PHASE COHERENT CHARGE-DENSITY-WAVE RESPONSE VERSUS METASTABLE STATES

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ABSTRACT

The frequency dependent response of the CDW condensate is examined in $K_{0.3}MoO_3$. At low temperatures the internal degrees of freedom play minor role in the dynamics of the collective mode, although the presence of metastable pinned configurations can be seen by sensitive methods. In contrast, at temperatures where screening by normal carriers allows internal deformations the long range phase coherence is lost and a low frequency mode emerges in the ac response.

INTRODUCTION

In semiconducting CDW systems normal carriers excited through the single particle gap are frozen out at low temperatures. In the ground state the condensate is expected to be different than at temperatures where electrostatic screening is effective, since unscreened Coulomb forces may prohibit local CDW deformations and restore long range order even in presence of impurities. Indeed, experimental observations of a novel type of CDW conduction at low temperatures can be well described in terms of a phase coherent, "rigid" condensate.[1,2] We show that the rigid CDW picture is consistent also with the dielectric properties of the pinned state, and effects associated with inherent degrees of freedom may give corrections only on long time scale.

The dynamics is more complex in the temperature range where screening by normal carriers allows CDW deformations.[3] The possibility of local CDW polarization gives rise to an additional low frequency "peak" in the ac response. We suggest that this mode is the same

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excitation of the condensate as that which appears in glassy relaxation between metastable states. In the temperature range where deformations are allowed the long range phase coherence is lost but still remains certain velocity coherence.[4-6]

In this paper we discuss the essential difference between the responses where internal deformations play a role or are suppressed. Because these are intimately related to the temperature through the screening by normal electrons, we call these two behaviours high temperature and low temperature response.

LOW-TEMPERATURE DIELECTRIC RESPONSE

We have investigated the complex dielectric constant of $K_{0.3}MoO_3$ at $T = 4.2\,K$ in a wide frequency range. At low frequencies $\varepsilon(\omega)$ was determined from the time dependence of the reversible polarization[7] by Laplace transformation:

$$\varepsilon(\omega) = \varepsilon_\infty + \int p'(t) \exp(i\omega t)dt, \qquad (1)$$

where p'(t) is the time derivative of the polarizability (it is the charging current density following an electric field step of unity). Due to the huge dielectric constant the impedance of the sample is easily measurable at higher frequencies, thus standard methods were applied above 10 MHz . Figure 1. shows the real part of the dielectric

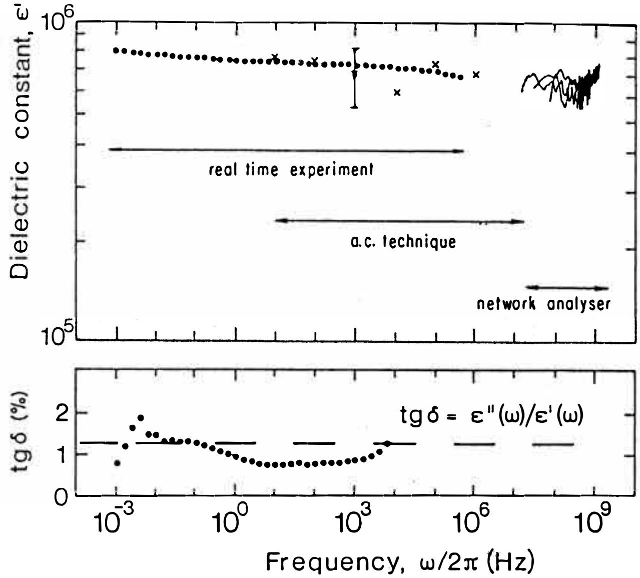

Fig. 1.
The dielectric constant and the loss factor of $K_{0.3}MoO_3$ at $T = 4.2\,K$ shown on logarithmic frequency scale.

constant and the loss factor ($tg\ \delta = \varepsilon''/\varepsilon'$) in the frequency range of 10^{-3} to 10^9 Hz. The dielectric response of the pinned condensate is characterized by virtually constant phase angle and weakly frequency-dependent dielectric constant.

