilon_F)/N_0$, where $N_0$ is the steady density of states.

Relevant for the magnetic quantum oscillations in the present case is a single 2D hole-like band, which coexists together with open 1D electron-like bands \[8\]. A priori, both bands are expected to contribute to the electronic transport. If, therefore, the conductivities originating from each band exhibit strong field and temperature dependences, one has to disentangle the different contributions from the measured $R$. For the present case, this means to extract the background resistance for the 2D band. With $R_b$ as shown in Fig. 1 (dashed line), a SdH signal $\Delta \sigma/\sigma$ is obtained which reasonably well agrees with the thermodynamic quantity $B^2dM/dB$ (see

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**FIG. 1.** Field dependence of the simultaneously measured torque signal (dotted line) and resistivity (solid line) of sample $A$. The dashed line is the estimated background magnetoresistance, $R_b$, resulting in the SdH signal $\Delta \sigma/\sigma$ which agrees with the derivative of the dHvA signal times $B^2$ (inset).

**FIG. 2.** Magnetoresistance of sample $B$ for different temperatures. The dashed line shows $R_b$ for $T = 0.55$ K resulting in the SdH signal $\Delta \sigma/\sigma$ (solid line in the upper part) which compares well with the derivative of the independently measured dHvA signal times $B^2$. 

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For this sample and sample $B$ (see below) a simple polynomial fit through the data would yield a smaller $R_0$ and, consequently, a too low $\Delta \sigma/\sigma$ at higher fields.

The estimated $R_0$ for sample $A$ is well inside the range of the resistance-oscillation amplitude. This is different for samples which reveal smaller-amplitude oscillations, i.e., which are of less-high quality. $R_0$ for sample $B$ has to be set at about the maxima of the measured $R$ in order to reproduce in $\Delta \sigma/\sigma$ approximately the field dependence of $B^2dM/dB$ (Fig. 2). For this sample $R$ was measured in Tallahassee and subsequently $\tau$ was measured in Grenoble (sample 2 in Ref. [10]), which explains the slight phase shift. It is obvious that without the knowledge of the dHvA signal the chosen $R_0$ seems to be rather arbitrary.

For the usual polynomial estimate of $R_0$, the $B$ and $T$ dependence of the resulting SdH signal would contradict the LK theory (see Refs. [9,15]). Although there remains an uncertainty in $R_0$ especially towards higher $B$, the comparison of the magnetic quantum oscillations extracted from thermodynamic and transport data indeed allows a reliable estimate of the magnetoresistance.

Finally, Fig. 3 shows the magnetoresistance of a high-quality sample $C$ with very large oscillation amplitudes in $R$ (three times larger than for sample $B$). Here, a polynomial fit for $R_0$ results in a reasonable field and temperature dependence of $\Delta \sigma/\sigma$. The inset of Fig. 3 shows the field dependence of $R_0$ for different temperatures. While for low fields $R_0$ decreases with decreasing $T$ in a metallic-like fashion, insulating behavior at high fields and low temperatures with $dR/dT < 0$ is observed. This suggests a metal-insulator transition as $T \to 0$. At high fields, $R_0$ grows approximately exponentially with field. This field-induced metal-insulator transition, recognized qualitatively already earlier [15], is a unique feature of the present organic metal.

It is important to note that the magnetoresistance cannot be explained by a conventional semiclassical theory. For charge carriers with a constant $\tau_0$ on the whole Fermi surface, the Boltzmann equation predicts that Kohler’s rule is obeyed. However, a Kohler plot of $R_0/R(0)$ vs $B/R(0)$ for different temperatures (inset of Fig. 3), where $R(0)$ for $B = 0$ is extrapolated from $R_0$ at fields above the superconducting phase transition, shows the failure of the semiclassical theory. This leads to the question whether the concept of Bloch states for interlayer transport still has a meaning for the present material [2] and how the observed field-induced metal-insulator transition and the interlayer transport in general can be understood.

