Field-Induced Metal-Insulator Transition in a Two-Dimensional Organic Superconductor

J. Wosnitza,[1] S. Wanka,[1] J. Hagel,[1] H. v. Löhneysen,[1] J. S. Qualls,[2,*] J. S. Brooks,[2] E. Balthes,[3] J. A. Schlueter,[4] U. Geiser,[4] J. Mohtasham,[5] R. W. Winter,[5] and G. L. Gard[5]

[1]*Physikalisches Institut, Universität Karlsruhe, D-76128 Karlsruhe, Germany*
[2]*National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32306*
[3]*Grenoble High Magnetic Field Laboratory, MPI-Festkörperforschung and C.N.R.S., F-38042 Grenoble, France*
[4]*Chemistry and Materials Science Divisions, Argonne National Laboratory, Argonne, Illinois 60439*
[5]*Department of Chemistry, Portland State University, Portland, Oregon 97207*

(Received 8 June 2000)

The quasi-two-dimensional organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ ($T_c \approx 4.4$ K) shows very strong Shubnikov–de Haas (SdH) oscillations which are superimposed on a highly anomalous steady background magnetoresistance, R_b. Comparison with de Haas–van Alphen oscillations allows a reliable estimate of R_b 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.

DOI: 10.1103/PhysRevLett.86.508 PACS numbers: 74.70.Kn, 71.30.+h, 72.15.Gd

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 [1,2]. Prominent examples are the cuprate superconductors [3] and organic metals [4] 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 τ_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 [5]. 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 [2,4]. 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 Ω 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 [1]. 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 τ_s, i.e., $\hbar/t_c \gg \tau_s$, 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 [1] 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 β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ ($T_c = 4.4$ K) [6], where BEDT-TTF stands for bisethylenedithio-tetrathiafulvalene. The Fermi surface has been mapped out by de Haas–van Alphen (dHvA) [7], Shubnikov–de Haas (SdH) [8], and angular-dependent magnetoresistance oscillations (AMROs) [9]. High-field dHvA measurements proved the existence of an ideal 2D Fermi surface [10]. In line with an incoherent transport mechanism [1] 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 [11]. At zero field, the material seems to be close to an insulating phase, since replacing CH_2CF_2 in the anion with CH_2 yields an insulator [12].

Although the measured dHvA oscillations of β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ [10] could quantitatively be understood by a 2D theory [13,14], the SdH oscillations seemed to show strong deviations in the field and temperature dependence from the usually expected behavior [9,15]. Similar observations have been reported for the SdH oscillations in other organic metals [16–19]. A principal problem inherent to transport data is the correct extraction of the SdH signal, which is given by the relative conductance oscillations $\Delta\sigma/\sigma = \sigma/\sigma_b - 1$,

 0031-9007/01/86(3)/508(4)$15.00

with σ_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 σ_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 β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ single crystals (labeled A to C hereafter) were grown by electrocrystallization [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 [9]. 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 effects of a new and better-quality sample (A).

Figure 1 shows the torque signal, τ (dotted line), and the magnetoresistance (solid line) measured during falling field which was tilted by about 0.4° in order to resolve a nonzero dHvA signal. In line with previous results [10] the dHvA signal shows an "inverse sawtooth" wave form — after subtraction of a quadratic background magnetization — 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 $B^2 dM/dB$, i.e., in the derivative of the dHvA signal times B^2 (inset in 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

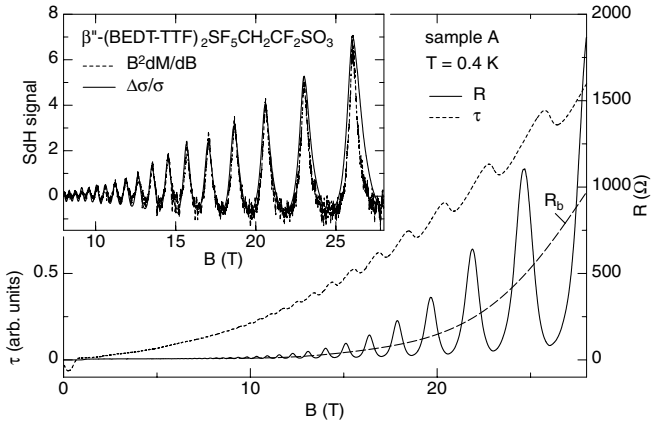

FIG. 1. Field dependence of the simultaneously measured torque signal (dotted line) and resistivity (solid line) of sample A. The dashed line is the estimated background magnetoresistance, R_b, resulting in the SdH signal $\Delta\sigma/\sigma$ which agrees with the derivative of the dHvA signal times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

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 holelike band, which coexists together with open 1D electronlike 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 $B^2 dM/dB$ (see inset). For this sample and sample B (see below) a simple polynomial fit through the data would yield a smaller R_b and, consequently, a too low $\Delta\sigma/\sigma$ at higher fields.

