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ac response of pinned-charge-density-wave conductors

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The ac response of the pinned-charge-density-wave conductor orthorhombic TaS_3 at low frequencies is found to be strongly ac-amplitude dependent for amplitudes well below the dc threshold field. In the low-amplitude regime, the behavior is strongly temperature dependent suggesting the existence of thermally activated relaxation processes. The ac-amplitude dependence reflects the presence of a distribution of local pinning fields.

Quasi-one-dimensional conductors such as $NbSe_3$ and TaS_3 form a spatially periodic charge density along the chain direction below the Peierls transition temperature. Below a dc threshold field, this charge-density-wave (CDW) condensate is pinned by impurities.[1] However, at finite frequencies or above the threshold field E_T, the CDW slides yielding a unique mechanism of electronic transport. The behavior of the collective mode in the pinned regime has been studied extensively in recent years through the measurements of ac conductivity and dielectric constant. Early measurements[2] were found to be in qualitative agreement with a rigid CDW model[3] which predicts a classical overdamped oscillator response. However, recent low-frequency measurements in orthorhombic TaS_3 (Refs. 4 and 5) and the blue bronze[6,7] ($K_{0.3}MoO_3$) found signigicant departure from the expected low-frequency response:

$$\sigma'(\omega) \sim \omega^2;\quad \sigma''(\omega) \sim \omega \quad , \tag{1}$$

where σ' and σ'' are the real and imaginary parts of the complex ac conductivity. The results reflect strong disorder: A broad distribution of relaxation times is present in both materials. The temperature dependence of the relaxation-time spectrum in the latter material[6] indicates an activated behavior, very similar to that reported recently in the orientational glass $(KBr)_{0.5}(KCN)_{0.5}$.[8]

In this Rapid Communication, we report measurements of the ac conductivity in orthorhombic TaS_3 between 10 Hz and 10 MHz over a wide temperature range. We demonstrate that the dynamics of the pinned CDW is considerably more complicated than has previously been recognized. We have found that in the pinned state, the ac conductivity is strongly nonlinear for ac amplitudes well below the dc threshold field. Moreover, in the linear response regime, the results are significantly different from what have been reported.[4,5] In what follows, we demonstrate that the low-frequency ac response in the small amplitude regime is caused by relaxational processes associated with thermally activated transitions among metastable states. We also describe the ac nonlinearity at higher amplitudes and propose that its frequency dependence provides evidence of a distribution of local pinning forces.

Measurements were made with an HP4192A impedance analyzer on samples in a two-probe configuration with platinum paint contacts. Figure 1 shows the frequency dependence of σ' and σ'' at 120 K for different values of the ac amplitude ($V_{ac} = V_0 \cos\omega t$). Clearly, both σ' and σ'' depend on the ac amplitude for amplitudes as small as 5% of the dc threhsold voltage, V_T. (For this sample, $E_T = 600$ mV/cm at 77 K.) This is in contrast with previous measurements[4] where no amplitude dependence was reported to exist until the ac amplitude was comparable to V_T. These measurements also indicated a power-law dependence of σ' and σ'' on ω. The exponents for σ' and σ'' were found to be nearly equal and temperature independent and suggested a divergence of the dielectric constant, ϵ, at low frequency.