The absence of dielectric cutoff in $\varepsilon(\omega)$ up to $\omega/2\pi = \tau^{-1} \simeq 2$ GHz and its magnitude of about $\varepsilon \simeq 8 \times 10^5$ signifies low damping.[8] In a simple single degree of freedom picture (i.e. with $\sigma(\omega)$ described by a single harmonic oscillator) these numbers are suggestive for a CDW conductivity of $\sigma_{CDW} \simeq \varepsilon/\tau > 10^5\ \Omega^{-1}cm^{-1}$ This estimate is consistent with the low damping in the sliding state, observed experimentally above the depinning threshold.[1]

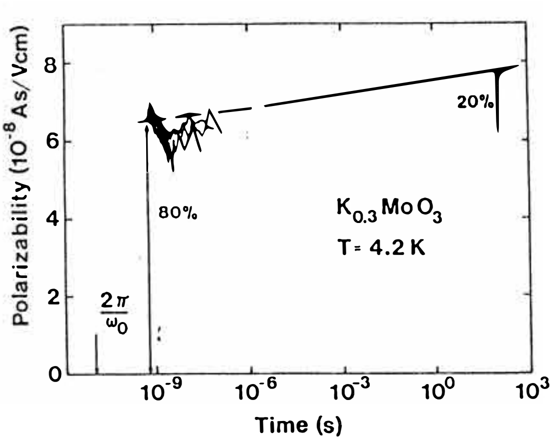

Fig. 2.
Time dependence of CDW polarization at T = 4.2 K There is a logarithmic term superimposed on the rapid response.

When an electric field step is applied, the polarization should develop exponentially with the above time constant of $\tau < 500$ ps and no further variation is expected - at least in the single degree of freedom picture. This is not the case, and polarization experiments performed on long time scale are suggestive for a slow rearrangement of the pinned condensate.[7] Figure 2 shows the time dependence of the polarization (the same data as in Fig. 1 in a different representation). The glassy relaxation of the polarization extends up to several minutes, and reflects deviations from the phase-coherent, single degree of freedom picture. We emphasize, however, that this additional relaxation gives only about 20 % contribution to the total polarization at any reasonable experimental time scales ($t < 10^3 - 10^4$ s), and in most cases it means only a small correction to the rapid response.

We believe that the glassy nature observed in the low-temperature thermal properties (including weak relaxation following a temperature step)[9-11] has the same origin as the slowly varying term in the polarization. It is important to note that the density of states involved in this additional specific heat term is extremely small.[9] The magnitude of the correction due to metastable CDW states is as small as the nuclear hiperfine contribution to the specific heat.[10,11]

In summary, at low temperatures, in absence of screening carriers, the dielectric studies suggest a phase-coherent CDW. The role of the internal degrees of freedom are strongly suppressed in the dynamic properties, and corrections due to metastable states may appear only on long time scales.

THE HIGH TEMPERATURE DIELECTRIC RESPONSE

Thermally excited carriers fundamentally modify the nature of the condensate by the electrostatic screening they provide. The reduction of the Coulomb interaction allows CDW deformations and the long range phase coherence is removed. The local neutrality requires screening currents for any deformation.[12,13] In the dynamic properties the dissipation of these currents determines not only the effective damping, but also the characteristic time of the excitations. The coupling between single particle and the collective excitation in this temperature range has been experimentally demonstrated both for the sliding state above the threshold[14-16] and for the low field ac response.[3,17]

The dielectric behaviour resembles to that of glassy systems. The smeared out peak of $\varepsilon''(\omega)$ observed around liquid nitrogen temperature was described by introducing a wide distribution in the relaxation times.[17] The similarity to glasses is more obvious at somewhat lower temperature, where real time experiments suggest a stretched exponential relaxation for the polarization:[18]

$$p(t) = p_0 \exp\left\{-\left(\frac{t}{\tau_n}\right)^{1-n}\right\} , \qquad (2)$$

with $n = 0.3$ Equation(2) is a phenomenological expression, which is often applied for relaxation processes in systems of many metastable states with slight energy separation. In our case the above time dependence describes the rearrangement of a deformed condensate through metastable pinned configurations.

