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SUPERCONDUCTIVITY AT 10 K AND AMBIENT PRESSURE IN THE ORGANIC METAL $(BEDT-TTF)_2Cu(SCN)_2$

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We confirm the observation of superconductivity at ambient pressure above 10 K in the organic metal $(BEDT-TTF)_2Cu(SCN)_2$ as reported recently by Urayama et al [12]. In addition we have measured ESR, ac-susceptibility and thermopower in crystals of $(BEDT-TTF)_2Cu(SCN)_2$ and have shown that in contrast to other organic superconductors here a relatively sharp superconducting transition even in the ac-susceptibility can be observed which saturates already around 8 K. The thermopower measurements indicate a clear metal-metal phase transition at 100 K and a possible second phase transition at around 50 K, while from the temperature dependence of the resistivity and susceptibility (ESR) these phase transitions cannot be observed.

Since the observation of superconductivity at temperatures up to 100 K in the high T_c ceramic Cu-oxides - first discovered by Bednorz and Müller [1] - superconductivity has attracted again a lot of interest. Compared to the high T_c anorganic superconductors the transition temperatures into a superconducting state in organic materials is still rather low. Nevertheless, the development of new superconductors in this class of materials is quite fast. Since the first observation of superconductivity in an organic metal – the Bechgaard-salts – in 1980 with transition temperatures around 1 K [2, 3] quite a number of different organic radical salts showed superconductivity [4-11]. The highest observed transition temperature in a stable superconducting state at ambient pressure in α_t-$(BEDT-TTF)_2I_3$ was found at 8 K [9, 10]. Just recently Urayama et al [12, 13] reported for the first time the synthesis of a new organic superconductor $(BEDT-TTF)_2Cu(SCN)_2$ with a transition temperature above 10 K (T_c=10.4 K). The superconductivity was established by resistivity, dc-susceptibility and critical field measurements.

In this letter we confirm this observation of superconductivity above 10 K in $(BEDT-TTF)_2Cu(SCN)_2$ of Urayama et al [12], and in addition we report ESR, thermopower and ac-susceptibility measurements.

Crystals of $(BEDT-TTF)_2Cu(SCN)_2$ were grown by electrochemical methods similar as described in [12].

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In contrast to [12] the crystals were grown under constant voltage conditions with a current density of about 1 $\mu A/cm^2$. Distorted hexagon shaped crystals of a typical size of 1.5 x 0.5 x 0.05 mm^3 were obtained, and the largest crystals had the size of 3 x 1 x 0.05 mm^3.

Dc- and ac-conductivity measurements were performed with the usual four point method. Fig. 1a shows the temperature dependence of the resistivity of $(BEDT-TTF)_2Cu(SCN)_2$ crystals. The resistivity is normalized to the room temperature value ($\rho_{300} \approx 0.05\ \Omega cm$). Similar as in [12] the resistivity increases first until around 100 K, but in contrast to [12] the increase is weaker (only a factor of 2.5, and in two other crystals even only 1.5). Below 70 K the resistivity starts to decrease rapidly and the ratio $\rho_{100K}/\rho_{15K} \approx 500$. Fig 1b shows the low temperature region of the resistivity of our crystals of $(BEDT-TTF)_2Cu(SCN)_2$. The onset temperature for superconductivity lies at 11.1 K. The superconducting transition temperature as evaluated by the center of the resistive transition amounts to 10.2 K. At 7.8 K the superconducting transition - as measured by resistivity - is complete and the resistivity zero within the experimental possibilities.

Ac-susceptibility measurements at 3 MHz ($\approx$ 0.2 Gauss) were done as described earlier [14]. Fig. 2 shows the increase in resonance frequency of the LC-circuit due to exclusion of the rf-field by diamagnetic shielding currents in the $(BEDT-TTF)_2Cu(SCN)_2$ crystals. There is a clear evidence of an onset of diamagnetic shielding around 10 K, somewhat lower than the observed onset in resistivity [12]. A similar lower transition temperature in the ac-susceptibility was already observed before for β-$(BEDT-TTF)_2I_3$ [15] and α_t-$(BEDT-TTF)_2I_3$ [10] crystals. In contrast to β-$(BEDT-TTF)_2I_3$ and α_t-$(BEDT-TTF)_2I_3$ the superconducting transition here in $(BEDT-TTF)_2Cu\cdot(SCN)_2$ as observed in the ac-susceptibility is rather sharp and starts to saturate already at 8 K. Urayama et al [12] reported a diamagnetic signal measured by a Faraday susceptometer of about 5 % with respect to a perfect diamagnet. With the ac-susceptibility we

