Frequency Dependent Conductivity of the Quasi-One-Dimensional Organic Charge-Density-Wave Conductor (Fluoranthene)$_2$PF$_6$

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Abstract

Frequency dependent conductivity measurements on the organic charge-density-wave (CDW) conductor (fluoranthene)$_2$PF$_6$ are reported in the frequency range from 10Hz to 3GHz for temperatures between 300 and 40K. We identify two processes associated with the CDW ground state of the system: (1) the dynamics of internal deformations of the collective mode, appearing at radio frequencies, together with (2) the onset of the pinned mode, which can be estimated to be of the order of 100GHz.

1. INTRODUCTION

The electrodynamics of the CDW ground state have been studied intensely in recent years by frequency dependent conductivity measurements on inorganic compounds like $K_{0.30}MoO_3$, $(TaSe_4)_2I$ and $NbSe_3$ [1,2]. In contrast to superconductors, where practically all relevant excitations are above the single particle excitation gap, in CDW condensates several features are observed for frequencies far below the single particle energies [3]. The most important resonance at the so-called pinning frequency (usually in the microwave to millimetre-wave range) is due to interactions of the CDW with the lattice and impurities, which shifts the collective mode excitation from zero to finite frequencies. In the radio frequency range internal deformations of the condensate lead to a low frequency contribution to $\sigma(\omega)$, which strongly depends on the stiffness of the CDW and on screening effects caused by uncondensed electrons. The optical response in the FIR range due to phonons is also influenced by the CDW formation through electron-phonon coupling.

In this paper we give a short overview of our frequency dependent experiments on the organic CDW conductor (fluoranthene)$_2$PF$_6$ and compare our results to inorganic systems. (Fa)$_2$PF$_6$ is a radical cation salt based on the organic donor molecule fluoranthene (Fa), which is a pure aromatic hydrocarbon ($C_{16}H_{10}$). Due to the chainlike crystal structure the anisotropy of the conductivity is very high ($\sigma_{\parallel}/\sigma_{\perp}\cong 10^4$) and the material undergoes a metal-insulator transition, a Peierls transition, at 182K to a CDW ground state [4]. Direct evidence for a Peierls transition came from x-ray investigations, where diffuse lines above and sharp spots below the transition temperature gave clear evidence for the existence of a $2k_F$ superlattice with a wavevector $\mathbf{q}=(\frac{1}{2},0,0)$ due to a CDW [5]. Like in inorganic compounds in the CDW state of (Fa)$_2$PF$_6$ several collective transport phenomena have been observed [6]. These are nonlinear conductivity above a small sample dependent threshold field of 0.1-1V/cm, conduction noise in the nonlinear state and the decay of metastable CDW configurations.

These investigations indicate that - in contrast to most inorganic CDW conductors - apart from defect pinning also commensurability pinning of the CDW with the underlying lattice plays a major role in the CDW dynamics of the system [6].

2. EXPERIMENT

The frequency dependent conductivity $\sigma(\omega)$ was measured over a broad spectral range from 10Hz to 3GHz between 40K and 300K by combining three different experimental techniques. The range from 10Hz to 10MHz was covered by a frequency response analyzer (Schlumberger SI1260) and an impedance analyzer (Hewlett Packard 4194A). In the high frequency range from 10MHz to 3GHz the sample was mounted at the end of a 50Ω wave guide of a network analyzer (Hewlett Packard 8753C). The temperature range was limited by the resolution ($\tan\delta\cong 10^{-3}$) and the input resistances of the analyzers.

The experiments were performed in a two-probe configuration on single crystals with lengths of 0.2-1mm and cross sections of typically 1x1mm^2. Only single crystals from the same batches were used for the different frequency ranges, so that the agreement of results gained by different techniques was satisfactory. The oscillator amplitudes applied in various methods were always small to avoid nonlinear effects.

3. RESULTS AND DISCUSSION

The electrical conductivity parallel to the needle axis, measured at several frequencies between 10Hz and 3GHz, is displayed in fig. 1. Though the room temperature conductivity obtained by the two-probe configuration is more than one order of magnitude lower than the values received by the four-probe method (σ(300K)=10^2-10^3(Ωcm)$^{-1}$), the signature of the Peierls transition is clearly visible for all frequencies (see ↙ in fig. 1). Below this temperature, when the sample resistance is rapidly growing with decreasing temperature, the influence of contacts can be neglected and the response originates from the specimen. Below about 150K the curves for different frequencies begin to separate and the conductivity at high frequencies soon leads into a plateau, whereas the data at low frequencies follow the dc conductivity down to low temperatures. Consequently, at 40K the difference in conductivity between 3GHz and 10Hz is about seven orders of magnitude.

In fig. 2 the conductivity is plotted as a function of the frequency for different temperatures. At room temperature there is practically no ac contribution in the whole frequency range, as is expected for a one-dimensional metal. Below T_P a frequency dependent contribution develops, whose onset is shifted towards lower frequencies with decreasing temperature. We identify two different contributions: (1) a temperature dependent process at low frequencies (indicated by ↘ in fig. 2) and (2) a temperature independent resonance with a much larger slope at high frequencies (↑ in fig. 2). The low frequency mode can be attributed to internal deformations of the CDW. It is strongly temperature dependent because of screening effects by uncondensed electrons. This contribution will be discussed in more detail in a forthcoming paper [7].

