DC and high-frequency conductivity of the organic metals \( \beta''-(\text{BEDT-TTF})_2\text{SF}_5R\text{SO}_3 \) \((R = \text{CH}_2\text{CF}_2 \text{ and CHF})\)

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Received: September 12, 2008

Abstract. The temperature dependences of the electric-transport properties of the two-dimensional organic conductors \( \beta''-(\text{BEDT-TTF})_2\text{SF}_2\text{CH}_2\text{CF}_2\text{SO}_3, \beta''-(\text{d}_{\text{B}} \text{BEDT-TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3, \) and \( \beta''-(\text{BEDT-TTF})_2\text{SF}_5\text{CHFSO}_3 \) are measured by dc methods in and perpendicular to the highly-conducting plane. Microwave measurements are performed at 24 and 33.5 GHz to probe the high-frequency behavior from room temperature down to 2 K. Superconductivity is observed in \( \beta''-(\text{BEDT-TTF})_2\text{SF}_2\text{CH}_2\text{CF}_2\text{SO}_3 \) and its deuterated analogue. Although all the compounds remain metallic down to low-temperatures, they are close to a charge-order transition. This leads to deviations from a simple Drude behavior of the optical conductivity which become obvious already in the microwave range. In \( \beta''-(\text{BEDT-TTF})_2\text{SF}_2\text{CH}_2\text{CF}_2\text{SO}_3 \) for instance, charge fluctuations cause an increase in microwave resistivity for \( T < 20 \text{ K} \) which is not detected in dc measurements. \( \beta''-(\text{BEDT-TTF})_2\text{SF}_5\text{CHFSO}_3 \) exhibits a simple metallic behavior at all frequencies. In the dc transport, however, we observe indications of localization in the perpendicular direction.

PACS. 71.10.Hf Non-Fermi-liquid ground states – 71.30.+h electron phase diagrams and phase transitions in model systems, Metal-insulator transitions and other electronic transitions – 74.70.Kn Organic superconductors

1 Introduction

Superconductivity has been discovered in organic crystals almost three decades ago; nevertheless they are still considered as novel materials with exotic properties because many questions remain unanswered in spite of the enormous progress achieved over these years \cite{1}. It became clear that the normal-state properties are distinct from those of conventional metals and severely influence the superconducting ground state. Several issues of crucial importance for the understanding of low-dimensional electron systems have been identified that are currently investigated in few distinct families of organic conductors, which serve as prototypes \cite{2}.

The \( \kappa \)-phase BEDT-TTF salts (where BEDT-TTF or ET stands for bis-(ethylenedithio)tetrathiafulvalene), for instance, are two-dimensional metals with a half-filled conduction band and a superconducting transition temperature \( T_c \) of almost 13 K. The system is close to a metal-insulator transition driven by electronic correlations \( U \). At room temperature the transport reflects more a semiconducting behavior, often described as a bad metal, and only below \( 100 \text{ K} \) coherent transport starts to emerge with a narrow Drude-like contribution developing as the temperature is reduced further \cite{3,4,5}. By now there is a good agreement between experiment and theory \cite{6} that the \( \kappa \)-salts represent a prime example of a band-width controlled Mott metal-insulator transition in two dimensions, and the correlated carriers in the narrow zero-frequency contribution of the optical conductivity exhibit Fermi-liquid behavior.

The \( \alpha \)-phase BEDT-TTF compounds are quarter-filled systems, which are subject to charge order depending on the ratio of nearest-neighbor interaction \( V \) to bandwidth \( W \) \cite{7}: while \( \alpha-(\text{BEDT-TTF})_2\text{I}_3 \) becomes completely insulating below a sharp metal-insulator transition at 135 K \cite{8}, \( \alpha-(\text{BEDT-TTF})_2(\text{NH}_4)_4\text{H}_2(\text{SCN})_4 \) is an ambient pressure superconductor with \( T_c \approx 1 \text{ K} \) \cite{10}. If in the anions

\[ \begin{align*}
\kappa & \quad \alpha \\
\beta'' & \quad \beta''
\end{align*} \]

Fig. 1. The BEDT-TTF molecules are arranged in different patterns for the various phases.
(NH)₄ is substituted by Tl, Rb, or K, superconductivity is suppressed but metallic properties are conserved all the way down to lowest temperatures. Accordingly, the optical conductivity exhibits a narrow Drude-like component within a charge-order pseudogap [11][12][13]. It was proposed that in these systems superconductivity is mediated by charge-order fluctuations [13], and recent experiments [13][15] provide strong evidence pointing in this direction.