The estimated R_b 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_b 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 $B^2 dM/dB$ (Fig. 2). For this sample R was measured in Tallahassee and subsequently τ 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_b seems to be rather arbitrary. For the usual polynomial estimate of R_b, 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_b 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.

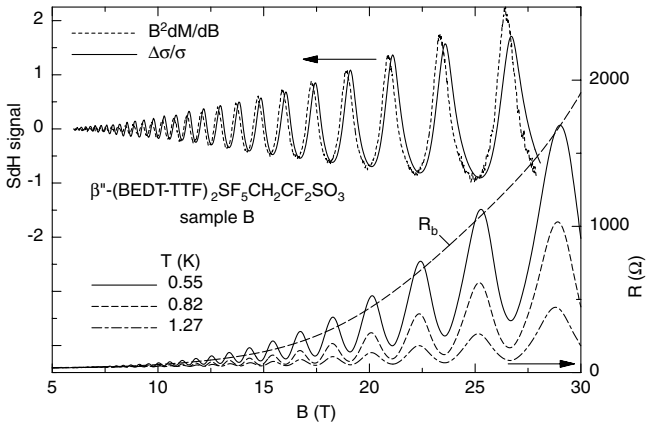

FIG. 2. Magnetoresistance of sample B for different temperatures. The dashed line shows R_b for $T = 0.55$ K resulting in the SdH signal $\Delta\sigma/\sigma$ (solid line in the upper part) which compares well with the derivative of the independently measured dHvA signal times B^2.

Finally, Fig. 3 shows the magnetoresistance of a high-quality sample (C) with very large oscillation amplitudes in R (3 times larger than for sample B). Here, a polynomial fit for R_b results in a reasonable field and temperature dependence of $\Delta\sigma/\sigma$. The inset in Fig. 3 shows the field dependence of R_b for different temperatures. While for low fields R_b decreases with decreasing T in a metalliclike 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_b 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 τ_s on the whole Fermi surface, the Boltzmann equation predicts that Kohler's rule is obeyed. However, a Kohler plot of $R_b/R(0)$ vs $B/R(0)$ for different temperatures (inset in Fig. 4), where $R(0)$ for $B = 0$ is extrapolated from R_b 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 κ is a composed critical exponent [26–28]. A first attempt to scale R_b (inset in Fig. 3) directly as a function of $(B - B_0)/T^\kappa$ did not yield a satisfactory result. However, the data do collapse onto one single curve when normalizing R_b by its value at $B = 0$ (Fig. 4). Thereby, a critical

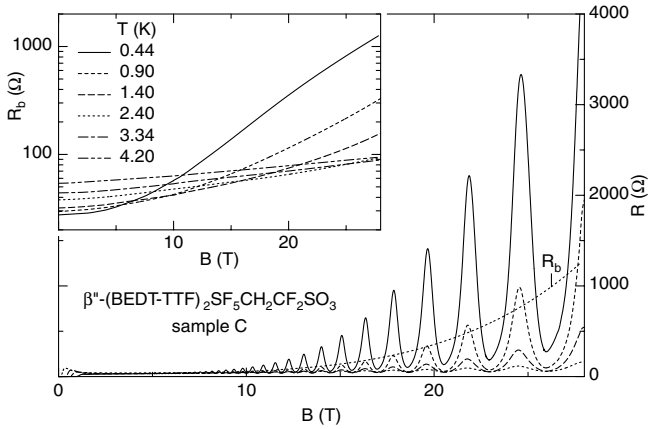

FIG. 3. Magnetoresistance of sample C for different temperatures. The dotted line shows R_b at $T = 0.44$ K which is also shown in a semilogarithmic scale in the inset together with R_b for other temperatures.

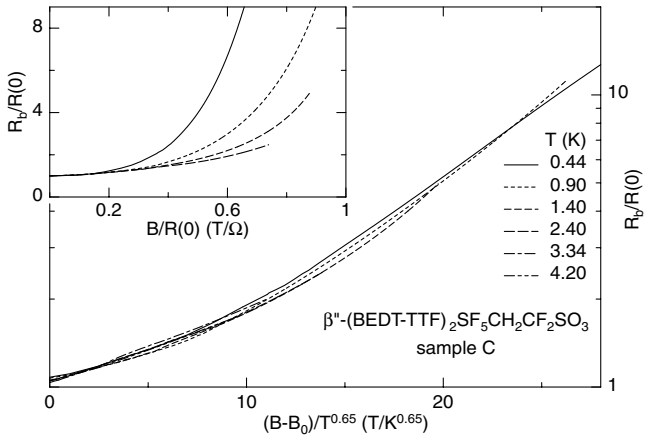

FIG. 4. Scaling plot of the data shown in the inset in Fig. 3. For $B_0 = 3.5$ T and an exponent $\kappa = 0.65$ the data collapse on a single curve. A Kohler plot of the same data (inset) reveals the significant deviation from the semiclassical theory.

field of $B_0 = 3.5$ T and a critical exponent of $\kappa = 0.65$ was chosen. $R(0)$ lies between 27.5 Ω for $T = 0.44$ K and 54 Ω 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 nonzero temperatures above B_0 a finite conductivity should be possible only due to hopping processes. Ideally, for $T \rightarrow 0$ the magnetoresistance for large enough B should diverge. However, towards lower temperatures the scaling fails and R_b rather seems to saturate [15,29]. 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 [6], 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 κ can be written as $1/z\nu$. 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 β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ 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.