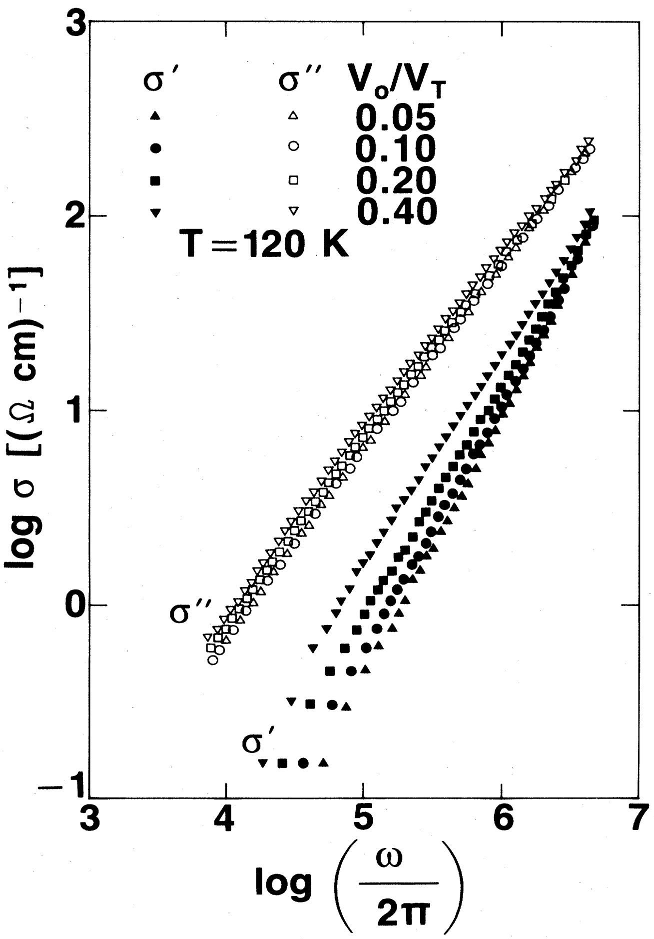

FIG. 1. Frequency dependence of the real and imaginary parts of the ac conductivity at 120 K for various ac amplitudes.

Our low-amplitude results are inconsistent with all these findings.

As shown in Fig. 1, σ' rises faster with ω than σ'' at low amplitudes. A power-law analysis of the data yields a higher exponent for σ' than for σ'' (about 1.35 and 0.95, respectively). However, a simple power-law description of the frequency dependence is incorrect, as we show below. The frequency dependence clearly changes with amplitude in Fig. 1. The slope of σ' changes more rapidly than that of σ'', and at high amplitudes, they are almost parallel. Since this is the behavior reported in previous measurements,[4,5] they are likely to have been made in this high-amplitude regime and do not reflect the low-amplitude behavior.[9]

We now focus our attention on the temperature dependence of the ac response. Here the differences with earlier studies[4,5] are even more striking. A more instructive way to display the results is to plot the real and imaginary parts of the dielectric constant $\epsilon'=\sigma''/\omega$ and $\epsilon''=\sigma'/\omega$. Figure 2 shows the frequency dependence of ϵ' and ϵ'' at various temperatures in the low-amplitude regime. In strong contrast with earlier studies[4,5] we find the frequency dependence changes rapidly with temperature.[10] At high temperatures, e.g., 180 K, ϵ' is essentially frequency independent in the frequency range shown, while ϵ'' increases linearly with frequency [this implies $\sigma'\sim\omega^2$ and $\sigma''\sim\omega$ as in Eq. (1)]. As temperature decreases, frequency dependence appears in ϵ'. The value of ϵ' decreases rapidly above a frequency which decreases with decreasing temperature. Analogous behavior is seen in ϵ''. Clearly, a single power law cannot fit these data, and ϵ' remains finite at low frequency.

This behavior is similar to what has been observed[6] in $K_{0.3}MoO_3$, where it was attributed to a distribution of relaxation times. Our measurements on TaS_3 suggest that this is a generic feature of CDW dynamics. Within a relaxation-time description, the conductivity σ' is given by

$$\sigma'\sim\int\frac{\omega^2\tau g(\tau)\,d\tau}{1+(\omega\tau)^2}\ . \tag{2}$$