In order to clarify how general this proposal is, other quarter-filled compounds have to be investigated that also show charge order, like the θ or ρ′ phases of the BEDT-TTF salts. In the case of the ρ′′ phase, there is only one kind of stack with two BEDT-TTF molecules per unit cell [10] as depicted in Fig. 1. The planar BEDT-TTF kind of stack with two BEDT-TTF molecules per unit cell [16] causes a metal to insulator transition near 180 K, while showing charge order, like the lic compound (d₄)-(BEDT-TTF)₃SF₅(CH₂CF₂)SO₃. Single crystals of the β″- and deuterated analog (d₈)-(BEDT-TTF)₃SF₅(CH₂CF₂)SO₃, as well as the metallic compound (d₄)-(BEDT-TTF)₂SF₅(CH₂CF₂)SO₃, have been grown via electrochemical techniques in an H cell at Argonne National Laboratory as described in Ref. [12]. The crystals have typical dimensions of 0.5 × 1.5 × 0.25 mm² and form plates with a large face containing the conducting (ab) plane. We have performed temperature-dependent measurements of the dc resistivity of these crystals within the plane and perpendicular to it (c direction) using the standard four-point method with a typical current of 50 μA. The contacts were made by evaporating 50 Å thick gold pads on the crystal, then 15 μm gold wires were pasted on each pad with a small amount of carbon paint; in some cases the leads were directly put onto the crystal. The samples were slowly (0.2 - 0.5 K/min) cooled down to avoid cracks and ensure thermal equilibrium. Temperature-dependent measurements were conducted in a He exchange gas cryostat down to 2 K; in general data were acquired upon cooling and warming.

It is a well-known problem that the dc transport measurements of highly-anisotropic samples are hampered by contributions from other directions. This is particularly important when the resistivity is measured along the highly-conducting chains (or layers), because there is no guarantee that the same chain (or layer) is contacted by the four leads. Any interruption, stack fault, kink, step, terraces, or other discontinuity causes contributions of the interchain (or interlayer) resistivity. The second problem common to organic but also inorganic crystals is how to achieve low-resistance contacts which inject the current homogeneously but do not cause cracks and non-linearities. For that reason contactless microwave methods have been developed by Schegolev et al. for investigating the electronic transport in small and highly anisotropic crystals [22][23]. It should be noted, that dc measurements probe the voltage drop for a fixed electrical current and thus are sensitive to any additional resistance added by interchanging the conducting chains (or layers), for instance; i.e., they measure the highest resistance for a given conductance path. In contrast, microwave methods are sensitive to the current induced by the electric field and thus probe the highest conduction, weighted by the respective volume fraction, of course. An interruption of a metallic sample by cracks, for instance, does not influence the microwave properties since the high frequencies easily bridge small gaps.

The microwave conductivity was obtained by the cavity-perturbation method utilizing two different cylindrical copper cavities which resonate in the TE₀₁₁ mode at 24 and 33.5 GHz, respectively. They are fed by an Agilent E8257D analog signal generator via suitable rectangular waveguides and operate in the transmission mode. The coupling is about 10% and done through two holes in the sidewalls. The crystals are positioned in the maximum of the electric field placed onto a quartz substrate (0.07 mm thick) in such a way that the electric field is parallel to the b axis. The samples were cooled down slowly (0.2 K per minute) from 300 K to 2 K by coupling to the liquid helium bath with the help of low-pressure He exchange gas and by utilizing a regulated heater. The stability is better than 10 mK [24]. By recording the center frequency f and the width Γ (FWHM) of the resonance curve as a function of temperature and comparing them to the corresponding parameters of an empty cavity (f₀ and Γ₀), the complex electrodynamic properties of the sample, like the surface impedance, the conductivity and the dielectric constant, can be determined by cavity-perturbation theory; further details on microwave measurements and the data analysis are extensively discussed in Ref. [25][26]. Complementary ESR experiments have been performed down to T = 5 K.
3 Results and Discussion