One explanation for an anomalously large magnetoresistance is based on the existence of a periodic potential in each layer [24]. A magnetic field applied perpendicular to the layers then converts the in-plane periodic potential into a periodic potential along the field direction. When the period is incommensurate with the layer spacing and when this potential is stronger than the interlayer hopping rate the electron wave function would become localized. The strength of the potential increases with field resulting in an increasing magnetoresistance [24]. For the present material, there is however no indication for the existence of an in-plane periodic potential, caused e.g. by a density wave. Therefore, it is unclear whether this kind of incoherent transport is present here.

The metal-insulator transition has been intensively studied in a number of other materials. For these, usually a universal dynamic scaling relation can be found which describes the resistivity as a function of the tuning parameter and temperature [25]. Thereby, the tuning parameter controls the quantum phase transition, i.e., might be the charge-carrier concentration, pressure, disorder, or the magnetic field. A scaling variable which has been found to hold well for the magnetic-field-tuned superconductor-insulator transition of 2D films is $(B - B_0)/T^\kappa$, where $B_0$ is the critical field for the metal-insulator transition and $\kappa$ is a composed critical exponent [24,25]. A first attempt to scale $R_0$ (inset of Fig. 3) directly as a function of $(B - B_0)/T^\kappa$ did not yield a sat-
isfactory result. However, the data do collapse onto one single curve when normalizing $R_b$ by its value at $B = 0$ (Fig. 3). Thereby, a critical field of $B_0 = 3.5$ T and a critical exponent of $\kappa = 0.65$ was chosen. $R(0)$ lies between 27.5 $\Omega$ for $T = 0.44$ K and 54 $\Omega$ for $T = 4.2$ K. For sample $B$ an equally good scaling was obtained with $B_0 = 3.5$ T and $\kappa = 0.7$. The slight deviations of the different curves towards higher fields may originate from the uncertainty in $R_b$. The scaling works very well over about two decades in $R_b$ and about one decade in $B$ and $T$. For non-zero temperatures above $B_0$ a finite conductivity should be possible only due to hopping processes. Ideally, for $T \to 0$ the magnetoresistance for large enough $B$ should diverge. However, towards lower temperatures the scaling fails and $R_b$ rather seems to saturate. [25-29]

A similar deviation from scaling at low $T$ observed in superconducting 2D films was ascribed to the coupling of the system to a dissipative bath [27].

The field $B_0$ separates the insulating from the metallic behavior. Usually, the data for $B < B_0$ should scale onto a second branch when plotting $R_b/R(0)$ vs $|(B-B_0)/T|^\kappa$. In the present case, where $B_0 = 3.5$ T is approximately equal to the upper critical field for superconductivity [4], the resistivity below $B_0$ is strongly influenced by the vortex dynamics in the superconducting state. Therefore, a reliable scaling for $B < B_0$ is not possible. For the known quantum phase transitions the exponent $\kappa$ can be written as $1/2z\nu$. For the field-tuned metal-insulator transition of strongly disordered films a dynamical critical exponent $z = 1$ and $\nu \geq 1$ for the coherence-length exponent was predicted and found experimentally ($\nu \approx 1.3$) [26,27]. However, recent results deviate from that prediction and suggest a more complex scenario [28]. Although the exponent $\kappa = 0.65(5)$ found here is close to the result of [26,27], the investigated organic system is very clean and the metal-insulator transition will probably fall in a different universality class.

In conclusion, we have shown that for $\beta''(\text{BEDT-TTF})_2SF_5CH_2CF_2SO_3$ the apparent deviation of the SdH oscillations from conventional behavior depends on the sample quality and relies strongly on the correct determination of the background magnetoresistance $R_b$, which can be achieved by a direct comparison with dHvA data. $R_b$ exhibits a field-induced metal-insulator transition which semiclassical theory fails to describe. The magnetoresistance data for different temperatures can reasonably well be scaled onto a universal curve. It remains to be checked whether the unusual behavior of the SdH oscillations observed in other organic metals [16,18] might be understood by a similar scenario.

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