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# FREQUENCY-DEPENDENT CONDUCTIVITY IN PURE AND IODINE-DOPED α-(BEDT-TTF)$_2$I$_3$

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## ABSTRACT

Measurements of the microwave conductivity at 10 GHz show a plateau below the metal-insulator transition at 135 K in contrast to the further dropping dc conductivity. In pure material the plateau is independent of frequency (4 GHz, 10 GHz, and 22 GHz), but in the temperature range above 135 K strong frequency dependence is observed.

Iodine doping causes frequency dependence also in the plateau range. Particular doping rates give rise to several conductivity behaviors for different crystal directions. The results are discussed referring to CDW depinning and relaxation processes.

## INTRODUCTION

In contrast to β-(BEDT-TTF)$_2$I$_3$ salt which becomes superconducting at ambient pressure at $T_c$ = 1.03 ... 1.45 K [1-2] the α-phase is a two-dimensional metal with the metal-insulator phase transition at about 135 K [3]. An interesting behavior is the difference between microwave conductivity and dc conductivity at temperatures below 120 K. On cooling the dc conductivity continues to decrease approximately with an activited character (≈ 0.08 eV) after a rapid drop at 135 K while the microwave conductivity reaches a plateau keeping almost constant down to 30 K [3]. In this temperature range recently a metallic state followed by a transition to superconductivity at 3.1 ... 3.3 K was found after doping with iodine [4].

The present paper describes microwave measurements of the conductivity at different frequencies on pristine as well as iodine doped (with various iodine

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exposure times) crystals of $\alpha$-(BEDT-TTF)$_2$I$_3$ along the stack direction [100] and perpendicular to it, [010]. Cavity perturbation technique [5-6] was used at frequencies 4.6, 9.3, 10.3 and 23.5 GHz in the temperature range between 80 and 300 K (at 10.3 GHz the temperatures down to 3 K were available to measure). Iodine doping has been performed by holding the crystals in the saturated iodine vapour for specified exposure time. It was tried to keep one crystal throughout all measurements.

## RESULTS AND DISCUSSION

Fig. 1 shows the effect of iodine doping on the stack-direction conductivity $\sigma_a$ (along [100]) at 10.3 GHz. The values of pristine probes are consistent with published data [3]. There are no significant differences between the $\sigma_a$ (10.3 GHz) versus temperature behavior of pure crystals and those exposed to iodine no longer than 16 min. The ac conductivity has metallic character down to 135 K, then undergoes a shorp metal-insulator phase transition. For temperatures be-

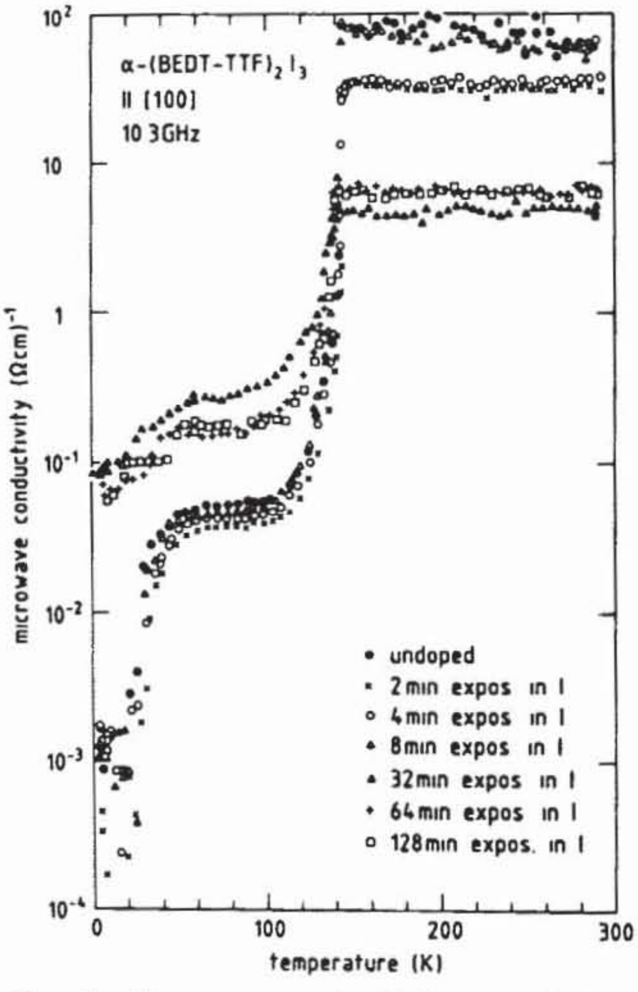

Fig. 1. Microwave conductivity $\sigma_a$ along stack direction [100] at 10.3 GHz versus temperature for various exposure times in iodine (first 5 items sample no. 5, last 2 items sample no. 7).

tween 120 and 40 K, $\sigma_a$ reaches a plateau lying 3 orders of magnitude lower then the metallic conductivity region. Below 40 K the conductivity falls down suggesting another phase transition. Exposing to iodine more than 30 min lowers the conductivity in the metallic range and raises the conductivity in the plateau region. The absolute values of $\sigma_a$ (10.3 GHz) in the plateau range (40 K < T < 120 K) are about 4-5 times larger than those of undoped probes. There is no rapid drop of the conductivity below 30 K. A metallic or even a superconducting state below 120 K is not observed. Further iodine doping (> 2 h) has no influence on the $\sigma$ versus T behavior. The effect of iodine doping on the dielectric constant is neglectable.

