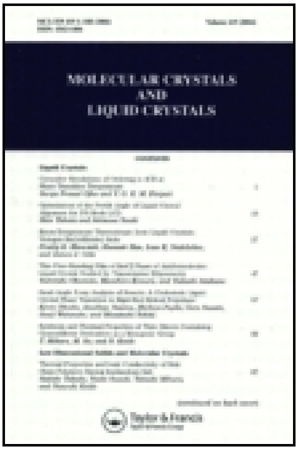

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# Explanation for the Anomalous IR Dielectric Constant of TTF-TCNQ

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# EXPLANATION FOR THE ANOMALOUS IR DIELECTRIC CONSTANT OF TTF-TCNQ

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The recent data of Tanner et al indicates an anomalous behavior of the dielectric constant $\varepsilon_1$ of TTF-TCNQ. The large and *negative* value of $\varepsilon_1$ found experimentally is accounted here by applying the phonon-drag theory. The positive value of $\varepsilon_1$ found for damaged samples is accounted by a broken-chain model.

Recent data by Tanner et al (1) raised again the question whether *collective* effects (2,3,4) or single particle theory (5,6,7) can account for transport phenomena in quasi-one-dimensional organic metals.

Tanner et al (1) obtained the ac conductivity $\sigma(\omega)$ and the dielectric constant $\varepsilon_1$ in the far infrared region ($\omega<400\ cm^{-1}$), for $25\ K < T < 160\ K$ . The main features of their results are: (i) Rapid decrease of the conductivity with increasing frequency at $\omega \simeq 20\ cm^{-1}$ (ii) Large (in absolute value) and *negative* dielectric constant ($\varepsilon_1 \simeq -10^4$ ).

These experimental results are in sharp contradiction with previous results of Gunning et al (8) who found a *positive* dielectric constant $\varepsilon_1 \simeq +2500$. This positive dielectric constant invoked the collective approach which yields a positive $\varepsilon_1$ . Another possible explanation for the positive dielectric constant was given by using a broken chain (9,10) model. This model applies to low-quality samples with damaged surfaces.

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In order to test which explanation is correct, one needs to measure $\varepsilon_1$ for very high quality samples. Indeed, the measurements by Jacobsen (11) and Tanner et al (1) for very high quality samples indicate a negative dielectric constant. Thus, the positive (8) $\varepsilon_1$ may be attributed to damaged areas near the surface which yield a large positive $\varepsilon_1$.

While the negative *sign* of $\varepsilon_1$ is properly accounted for by the Boltzmann-Drude theory, the *magnitude* is not. $(\varepsilon_1)_{Drude} \simeq -200$ at $T = 100$ K which is about *two orders of magnitude* smaller than the experimental value.

We apply here the phonon-drag theory (12) and get

$$\sigma(\omega) = \sigma_{dc}\left(1 + i\omega\tau_{dc} + \frac{\tau_{dc}}{\tau_{e\ell\to ph}}\ \frac{i\omega\,\tau_{ph\to e\ell}}{1+i\omega\tau_{ph\to e\ell}}\right)^{-1}$$

$$\varepsilon_1(\omega) = -4\pi\,\sigma_{dc}\,\tau_{dc}\ \frac{1 + \tau_{ph\to e\ell}/\tau_{e\ell\to ph} + \omega\tau_{ph\to e\ell}}{[1-\omega^2\tau_{dc}\tau_{ph\to e\ell}] + \omega^2[\tau_{ph\to e\ell} + \tau_{dc}(1+X)]^2}$$

where $X = \tau_{ph\to e\ell}/\tau_{e\ell\to ph}$ and $\tau_{e\ell\to ph}$ , $\tau_{ph\to e\ell}$ are the relaxation times for the electron (due to emission or absorption of a $2k_F$ phonon) and the $2k_F$ phonon (due to absorption or emission by an electron). At $\omega \to 0$ this yields $\varepsilon_1 \simeq -10^4$ in good agreement with experiment. $\tau_{dc}$ is the relaxation time of the electron due to quadratic coupling (the 2-phonon mechanism) which dominates in TTF-TCNQ (7).