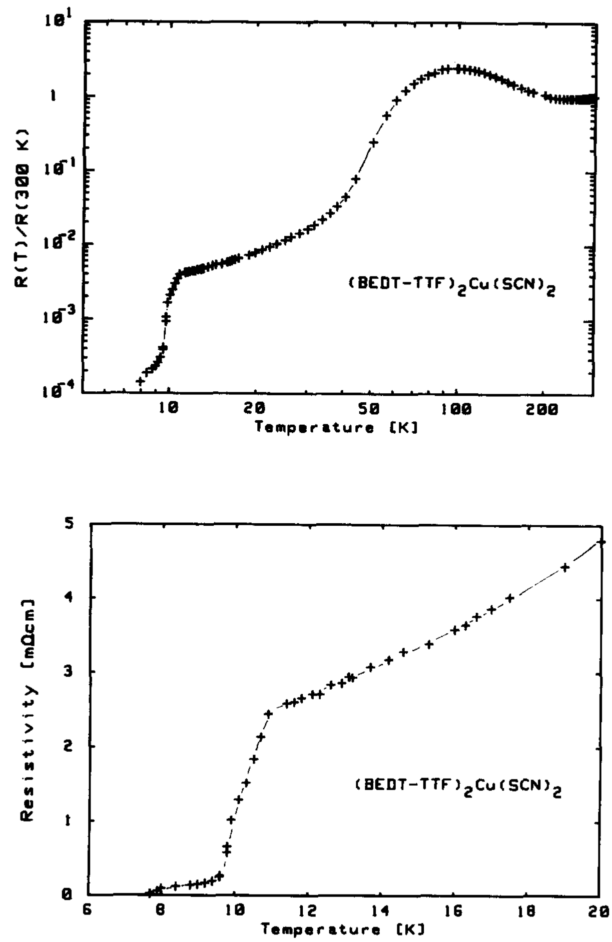

Fig. 1 a: Temperature dependence of the resistivity of $(BEDT-TTF)_2Cu(SCN)_2$ (resistivity normalized to the room temperature value). b: Low temperature region of the resistivity (linear scale) of $(BEDT-TTF)_2Cu\cdot(SCN)_2$.

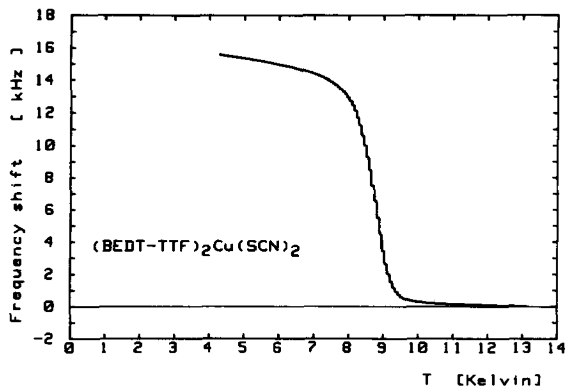

Fig. 2. Increase in resonance frequency of a LC-circuit due to exclusion of the rf-field by diamagnetic shielding currents in $(BEDT-TTF)_2Cu(SCN)_2$ (ac-susceptibility).