If the relaxation time τ reflects hopping between metastable states, it will generally be of the form $\tau\sim\tau_0\exp(B/k_BT)$, with B the barrier height and τ_0^{-1} the attempt frequency. A distribution $g(\tau)$ can arise from a distribution of barrier heights, a distribution of attempt frequencies, or a combination of both. In either case, as the temperature is lowered, the characteristic frequency will decrease. A distribution of barrier heights will also lead to broadening of the distribution of relaxation times as T decreases. Our data show both a decrease in the characteristic frequency and a broadening. The decrease in characteristic frequency is consistent with a typical barrier height of 600 K. However, this value is sample dependent and appears to increase with increasing E_T. A similar distribution of relaxation times and typical barrier height are observed in the "$1/f$" noise.[11] More work is needed to clarify the connection between these distributions and whether a fixed distribution of barrier heights can fit the data at all temperatures.

The temperature dependence of the low-frequency ϵ' is also interesting. While above 100 K, ϵ' scales approximately as the inverse of E_T,[12] below 80 K ϵ' drops precipitously. This drop cannot be accounted for by the increase in E_T. Instead, the frequency and temperature dependence of ϵ' is symptomatic of the onset of a glassy state,[8] where the relaxation times have become very long, i.e., the deformations of the CDW are frozen-in and cannot respond to the ac field. This behavior might also reflect the presence of an incommensurate-commensurate transition. Detailed studies of this temperature region will be presented elsewhere.

We now return to the ac-amplitude dependence. As the ac amplitude increases the value of ϵ'' at low frequencies rises more rapidly than that at higher frequencies (Fig. 1). Interpretation of the data in terms of a relaxation-time picture implies that the mean relaxation time increases. One model proposes that the relaxation time is activated in both temperature and field.[13] This implies a decrease in the relaxation time with increasing field, which is exactly opposite to the experimental trend. The ac-amplitude dependence has also been observed in $K_{0.3}MoO_3$,[14] where it has been attributed to field-assisted transitions among metastable states that are not accessible at lower fields. The barriers for these new transitions are assumed to be higher than at low fields, leading to longer relaxation times. While an increased ac amplitude will be able to excite the CDW over more of the low barriers between metastable states, this should only result in a decrease in ϵ'' at the high frequencies corresponding to these low barriers. It is difficult to understand how it would lead to the observed increase in ϵ'' at all frequencies. We propose a different explanation.

Figure 3 shows the amplitude dependence of the dielectric constant and the conductivity for different frequencies at 77 K. At low frequencies, amplitude dependence is not measurable until V_0 is close to V_T, and then the response increases rapidly. At higher frequencies ac-amplitude dependence is observed at lower amplitudes and increases more slowly. We note that the rigid CDW model of an overdamped particle in a sinusoidal potential also is intrinsically nonlinear. In this model the nonlinear part of the ac conductivity increases as V^2 until the amplitude reaches the pinning field where it increases more rapidly. In our experi-

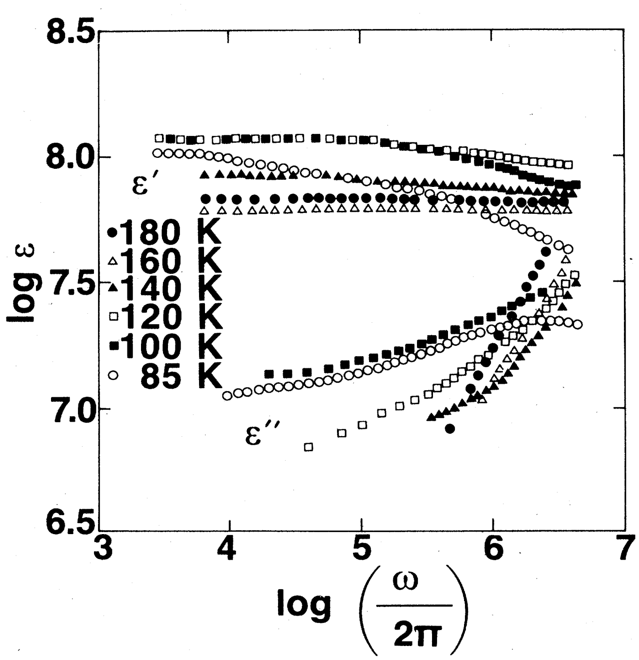

FIG. 2. Low-amplitude ac response at diffeent temperatures. ϵ' and ϵ'' represent the real and imaginary parts of the dielectric constant. See text for discussion.