The sharp difference of $\varepsilon_1$ between different samples supports the idea of the existence of broken chains for which we get:

$$\sigma(\omega) = \frac{\sigma_{bulk}}{1+(\omega/\omega_c)^2}\ \left((\omega/\omega_c)^2 + i(\omega/\omega_c)\right)$$

where $\omega_c = 4\pi d\sigma_{bulk}/\varepsilon_o\ell$ and $d$ is the width of the break, $\varepsilon_o$ the effective dielectric constant of the broken section and $\ell$ is the length of the chain. Thus, the large positive $\varepsilon_1$ is accounted for by this model.

Describing the bulk by the phonon-drag theory and the surface by a broken strand model with an effective medium calculation (taking into account also the transverse conductivity) we obtain good agreement with experimental results (Figs. 1, 2).

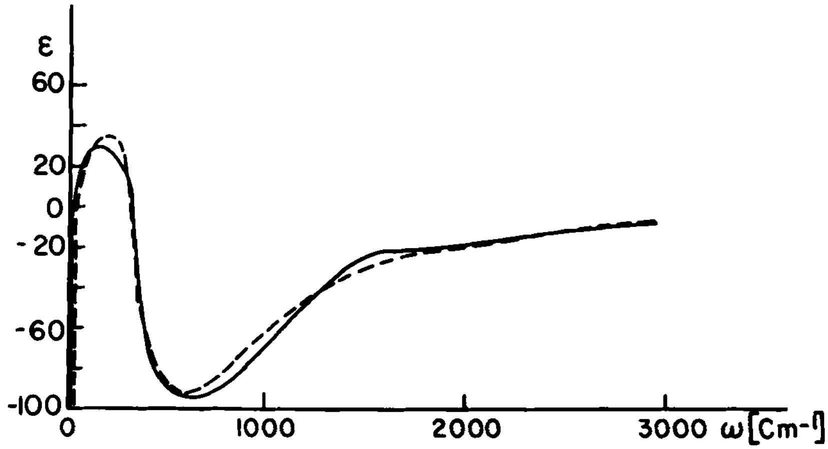


FIGURE 1 Dielectric constant $\varepsilon_1(\omega)$ at T=100 K. Full line-experiment (ref. 11). Dashed line-present theory.

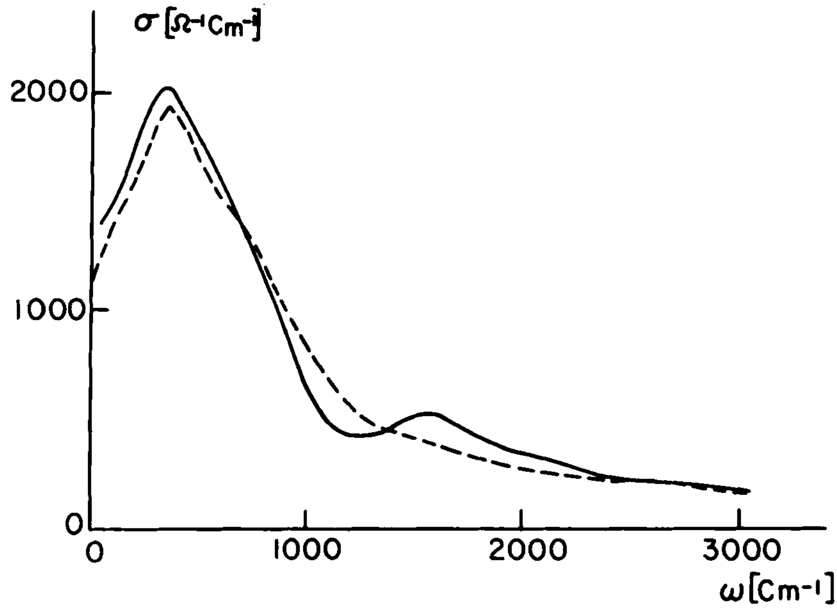


FIGURE 2 Conductivity $\sigma(\omega)$ at T=100 K. Full line - experiment (ref. 11). Dashed line - present theory.

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