In a second stage the conductivity was measured on one crystal as function of temperature and frequency for a fixed exposure time in iodine. Fig. 2 shows the results of the pristine crystal, while Figs. 3-4 represent the behavior of the same samples after 1 h iodine doping.

In the metallic range (T > 135 K) a significant frequency dependence is found as well in the pure (Fig. 2) as in the doped sample (Figs. 3-4). Iodine doping causes decrease of the conductivity $\sigma_a$ along the stack, especially at low frequencies. On the other hand the conductivity $\sigma_b$ along [010] is increased. That means iodine doping makes the initial two-dimensional conductivity more one-dimensional (anisotropy factor $\sigma_b/\sigma_a \approx 3.8$).

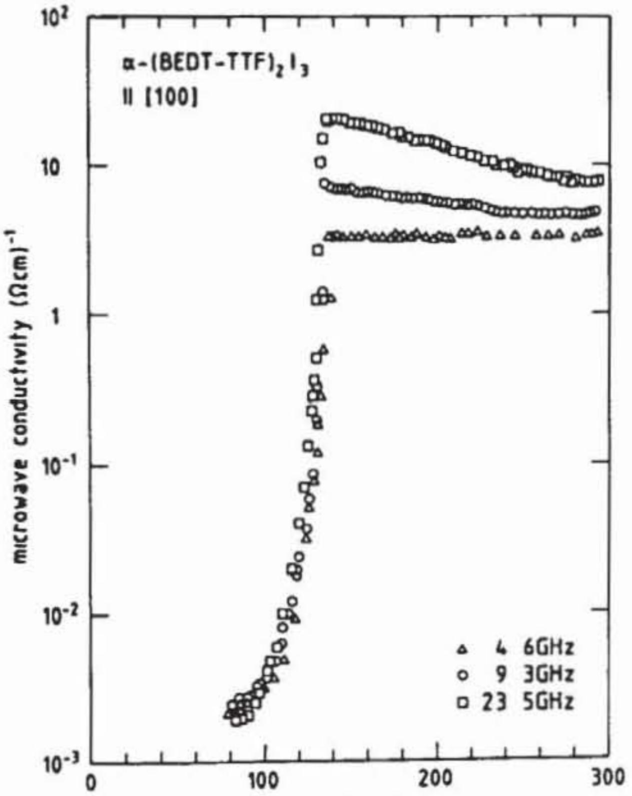

Fig. 2. Microwave conductivity $\sigma_a$ along stack direction [100] versus temperature at different frequencies for a pristine probe (sample no. 10).

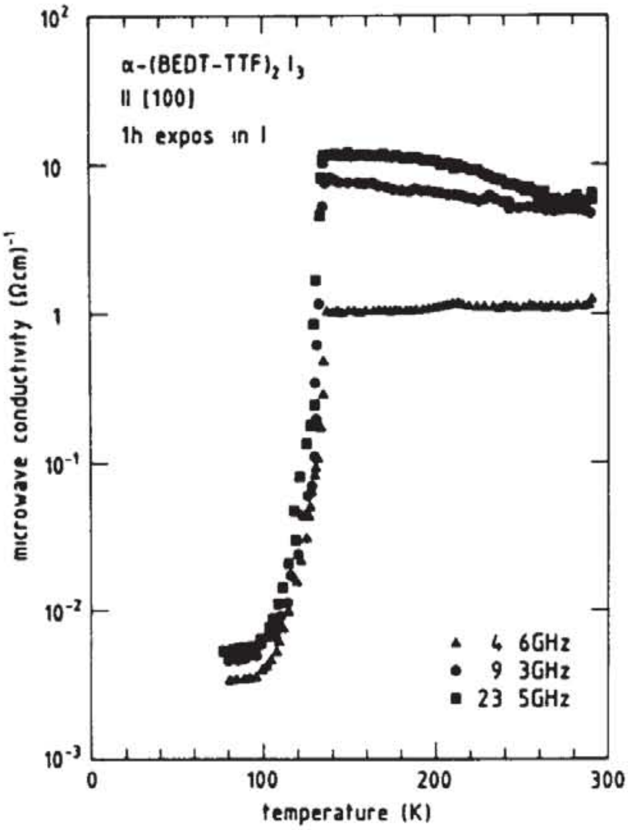

Fig. 3. Microwave conductivity $\sigma_a$ along stack direction [100] versus temperature at different frequencies after 1 h iodine doping (sample no. 10).

