# Low-Frequency Behavior of Pinned Charge-Density-Wave Condensates

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Low-frequency conductivity measurements (100 Hz to 100 MHz) are reported in the charge-density-wave state of orthorhombic $TaS_3$. The observed frequency-dependent response is in clear disagreement with recent predictions which are based on the Fukuyama-Lee model. The results can be described by $\sigma(\omega) = A(i\omega)^\alpha$, with $\alpha < 1$, in a wide frequency range, suggesting relaxational dynamics associated with the collective mode.

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The electron-hole condensate called the charge density wave (CDW) does not contribute to dc electrical conductivity at low electric fields because interactions between the collective mode and impurities lead to pinning of the phase $\phi$ of the condensate. However, the interaction can be weak, for small impurity concentration, leading to strong frequency $\omega$ and electric field $E$ dependent response,[1] as observed recently in a broad range of inorganic chain compounds with a CDW state. Phenomenological models[2,3] have been proposed which account in a general way for a broad variety of observations in these materials and which make explicit predictions for the frequency dependence of the complex conductivity, $\sigma = \sigma' + i\sigma''$. While one of these models is classical and the other quantum mechanical, both predict in the low-frequency limit

$$\sigma' \propto \omega^2, \quad \sigma'' \propto \omega, \tag{1}$$

analogous to the response of a single harmonic oscillator. Previous experiments,[1,4,5] conducted on various CDW solids between 1 MHz and 10 GHz, found broad qualitative agreement with the phenomenological models, although with clear deviations in some details. The phenomenological models treat the CDW as a single rigid entity, neglecting internal deformations of the CDW. Such deformations are described by a microscopic model,[6,7] and the frequency-dependent response of the system has been examined in detail (see below). The predictions based both on the phenomenological and on the microscopic models motivated us to extend our measurements to lower frequencies, where we find power-law behavior

$$\sigma(\omega) = A(i\omega)^\alpha, \tag{2}$$

with $\alpha < 1$ over a very broad frequency range. This result is in clear disagreement with both the phenomenological and the internal-deformation models, suggesting instead slow relaxation phenomena, and "glassy" behavior in general, which is not included in the theories.

The experiments were performed on orthorhombic $TaS_3$, a widely investigated model compound for CDW transport phenomena. Samples were prepared by standard temperature-gradient growth method from high-purity starting materials. Both the sharpness of the transition and the observed threshold field ($E_T \approx 200$ mV/cm at 120 K) indicate a small impurity concentration. In order to cover a wide frequency range, different techniques with overlapping bandwidths were applied. A carefully designed ac bridge circuit was used in the 0.5–100-MHz frequency region. Between 5 and 500 kHz, a HP 4800A vector impedance meter was used. Below 50 kHz, the conductivity $\sigma_{dc}$ due to single-particle excitations across the gap becomes dominant. To subtract this contribution we applied a dc bridge circuit in the frequency range of 100 Hz–50 kHz. At low frequencies $[\mathrm{Re}\sigma(\omega) - \sigma_{dc}]/\sigma_{dc}$ as small as $10^{-5}$ was measured by this method. Although we have found that contact resistances were two orders of magnitude smaller than the sample resistance, they might influence the ac response at such low frequencies. Four-probe bridge experiments were performed to exclude this possibility. To reduce the effect of spurious capacitances associated with long leads, a $^4$He gas flow system was used.

During the experiments the peak-to-peak voltage drop on the sample $V_{ac}$ was kept below $V_T/10$, where $V_T$ is the threshold field of the nonlinear conduction. We have investigated the voltage dependence of $\sigma(\omega)$ and found no significant changes even if $V_{ac}$ was comparable to (but less than) $V_T$. (In the case of two-probe measurements, the second harmonic of the signal appears at these higher voltages, probably due to rectification effects at the contacts.) Our results proved also to be insensitive to a dc bias field, if the applied dc voltage was below threshold. Experiments were performed at several temperatures well below the Peierls transition temperature $T_P = 220$ K. Previous conductivity[4] and also broad-band and narrow-band noise[8] experiments suggest that at these temperatures disorder is important. The highly coherent response, observed in the current carrying state near to the Peierls transition (as evidenced by the narrow-band noise),[6] is replaced at these temperatures with a distribution of narrow-band-noise components, strongly suggesting incoherent effects.