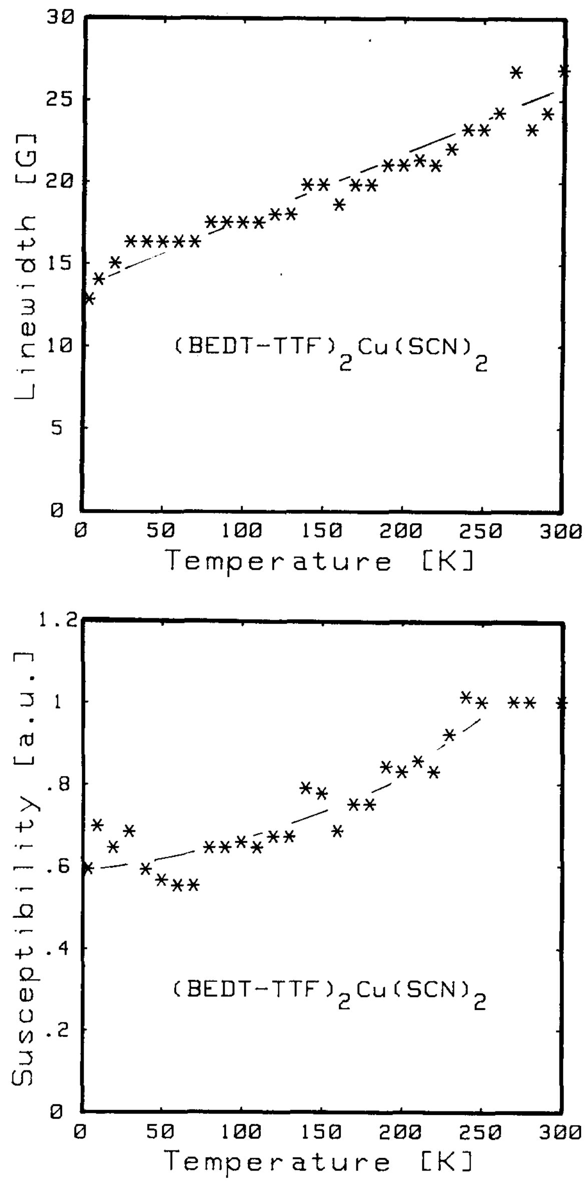

Fig. 3 a: Temperature dependence of the ESR-linewidth of $(BEDT-TTF)_2Cu(SCN)_2$ crystals. b: Temperature dependence of the susceptibility - as obtained by ESR - of $(BEDT-TTF)_2Cu(SCN)_2$.

find a much stronger effect and see a diamagnetic shielding signal for our crystals of at least 70 % with respect to a perfect superconductor indicating a strong volume effect (this strong effect can also be seen on a powdered sample).

Figs.3a,b show the temperature dependence of the ESR-linewidth as well as of the susceptibility. At room temperature the ESR-linewidth is about 26 Gauss while at 10 K a linewidth of only 14 Gauss is observed. The ESR -linewidth measurement can be used for the identification of the crystals [16]. The ESR-line is more or less symmetric over the whole temperature range, and only below 20 K the typical Dyson-type lines as for metals with high conductivity are observed. As expected for a metal the susceptibility obtained by ESR is only weakly temperature dependent and looks very similar to the susceptibility measured with the Faraday susceptometer [12].

Thermopower measurements were done on three crystals as described earlier [17]. All three crystals showed very similar results. An example is shown in Fig. 4. In contrast to the resistivity and susceptibility the thermopower measurement indicates clearly that between 100 and 110 K a phase transition exists. In addition at 50 K a second phase transition might occur. Since above 100 K and between 50 and 80 K the thermopower depends linearly on the temperature, the phase transition at 100 K seems to be a metal-metal phase transition. At room temperature the anions are not linear but bent and positional disorder was observed [12] while at 100 K the disorder does not exist [13]. So, it seems that the ordering of the anions below 100 K results in a new electronic state as can be seen from the thermopower measurement.

Fig. 5 shows the low temperature region (<50 K) of the thermopower of $(BEDT-TTF)_2Cu(SCN)_2$. Between 18 und 50 K the typical behaviour of a metal is observed while below 12 K the thermopower is zero within the experimental error as expected for a superconductor.

In conclusion we have shown that $(BEDT-TTF)_2Cu(SCN)_2$ is a volume superconductor at ambient pressure above 10 K with even a relative sharp transition observed in the penetration depth measurement (ac-susceptibility). Thermopower measurements indicate a metal-metal phase transition at 100 K and a possible second phase transition around 50 K.

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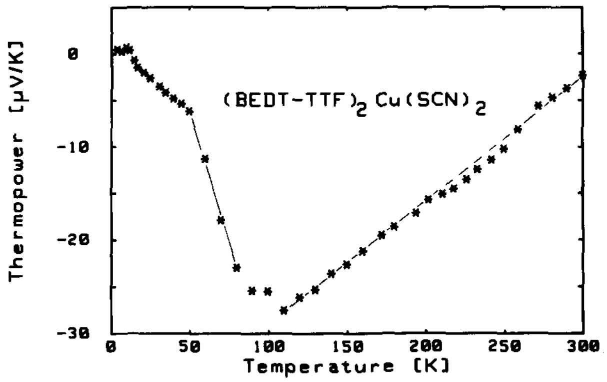

Fig. 4. Temperature dependence of the thermopower of $(BEDT-TTF)_2Cu(SCN)_2$.

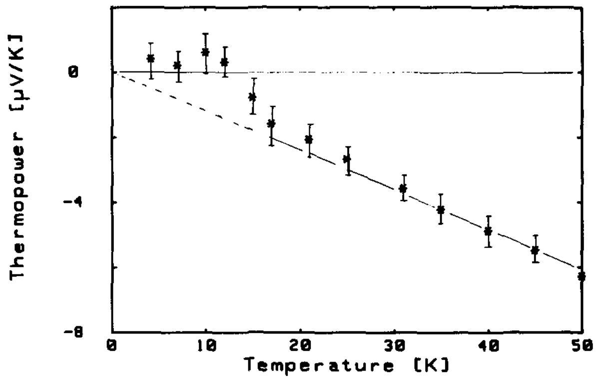

Fig. 5. Low temperature region of the thermopower of $(BEDT-TTF)_2Cu(SCN)_2$ (see text).

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