The process at GHz frequencies can be identified as the onset of the collective CDW mode at the pinning frequency ω_p. This resonance is due to the oscillatory response of the whole CDW in the periodic pinning potential.

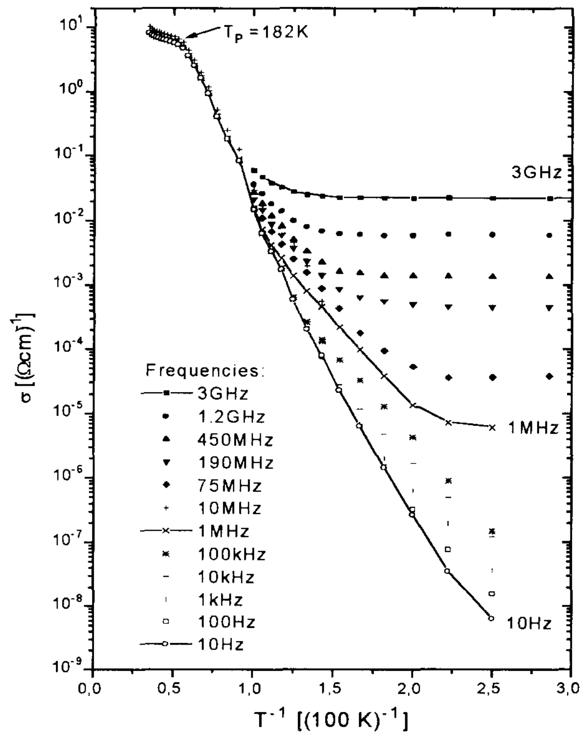

Figure 1: Temperature dependence of the electrical conductivity of a $(Fa)_2PF_6$ single crystal for different frequencies. Data from 10Hz to 1MHz were measured with the SI1260 and from 10MHz to 3GHz with the HP8753C.

According to the model of Grüner et al. [8] the pinning frequency can be estimated from:

$$\omega_p^2 = \frac{2\pi}{\lambda_p}\cdot\frac{eE_T}{M^*} \qquad (1)$$

where λ_p is the wavelength of the pinning potential, E_T the electrical threshold field for nonlinear conductivity and M^* the effective mass of the CDW condensate. M^* is given by:

$$M^* = m^*\left(1+\frac{1}{\lambda}\left(\frac{2\Delta}{\hbar\omega_0}\right)^2\right) \qquad (2)$$

with m^* the effective single particle band mass, 2Δ the single particle energy gap, ω_0 the $2k_F$ phonon frequency of the metallic system and λ the dimensionless electron-phonon coupling constant. With $m^*=m_0$, Δ=90meV, λ=0.3 and $h\omega_0$=2meV [9] one yields $M^*\cong 2.7\cdot 10^4 m_0$. With the value of E_T=1V/cm, derived from nonlinear experiments, and λ_p=2a=6.6Å follows:

$$\omega_p/2\pi = 0.3\text{GHz}.$$

The high frequency results, together with earlier data from microwave conductivity measurements at 10.2GHz [10], suggest that the pinning frequency is above 10GHz. From the extrapolation in fig. 2 the value of ω_p can be estimated to be of the order of 10^{11}Hz. Thus the above calculation of the pinning frequency of 0.3GHz is certainly two orders of magnitude too low. An explanation might be that the threshold field for nonlinear conductivity in a commensurate system should be much higher than 1V/cm. So the non-linear conductivity observed in our experiments may be due to local CDW discommensurations or defects [6]. This is confirmed primarily by the fact that there is no narrow band noise in this system. And secondly, the increase in conductivity for electrical fields as large as $10E_T$ is always less than a factor of 10, whereas the high frequency conductivity is several orders of magnitude higher than the dc values.

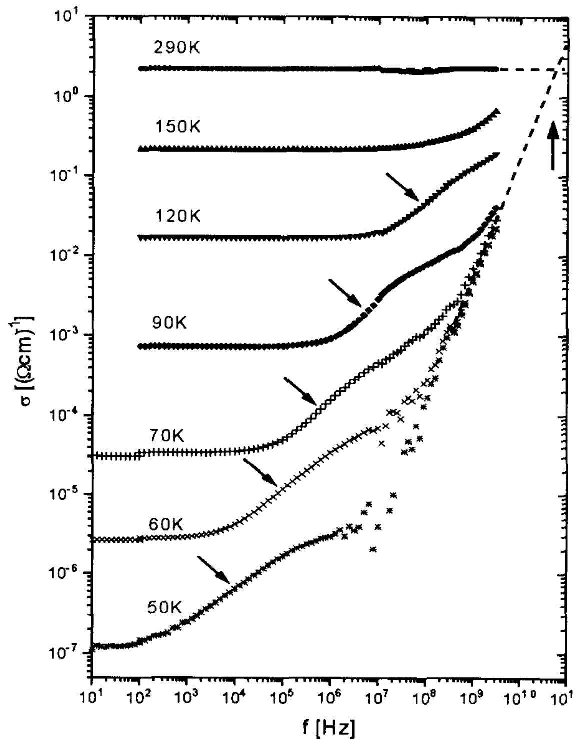

Figure 2: Frequency dependence of the electrical conductivity of another $(Fa)_2PF_6$ crystal between room temperature and 50K. Data from 10Hz to 100Hz were measured with the SI1260, from 100Hz to 10MHz with the HP4194A and from 10MHz to 3GHz with the HP8753C.

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