Figure 1 shows $\mathrm{Re}\sigma(\omega)-\mathrm{Re}\sigma(\omega\rightarrow 0)$ and $\mathrm{Im}\sigma(\omega)$ at $T=120$ K. Similar results were obtained at different temperatures below $T_{\mathrm{P}}$. At low frequencies both $\mathrm{Re}\sigma(\omega)$ and $\mathrm{Im}(\omega)$ approach a power law and between 100 Hz and 10 MHz both can be well described by Eq. (2) with $\alpha<1$. $\alpha$ appears to be independent of the temperature, while the magnitude of the frequency-dependent response displays a rather weak (as yet unexplained) temperature dependence.

These results conflict conspicuously with the phenomenological models, Eq. (1). While such models *may* be appropriate when the dimensions of the specimen are comparable to the intrinsic length scale of phase correlations (determined by the interplay of random pinning with the elastic stiffness of the condensate), in general the internal dynamics must play a crucial role. The commonly used microscopic model which describes the impurity pinning of the condensate is that of Fukuyama and Lee[8] and of Gorkov.[9] The Hamiltonian in one dimension is

$$H=\int\left[\frac{v_{\mathrm{F}}}{4\pi}(\nabla\phi)^2 - V\sum_k\delta(x-x_k)\cos(\phi+2k_{\mathrm{F}}x)\right]dx, \quad (3)$$

where $v_{\mathrm{F}}$ and $k_{\mathrm{F}}$ are the Fermi velocity and wave vector, and the impurity potential is assumed to be of short range, $V(x)=V\delta(x-x_0)$. The first term is elastic energy of the condensate, and the second term represents the interaction of the CDW with the randomly positioned impurities. Eq. (3), when supplemented with a coupling to the applied electric field, represents a complicated nonlinear problem, with reliable solutions obtained only in certain limits of the parameters involved. One such limit is the low-frequency ($\omega\rightarrow 0$) response of the condensate for small applied ac fields ($V_{\mathrm{ac}}\rightarrow 0$).

While the effective medium approximation to Eq. (3) leads to the real and imaginary parts of the conductivity $\mathrm{Re}\sigma(\omega)\sim\omega^2$ and $\mathrm{Im}\sigma(\omega)\sim\omega$ in the $\omega\rightarrow 0$ limit, a careful examination of the energy dependence of the density of states gives the result[9,10]

$$\mathrm{Re}\sigma(\omega)\sim\left(\frac{e^2u^2}{v_{\mathrm{F}}}\right)\omega_0^{-1}\left(\frac{\omega}{\omega_0}\right)^2\ln^2\left(\frac{\omega_0}{\omega}\right), \quad (4)$$

which corresponds to the Mott-Berezinski[11] result for one-dimensional localization. Here $u=v_{\mathrm{F}}(m/m_{\mathrm{F}})^{1/2}$ is the phason velocity with $m$ and $m_{\mathrm{F}}$ the band mass and CDW mass. The characteristic frequency $\omega_0$ is related to the pinning energy $\epsilon_P$ and is given by

$$\omega_0=(u/v_{\mathrm{F}})(cv_{\mathrm{F}}V^2)^{1/3}, \quad (5)$$

where $c$ is the impurity concentration. Equation (4) is appropriate if $\omega_1<\omega<\omega_0$ with $\omega_1=Vu/v_{\mathrm{F}}$. Below this frequency the conductivity depends exponentially on the frequency, and[9]

$$\mathrm{Re}\sigma(\omega)\sim(\omega_0/\omega)^2\exp(-\omega_0/\omega). \quad (6)$$

$\omega_0$ and $\omega_1$ can be estimated using the parameters which enter into Eq. (3). With the phason velocity $u\sim 10^7$ cm/sec, $v_{\mathrm{F}}\sim 10^8$ cm/sec, impurity potential $V\sim 10^{-3}$ eV, and $c=100$ ppm, one determines that both $\omega_0$ and $\omega_1$ are of the order of $10^{10}$ $\mathrm{sec}^{-1}$. We note that with these parameters the characteristics Lee-Rice-Fukuyama[6] length $L=(v_{\mathrm{F}}^2/cV^2)^{1/3}$ $\sim 10$ $\mu$m and the ratio between the pinning and elastic energy $\epsilon=V/cv_{\mathrm{F}}\sim 0.3$, corresponding to weak impurity pinning. Accordingly Eq. (4) is expected to have a very limited range of validity, and