3.1 Organic Superconductor Close to Charge Ordering: $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$

In Fig. 2 the in-plane and out-of-plane dc resistivity of $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is plotted as a function of temperature. The absolute value of $\rho_a = 2.6 \, \Omega \cdot \text{cm}$ at room temperature is comparable to the one reported in literature [27,28]. Previous experiments [29,30,31,32] found a metallic temperature dependence of the $c$-axis resistivity very similar to our data, except for a higher absolute value. The room-temperature anisotropy within one crystal is $\rho_c/\rho_a \approx 140$. Analyzing the asymmetry factor of the Dysonian line shape obtained in X-band ESR experiments, Wang et al. [33] estimated an in-plane anisotropy $\sigma_{\text{max}}/\sigma_{\text{min}}$ of 1.35, with the maximum conductivity in $b$ direction and $\sigma_{\text{min}}$ basically parallel to the stacks [34]. This behavior is confirmed by optical measurements [27,35] and expresses the fact that the orbital overlap between the stacks is stronger than along the stacks. The resistivity ratio $R(300 \, \text{K})/R(6 \, \text{K})$ is approximately 50 and 150 for the $a$ axis and 150 for interlayer transport. At low temperatures, $\rho(T)$ follows a linear behavior. Hagel et al. [32] report a $T^2$ behavior below 10 K which becomes linear above; the temperature region of the quadratic dependence could be extended upon applying external pressure. The critical temperature of the superconducting transition $T_c = 5.9 \, \text{K}$ is determined by the resistivity drop in both directions; its comparably high value [19,20] evidences the excellent quality of the single crystals. As shown in the insets, the width of the superconducting transition is $\Delta T_c \approx 1 \, \text{K}$.

It is interesting to note that for both directions the resistivity $\rho(T)$ increases more or less linearly with temperature up to approximately 125 K. It was theoretically predicted [15,36] and for various $\alpha$-salts experimentally observed [13] that the $T^2$ dependence of a regular metal changes to a linear temperature dependence of the scattering rate when charge-order fluctuations become important. For $\rho_c(T)$ we may identify a second change in slope around $T = 200 \, \text{K}$. From optical experiments, we see first indications that the infrared-active vibrational features $\nu_{3\sigma}(\text{B})$ split already around 200 K [37]; at $T = 150 \, \text{K}$ two Raman modes are clearly distinguished, which evidences charge disproportionation [35]. Other indications of charge fluctuations come from the electronic bands.

The microwave properties obtained at $f = 24.0$ and 33.5 GHz along the $b$ axis are presented in Fig. 3 the data are normalized to the room-temperature value because the uncertainty in absolute values is large (and can exceed a factor of 10) due to errors in the depolarization factor caused by the irregular shape of the samples. Nevertheless, from our data we roughly estimate $\rho(300 \, \text{K}) = 30 \, \text{m}\Omega \cdot \text{cm}$ which is about one order of magnitude less than the dc values. Starting at ambient temperature, the resistivity drops continuously down to $T = 30 \, \text{K}$ as expected for a metal. This change is larger for lower frequencies. A similar tendency is in general observed in unconventional metals and indicates a strong frequency dependence even in the microwave range.

In both microwave measurements, at 24 and 33.5 GHz, the resistivity exhibits a minimum around $T = 20$ to 30 K and turns up by approximately 20 to 30% when cooled down further; this feature is robust. We relate the resistivity upturn to charge-order fluctuations which may be even responsible for superconductivity. From optical experiments (infrared and Raman vibrational spectroscopy) it is known that $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is subject to charge ordering around 125-150 K [35]. From that we would expect that a pseudogap opens which causes a reduction of the density of states and hence a lower conductivity. However, the coherent-particle zero-frequency transport is not affected; and the low-frequency optical measurements also give clear evidence for a Drude-like peak. Alternatively, the increased microwave resistivity...
behavior infers charge fluctuations being responsible for this behavior.

3.2 Organic Superconductor: $\beta''$-(d$_8$-BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$

The dc-transport properties of the deuterated organic superconductor $\beta''$-(d$_8$-BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ are displayed in Fig. 4, the current is either directed within the planes or perpendicular to them, as indicated. For the highly-conducting $b$ direction a simple metallic response is observed with a resistivity ratio $R(300\, K)/R(6\, K) = 100$. The exponent $\alpha$ in the power law $\rho(T) \propto T^\alpha$ is slightly below 2. Measurements of magnetic quantum oscillations evidenced virtually no difference in the low-temperature band-structure parameters compared to the hydrogenated sister compound. At $T_c = 5.6\, K$ the onset of the superconducting transition is observed which is about 1 K broad; a somewhat lower critical temperature was reported previously based on ac-susceptibility measurements [38]. In the $c$ direction the resistivity is very large indicating the pronounced two-dimensionality of the material. Cooling down from ambient temperature the resistivity increases first, goes through a maximum around $T = 200\, K$ and then continuous to drop down to lowest temperatures in a linear fashion, before it exhibits superconductivity [39].