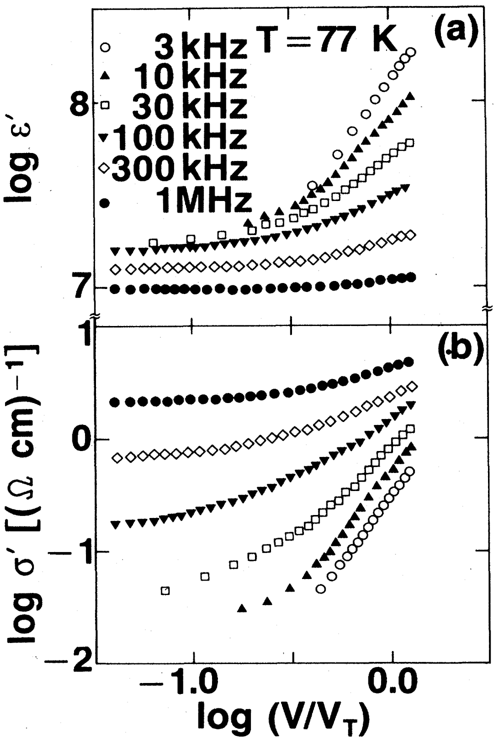

FIG. 3. Amplitude dependence of (a) the real part of the dielectric constant and (b) the real part of the conductivity at 77 K.

mental results the nonlinear component of σ rises more slowly than V^2, and the effective exponent decreases markedly with increasing frequency.

We propose that this behavior reflects the existence of a distribution of local pinning fields. In classical models of CDW conduction, impurities produce local pinning forces whose strength varies spatially. There is a coherence length for CDW motion due to the deformation energy of the condensate. At zero frequency, this coherence length diverges as $V \to V_T$, leading to a single sharply defined dc threshold representing an average of the local pinning strengths.[15] We propose that at finite frequencies the response samples the inhomogeneity in pinning strengths. At a finite frequency there is a maximum length for correlated motion because of the time needed for information to propagate. For the diffusive dynamics usually assumed, this length $L \propto \omega^{-1/2}$. At high frequencies this characteristic length is small, and the effective distribution of pinning fields is broad. Lowering the frequency is equivalent to a coarse graining of the system which leads to a sharper distribution function, as the central limit theorem would imply. This qualitative picture is consistent with Fig. 3. At high frequencies ac-amplitude dependence is appreciable at very low amplitudes, indicating that the distribution of local pinning fields extends to these low values. At lower frequencies the amplitude dependence becomes sharper and approaches that for a single pinning field. This variation of the local pinning field distribution should also affect the frequency dependence of the low-amplitude conductivity.

The qualitative behavior of the ac-amplitude dependence is the same at higher temperatures, although there are quantitative differences. At higher temperatures the ac-amplitude dependence at a given frequency moves to higher amplitudes and sharpens. A possible explanation of this behavior is that thermal energy helps to overcome the low-lying barriers, thereby providing a low-end cutoff for the distribution of pinning fields. However, other factors also change with temperature, such as the relaxation times, number of normal electrons excited across the gap, and the characteristic pinning frequency (~ 100 MHz). In addition, the conductivity of normal electrons increases with temperature, reducing the accuracy of measurements. What effect these factors have is not clear.

The results presented here raise several questions which need to be addressed theoretically. The effect of local inhomogeneities in pinning field on the ac conductivity and its amplitude dependence should be explored. Furthermore, a variety of experiments indicates that thermal effects are important. Theoretical studies are urgently needed to understand what thermal effects are essential in CDW dynamics.

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