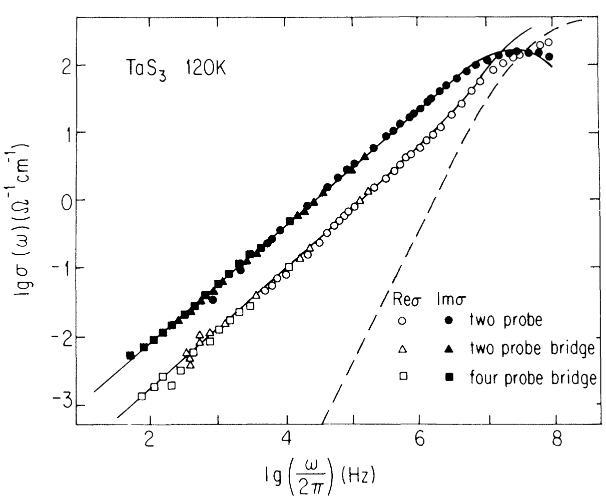


FIG. 1. $\mathrm{Re}\sigma(\omega)$ and $\mathrm{Im}\sigma(\omega)$ vs frequency at $T=120$ K. The dc conductivity has been subtracted from $\mathrm{Re}\sigma$. The full lines represent the Cole-Cole-like expression, Eq. (7), with $\alpha=0.87$ and $\tau^{-1}/2\pi=25$ MHz. The dashed line gives the low-frequency limit of the harmonic oscillator response, $\mathrm{Re}\sigma\propto\omega^2$.

$\mathrm{Re}\sigma(\omega)$ is expected to obey Eq. (6) in the $\omega \to 0$ limit.

In Fig. 2, the experimental behavior is contrasted with the predictions based on Eq. (3). The Mott-Berezinski law predicts that $\omega^{-1}[\mathrm{Re}\sigma(\omega)]^{1/2}$ versus $\ln\omega$ is linear, while if Eq. (6) is appropriate, $\ln[\omega^2 \mathrm{Re}\sigma(\omega)]$ is a linear function of $\omega^{-1}$. It is obvious from Fig. 2 that neither of these behaviors has been observed.

On the other hand, the low-frequency data are described excellently by Eq. (2), which emphasizes that the *same* power law $\omega^\alpha$ is found for both the real and imaginary parts. To account for the high-frequency behavior, the Cole-Cole-like expression

$$\sigma(\omega) = \sigma_\infty \frac{(i\omega\tau)^\alpha}{1+(i\omega\tau)^\alpha} \tag{7}$$

can be used, which reduces to Eq. (2) in the $\omega \to 0$ limit. The full lines of Fig. 1 are fits by Eq. (7) (parameters given in the figure caption). We stress, however, that Eq. (7) is regarded only as an empirical expression which reduces to the observed low-frequency limit, and which leads to saturation of $\mathrm{Re}\sigma(\omega)$ at high frequencies. Also, Eq. (7) would predict a divergent dielectric constant as $\omega \to 0$. It is possible that the expression breaks down below a cutoff frequency which appears to be below the lowest frequencies where the experiments were performed. Other descriptions of the high-frequency behavior have been discussed in Refs. 4 and 5.

Low-frequency conductivities similar to those reported here have been observed (although in a much more restricted frequency range) in a wide