Fig. 5 shows the microwave resistivity of $\beta''$-(d$_8$-BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ measured along the highly conducting $b$ direction at 24 and 33.5 GHz. At room temperature the absolute values are approximately 0.1 $\Omega\, cm$; i.e. they are comparable to the dc resistivity. While the resistivity ratio is similar in both experiments $[\rho(300\, K)/\rho(6\, K) \approx 10]$, at $f = 24\, GHz$ we observe a dramatic change at temperatures above 200 K while the higher frequency data drop gradually in a more or less quadratic temperature behavior. This confirms the general behavior already mentioned for $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, that changes in resistivity with temperature become smeared out as the measurement frequency increases. The transition to the superconducting phase is detected at 5.6 K. The microwave resistivity flattens out below 50 K and exhibits only a very weak temperature dependence. But most important, there are no indications of a resistivity upturn as observed in the hydrogenated analogue (cf. Fig. 3). With other words, since the deuterated crystals remain metallic even in their microwave properties, they seem to be less susceptible to charge fluctuations.

A careful investigation [38] of the isotope shift in a large number of BEDT-TTF samples revealed that for $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ the deuteration causes an increase of the superconducting transition temperature by about 0.3 K. From pressure-dependent measurements [81] we know that the superconducting transition temperature decreases with pressure by a rate of -1.3 K/bar. This is due to the weaker influence of the effective inter-site Coulomb repulsion $V/t$ as $t$ increases with pressure; the systems becomes more metallic [41]. The increase of
$T_c$ would therefore infer that $\beta''$-(ds-BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is closer to the charge-order transition. However, due to the known sample-to-sample variation, we cannot make any final statement in this regard and have to leave this discrepancy unresolved.

### 3.3 Organic Metal: $\beta''$-(BEDT-TTF)$_2$SF$_5$CHFSO$_3$

Replacing the (SF$_5$CH$_2$CF$_2$SO$_3$)$^-$ anions in the organic crystals by (SF$_5$CHFSO$_3$)$^-$ leads to a non-superconducting compound. From room temperature down to approximately 50 K, the electrical resistivity of $\beta''$-(BEDT-TTF)$_2$SF$_5$CHFSO$_3$ exhibits a metallic temperature dependence in both directions, parallel and perpendicular to the highly conducting (ab) plane. The temperature dependences are presented in Fig. 5. Along the $b$ axis, the absolute value $\rho_b(300 \text{ K}) = 2.3 \Omega \text{cm}$ is comparable to the resistivity measured for $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$; the ratio $\rho_b(300 \text{ K})/\rho_b(30 \text{ K}) = 15.3$. Below 50 K the resistivity seems to saturate. A rise of the low-temperature resistivity was reported by Ward et al. [20,21] without providing an explanation. They also mention that the spin susceptibility slightly increases below 12 K which indicates that electron localization takes place at very low temperatures. We cannot confirm this finding from X-band ESR measurements on our samples, as demonstrated in Fig. 7. In the perpendicular direction, $\rho_c(T)$ also shows a decrease in resistivity when cooled down, as plotted in Fig 6b. The room-temperature anisotropy $\rho_c/\rho_b$ of $\beta''$-(BEDT-TTF)$_2$SF$_5$CHFSO$_3$ is approximately 200, i.e. somewhat higher than for $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. For $T < 50 \text{ K}$ an insulating behavior is observed with a rapid increase in resistivity well above the room-temperature value; no simple thermally activated behavior can be determined; nevertheless the initial slope corresponds to an activation energy $E_a = 25 \text{ meV}$, when fitted by $\rho_c(T) \propto \exp{E_a/k_BT}$.

It should be noted that for some of the samples (two out of eight) a different temperature behavior of the in-plane resistivity was measured which resembles previous reports [20]. Starting from approximately the same ambient-temperature value, $\rho_b(T)$ decreases slightly when the sample is cooled down to 250 K. Then the electrical resistivity becomes semiconducting and gradually increases by about a factor of 4 all the way down to 70 K where a broad maximum if found (Fig 8). While Ward et al. [20] could identify an activated behavior between $T = 100$ and 300 K with $E_a = 56 \text{ meV}$, the increase we observe does not correspond to a single activation energy; only between 150 and 200 K the behavior may be described by $\rho_b(T) \propto \exp{E_a/k_BT}$ with $E_a = 26 \text{ meV}$. The drop in $\rho_b(T)$ observed at lower temperatures exhibits a kink around 16 K which might be an indication of a phase transition. At this point we do not have a definite expla-
nation, but the absence of any change in the ESR data around this temperature (Fig. 4) is an argument against magnetic order \[12\].