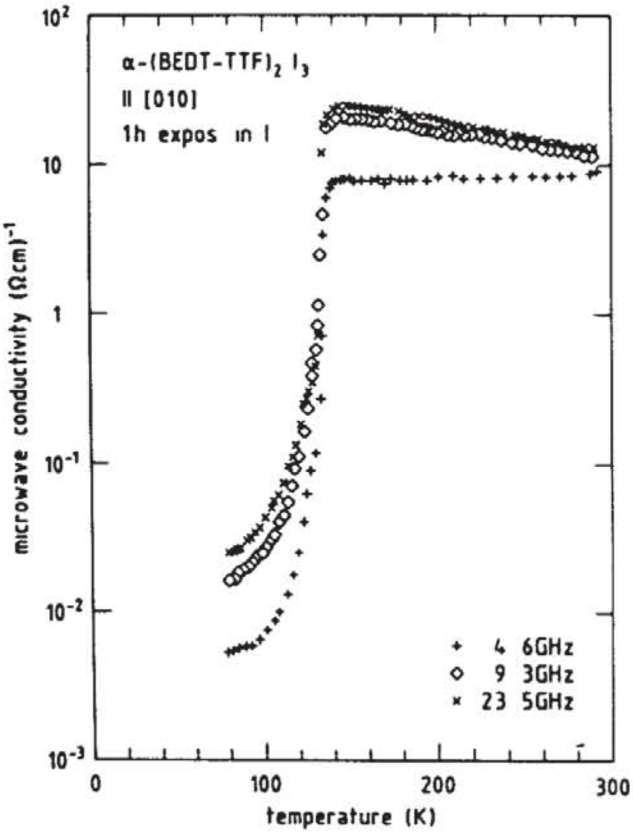

Fig. 4. Microwave conductivity $\sigma_b$ perpendicular (≈ [010]) to the stack direction versus temperature at different frequencies after 1 h iodine doping (sample no. 10).

In the plateau range (T < 100 K) there is practically no frequency dependence in the pure crystal (Fig. 2). Iodine doping leads to somewhat higher conductivity $\sigma_a$ and little frequency dependence (Fig. 3). But the $\vec{b}$-axis conductivity $\sigma_b$ increases significantly showing also an important frequency dependence (Fig. 4). The conductivity increases the higher the frequency.

There are arguments to assume that the frequency dependence of the conductivity in the metallic state (T > 140 K) is due to the quality of the crystals. Grain boundaries and microscopic cracks lead to a relaxation process. Fig. 5 shows conductivity and dielectric constant $\varepsilon'$ versus frequency calculated for a simple relaxation model with a reasonable set of data. The iodine doping may be interpreted by diffusion of iodine into the grain boundaries. Then the broadening of the boundaries causes an increase of the relaxation frequency.

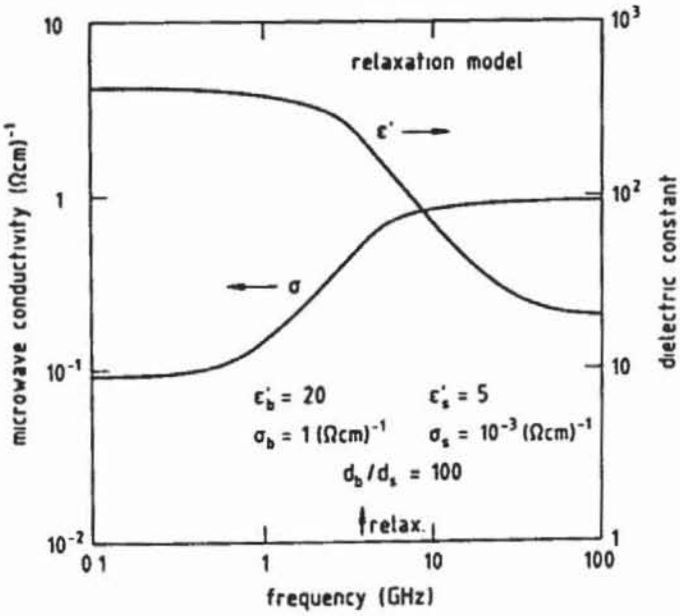

Fig. 5. Microwave conductivity and dielectric constant $\varepsilon'$ calculated from a relaxation model. Assumed $\sigma$, $\varepsilon'$ and thickness d values for the bulk (b) grains and the boundaries (s) are inserted.

The frequency dependence of the conductivity below the phase transition (T < 130 K) might be accounted for by collective modes represented by a periodic modulation of charge i.e. charge density waves (CDW), which can be pinned by impurities or disorder. The pinning energy estimated for $\alpha$-(BEDT-TTF)$_2$J$_3$ would be of the order of $10^{-2}$ eV, characteristic for strong pinning forces.

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