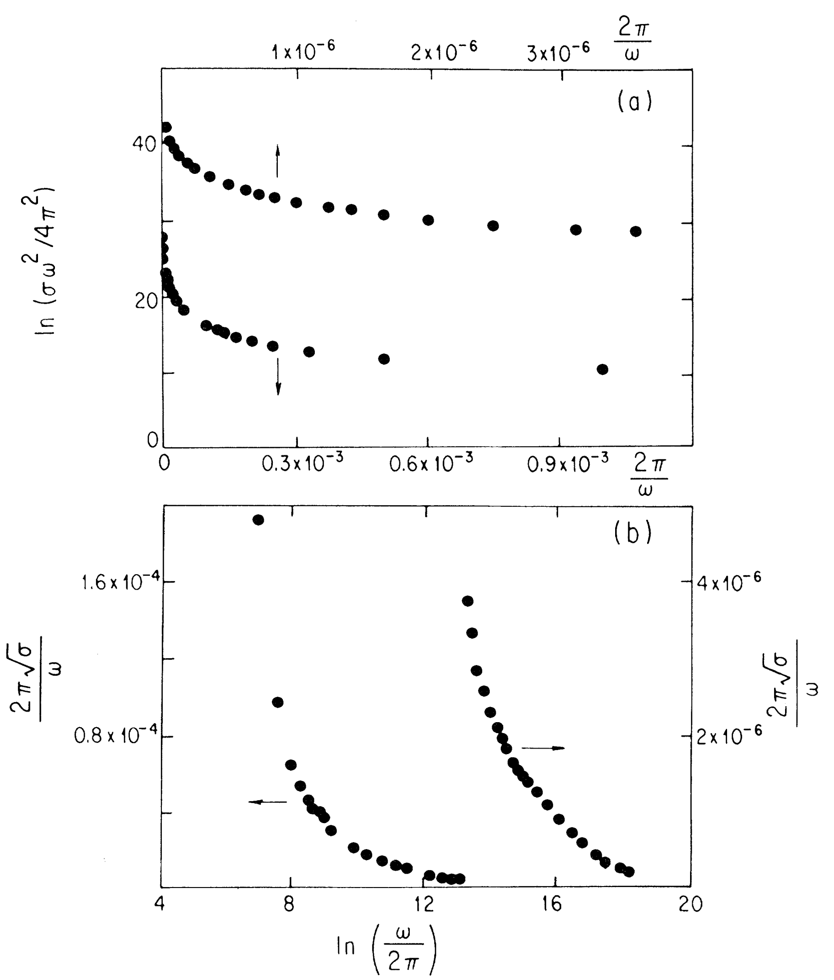


FIG. 2. Frequency dependence of (a) $\omega^2\mathrm{Re}\sigma$ and (b) $\omega^{-1}(\mathrm{Re}\sigma)^{1/2}$. If Eqs. (6) or (4) were correct, the experimental points would fall on a straight line in (a) or (b), respectively. Two scales are used to demonstrate that a straight line is not found over any range of frequency.

range of solids where disorder plays an important role.[12] Power-law behaviors are traditionally accounted for with a broad distribution of relaxation times, associated with transitions between states close in energy but nearly orthogonal in configurational space. Such arguments have recently been advanced to discuss long-time behaviors in spin glasses.[13] While such general arguments can also be applied to describe pinned CDW systems, where the inherent randomness is associated with the positions of the impurities, recent calculations which treat the dynamics of random one-dimensional models have a closer relation to our findings. Monte Carlo simulations of the dynamics of randomly pinned CDW's by Littlewood and Varma[14] lead to $\sigma(\omega)=A(i\omega)^\alpha$ with $\alpha=\frac{1}{2}$. Computer simulations based on the Frenkel-Kontorova model by Coppersmith[15] also lead to an $\omega^\alpha$ (with $\alpha<1$) behavior in the limit where the mode is pinned due to strong lattice interactions. Such behavior also follows from extended calculations[16] based on a simple one-dimensional rate equation connecting excitation amplitudes $P_n$ at different sites by

$$dP_n/dt = W_{n,n+1}(P_{n+1}-P_n) + W_{n-1,n}(P_{n-1}-P_n), \tag{8}$$

with $W_{n,n\pm1}$ representing random nearest-neighbor transfer rates. For a density of transfer rates which diverges as $W^{-\alpha}$ for $W\rightarrow 0$, Eq. (2) is recovered in the low-frequency limit.

In all of these approaches the distribution of relaxation times, or transfer times between various configurations in close proximity in energy, determines the low-frequency transport properties. This is in contrast to arguments which lead to Eqs. (4) and (6) which are based on evaluation of the density of states, and assume instantaneous transitions between these states, with $\sigma$ evaluated from the absorption probability. Our experiments suggest that a relaxation-dynamics approach is more appropriate than the density-of-states analysis for interpreting the dynamics of charge density waves.

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