Microwave measurements of \(\beta''\)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\) in the highly-conducting plane reveal a metallic behavior down to lowest temperatures (Fig. 9). No indication of charge order could be detected. Again, for \(f = 33.5\) GHz the temperature dependence of the resistivity changes more gradually (almost linearly), while for 24 GHz we observe a fast drop below room temperature with only little variation for \(T < 150\) K. At low temperature we did not find an upturn in resistivity as present in the dc data (cf. Fig. 9). This basically implies that the semiconducting behavior shows up only for \(\omega \rightarrow 0\) while in ac-transport and optical experiments the charge localization can be overcome.

It should be pointed out that for the superconducting crystals \(\beta''\)-(BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) the tendency was opposite: there charge fluctuations led to a rise in high-frequency resistivity upon cooling below \(T = 20\) K prior to the superconducting transition (see Sec. 3) and in particular Fig. 3 while the dc resistivity remains purely metallic. In the present case of \(\beta''\)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\), we feel that the strongly insulating behavior observed in the perpendicular direction (Fig. 6) also affects \(\rho_b\) because due to unavoidable crystal imperfections (e.g. cracks) the current path might have to change planes. This is not the case for microwave measurements which are not sensitive to these imperfections and probe the electrical conduction more directly as discussed in Sec. 2.

4 Conclusions

A comprehensive investigation of transport properties of different \(\beta''\)-phase organic conductors and superconductors has been presented, utilizing dc and microwave methods at different frequencies (24 and 33.5 GHz) from room temperature down to 2 K. At ambient conditions the inplane dc conductivity is of the order 1 (\(\Omega\)cm\(^{-1}\)); the anisotropy \(\sigma_\parallel/\sigma_\perp\) exceeds 10\(^2\). Hence, the systems are highly anisotropic two-dimensional metals. All compounds exhibit a more or less metallic behavior upon cooling with \(\beta''\)-(BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) and the deuterated analog \(\beta''\)-(d\(_8\)-BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) becoming superconducting around 5 K. For \(\beta''\)-(BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) clear signatures of charge fluctuations are present for \(T < 30\) K. While they do not influence the dc properties, they lead to an increase of the high-frequency resis-
T emperature-dependent resisitivity of \( \beta'' \)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\). The resistivity increases to a maximum around 70 K, below which it decreases with a kink at 16 K.

**Fig. 8.** Some of the \( \beta'' \)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\) samples exhibit an insulating behavior in \( \rho(T) \). The resistivity increases to a maximum around 70 K, below which it decreases with a kink at 16 K.

**Fig. 9.** Temperature-dependent resistivity of \( \beta'' \)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\) measured at 24 and 33.5 GHz. The data are normalized to the respective room-temperature value.

activity. These effects are not seen in the deuterated analog, maybe due to the better metallic behavior.

Considering the frequency dependence of the conductivity as derived from our experiments, the general behavior certainly deviates from that of simple metals. We observe a more gradual temperature dependence for higher frequencies, indicating a significant frequency dependence of the electrodynamic response even in the microwave range. Since this happens at elevated temperatures (\( T > 100 \) K), we can rule out effects due to conventional phonon scattering. Instead we are inclined to trace it back to the peculiar bandstructure and correlation effects. The difference in frequency dependence is strongest in \( \beta'' \)-(BEDT-TTF)\(_2\)SF\(_5\)CHFSO\(_3\) and weakest in \( \beta'' \)-(BEDT-TTF)\(_2\)-SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\). Our investigations provide evidence that the vicinity to charge order and the presence of charge fluctuations strongly alter the dynamical response of the electronic system.

**Acknowledgements**

The dc measurements were done with the support of E. Rose. We would like to thank G. Untereiner for the careful sample preparation. The work at Stuttgart was supported by the Deutsche Forschungsgemeinschaft (DFG). The crystal growth at Argonne National Laboratory was performed under the auspices of the Office of Basic Energy Sciences, Division of Material Sciences of the U.S. Department of Energy, Contract No. DE-AC02-06CH11357.

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M. Gied et al.: High-frequency conductivity of the organic metals

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