The characteristic time, τ_n, is strongly temperature dependent, and the excitations slow down exponentially with decreasing temperature[17,18]. The peak in $\varepsilon''(\omega)$, which appears in the 10^4 - 10^6 Hz frequency range at $T = 77$ K, is expected to shift to a few Hz around 30 K, where the weak relaxation was found in the real time experiments. In fact, if the complex dielectric constant is derived from the polarization data by applying Eq(1), its frequency dependence is the same as that found by ac methods at higher temperatures. In Fig. 3 we show $\varepsilon(\omega)$ in a wide temperature range, together with the fit to the Laplace transform of the stretched exponential expression. Note, that all the fitted curves (solid lines) use $n = 0.3$, and the only free parameter is the magnitude of $|\varepsilon|$ Although the applicability of a linear transformation theory to the relaxation data is not verified yet experimentally, we believe that the data shown on Fig. 3 are convincing that the weak relaxation process and the smeared response of the ac excitation is the same phenomenon. This has already been suggested on theoretical grounds by Littlewood and Rammal.[19]

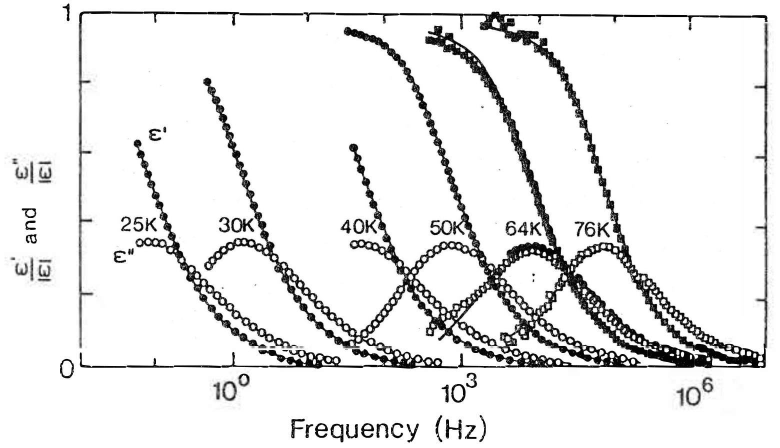

Fig. 3. Real and imaginary part of the dielectric constant (■ from Ref. 17, ● : Laplace transformed data of Ref. 18). The solid line corresponds to $\varepsilon(\omega)$ calculated from Eq(2).

While the dielectric behaviour clearly reflects a rich spectrum of the internal CDW excitations, certain coherence still exists in the current carrying state. Most of the relevant experiments[4-6] are based on narrow band noise studies. These measurements show the continuity of the CDW current density, which remains constant even in strongly perturbed circumstances (e.g. in presence of a temperature gradient). Such a nonlocal behaviour is suggestive for a long range velocity-velocity coherence, but does not exclude inherent phase excitations. As phason excitations do not require local CDW → normal-electron conversion the narrow band noise frequency is not influenced by the CDW deformations. While these inherent excitations certainly destroy the phase coherence, leading to a finite phase-phase coherence length L_o, the velocity coherence may well exceed L_o.[4]

In summary, in the temperature range where normal carrier screening is important the low frequency dielectric behaviour is determined by the internal dynamics of a deformable CDW. The phase coherence is lost and the polarized CDW relax through various metastable pinned configurations.

CONCLUSION

We have studied two, basic aspects of CDW phenomena; the macroscopic coherence effects and the presence of metastable states. We argue that their relative importance is different in the low-temperature and in the conventional CDW dynamics. At low temperatures the internal degrees of freedom are suppressed, and a phase coherent approach gives a good phenomenological description for the dynamic properties. On the other hand, when screening by thermally excited normal carriers allows deformations, the situation is dramatically different, and in that case the role of the internal CDW excitations is dominant.

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