We thank R. H. McKenzie for enlightening discussions. The work at Karlsruhe was supported by the DFG and the TMR Programme of the European Community (Contract No. ERBFMGECT950077). J. S. Q. was supported by Grant No. NSF-DMR-10427. We acknowledge the National Science Foundation, the state of Florida, and the U.S. Department of Energy for support of the National High Magnetic Field Laboratory. Work at Argonne National Laboratory was supported by the U.S. Department of Energy (W-31-109-ENG-38). Work at Portland State University was supported by NSF (Che-9904316) and the Petroleum Research Fund (ACS-PRF No. 34624-AC7).

*Present address: Wake Forrest University, Winston-Salem, NC 27109.

[1] R. H. McKenzie and P. Moses, Phys. Rev. Lett. **81**, 4492 (1998); P. Moses and R. H. McKenzie, Phys. Rev. B **60**, 7998 (1999), and references therein.
[2] R. H. McKenzie *et al.,* Phys. Rev. B **57**, 11 854 (1998).
[3] P. W. Anderson, *The Theory of Superconductivity in the High-T_c Cuprates* (Princeton University Press, Princeton, 1997).
[4] S. P. Strong *et al.,* Phys. Rev. Lett. **73**, 1007 (1994); D. G. Clarke *et al.,* Science **279**, 2071 (1998).
[5] A. B. Pippard, *Magnetoresistance in Metals* (Cambridge University Press, Cambridge, England, 1989).
[6] S. Wanka *et al.,* Phys. Rev. B **57**, 3084 (1998).
[7] J. Wosnitza *et al.,* Physica (Amsterdam) **246B–247B**, 104 (1998).
[8] D. Beckmann *et al.,* Eur. Phys. J. B **1**, 295 (1998).
[9] J. Wosnitza *et al.,* Synth. Met. **103**, 2000 (1999).
[10] J. Wosnitza *et al.,* Phys. Rev. B **61**, 7383 (2000).
[11] X. Su *et al.,* Phys. Rev. B **59**, 4376 (1999).
[12] B. H. Ward *et al.,* Chem. Mater. **12**, 343 (2000).
[13] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, England, 1984).
[14] N. Harrison *et al.,* Phys. Rev. B **54**, 9977 (1996).
[15] F. Zuo *et al.,* Phys. Rev. B **60**, 6296 (1999).
[16] V. N. Laukhin *et al.,* Physica (Amsterdam) **211B**, 282 (1995).
[17] P. S. Sandhu *et al.,* Surf. Sci. **361–362**, 913 (1996).
[18] E. Balthes *et al.,* Z. Phys. B **99**, 163 (1996); E. Balthes *et al.,* Europhys. Lett. **47**, 70 (1999).
[19] N. Harrison *et al.,* Phys. Rev. B **58**, 10 248 (1998).
[20] E. N. Adams and T. D. Holstein, J. Phys. Chem. Solids **10**, 254 (1959).
[21] U. Geiser *et al.,* J. Am. Chem. Soc. **118**, 9996 (1996).
[22] The explanation [S. Tanuma and R. Inada, Prog. Theor. Phys. Suppl. **57**, 231 (1975)] for a similar effect found in Bi is not applicable for the present 2D material.
[23] The relation $\Delta\sigma/\sigma \propto B^2 dM/dB$ can easily be derived from Eqs. (4.42) and (2.166) of Ref. [13]; see also L. M. Roth and P. N. Argyres, in *Semiconductors and Semimetals* (Academic Press, New York, 1966), Vol. 1, p. 159.
[24] D. Yoshioka, J. Phys. Soc. Jpn. **64**, 3168 (1995).
[25] S. Sachdev and O. A. Starykh, Nature (London) **405**, 322 (2000); S. Waffenschmidt *et al.,* Phys. Rev. Lett. **83**, 3005 (1999), and references therein.
[26] M. P. A. Fisher, Phys. Rev. Lett. **65**, 923 (1990); A. F. Hebard and M. A. Paalanen, *ibid.* **65**, 927 (1990).
[27] N. Mason and A. Kapitulnik, Phys. Rev. Lett. **82**, 5341 (1999).
[28] N. Marković *et al.,* Phys. Rev. B **60**, 4320 (1999).
[29] J. Wosnitza *et al.* (unpublished).