Inertial Dynamics of Charge-Density Waves in TaS_3 and $NbSe_3$

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Frequency-dependent complex-conductivity measurements are reported in the charge-density-wave states of TaS_3 and $NbSe_3$, in the 10–100-GHz spectral region. A Drude-type behavior associated with damped inertial response of the condensate is observed for the first time. The fundamental parameters that characterize the dynamics of the collective mode (damping constant, pinning energy, and effective mass) are obtained. The results for the effective mass and damping are compared with microscopic theories.

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Strongly frequency-dependent response due to pinned charge-density-wave (CDW) condensates has been intensively studied[1] in materials which show transitions to the Peierls-Fröhlich state. In particular, for the transition-metal trichalcogenides $NbSe_3$ and TaS_3, measurements in the radio-frequency spectral range show that the conductivity increases with increasing frequency between 400 Hz and 10 GHz and that the dielectric constant is positive. This behavior can be understood, to a first approximation, in terms of a relaxation equation of motion $\dot{x}/\tau+\omega_0^2 x=(-e/m^*)E(t)$ for the displacement $x(t)$ of the CDW due to the driving electric field $E(t)$. τ, ω_0, and m^* are respectively the phenomenological damping constant, pinning frequency, and effective mass.

Inertial effects due to the acceleration of the CDW have not been clearly observed to date. Further, the existing experiments yield information regarding only one parameter, the "crossover" frequency $\omega_{co}=\omega_0^2\tau$, and reliable[2] experimental values for the fundamental parameters of the CDW mode—τ, ω_0, and m^*—have not been available. Consequently, it has not been possible to make meaningful comparison to microscopic theories which attempt to calculate these fundamental parameters.

In this communication we report new frequency-dependent complex-conductivity measurements in the hitherto unexplored 10–110-GHz frequency range in two model systems: orthorhombic TaS_3 and both CDW phases of $NbSe_3$. We find a Drude-type high-frequency behavior for

$$\sigma_{CDW}(\omega)=\mathrm{Re}\,\sigma_{CDW}(\omega)+j\,\mathrm{Im}\,\sigma_{CDW}(\omega)$$

given by

$$\sigma_{CDW}(\omega)=\sigma_{max}\frac{1}{1+j\omega\tau}. \qquad (1)$$

Equation (1) can be obtained from an equation of motion $\ddot{x}+(1/\tau)\dot{x}=(-e/m^*)E(t)$ which describes the damped inertial response of the CDW condensate (ignoring pinning). From the observed frequency dependence, the damping parameter τ is extracted. Furthermore, from the measured maximum conductivity $\sigma_{max}=ne^2\tau/m^*$ and the "crossover" frequency $\omega_0^2\tau$ obtained from lower-frequency (10 MHz–10 GHz) measurements, we evaluate m^* and ω_0.

The inertial response described by Eq. (1) is the high-frequency part of the complete dynamical response of the CDW which can be respresented[3] by

$$\sigma_{CDW}(\omega)=\frac{ne^2\tau}{m^*}\frac{j\omega}{\tau(\omega_0^2-\omega^2)+j\omega}. \qquad (2)$$

For the first time all the relevant phenomenological parameters—τ, m^*, and ω_0—which characterize this dynamical response are experimentally obtained. The measured effective masses are in order-of-magnitude agreement with those obtained from the theory of Lee, Rice, and Anderson.[4]

The samples used in these measurements were prepared at the University of California at Los Angeles by techniques described elsewhere.[5] The nominally pure orthohombic TaS_3 has a typical threshold field of 150 mV/cm at 160 K. The $NbSe_3$ samples had a typical threshold field of 30 mV/cm at 45 K. In both the materials the samples selected for conductivity measurements were $\sim 1\ \mu m^2$ in cross section.

The conductivities at 9 and 35 GHz were measured with a standard cavity perturbation method.[6] At higher frequencies, this method became impractical and a new waveguide technique was developed. This technique uses a millimeter-wave bridge to measure the complex impedance of a section of shorted waveguide containing the sample. The sample was electrostatically fixed to a quartz fiber and placed parallel to and at a maximum of the electric field. From measurements of the bridge parameters with and without the sample, the sample complex impedance Z_s was determined. This can be represented as

$$Z_s=jX_{inf}+\alpha/\sigma, \qquad (3)$$

where σ is the complex conductivity and α is a geometric factor determined by sample size. [All impedances in Eq. (3) are normalized to the characteristic waveguide impedance.[7]] X_{inf} is the purely reactive impedance of an equivalent infinitely conducting wire, identical in size and shape to the sample and placed at the same position. Thus X_{inf} is determined purely by geometry and is independent of the sample conductivity. One method of determining X_{inf} is to measure Z_s when the sample is highly conducting. For $NbSe_3$, this is the case for $T < 15$ K and X_{inf} was thus measured. For TaS_3, where σ is never high, X_{inf} was determined from Z_s at room temperature where the sample is metallic by setting $\mathrm{Im}\sigma = 0$. In both cases, α was determined by normalization of $\mathrm{Re}\sigma$ to σ_{dc} above the phase transition, i.e., in the metallic phase. By this technique, σ was determined at fixed frequencies 35, 60, 94, and 110 GHz in the temperature range 300 to 15 K. While both the in-phase and the out-of-phase componets of σ were measured in TaS_3, in $NbSe_3$, because of the large dc conductivity, only the real part could be evaluated. The bridge method yielded results at 35 GHz in agreement with those obtained with use of standard cavity perturbation techniques at the same frequency. Further, the validity of the bridge technique has been checked by extensive measurements on well-known materials such as quartz rods, metallic copper wires, and purely resistive materials, such as $TaSe_3$.

In this paper we present the frequency dependence of the conductivity at a few representative temperatures—160 K for TaS_3 and 45 and 120 K for $NbSe_3$, corresponding to the two CDW states for the latter. At these temperatures, all of the salient features of collective CDW dynamics, such as a sharp threshold field, narrow-band noise, Shapiro steps, etc., are well documented.[1] (Detailed results at other temperatures and detailed comparison to theories will be published elsewhere.)

Figures 1(a), 2(a), and 2(b) present the results for the real part of the CDW conductivity $\mathrm{Re}\sigma_{CDW}(\omega) = \mathrm{Re}\sigma(\omega) - \sigma_{dc}$ at various frequencies for TaS_3 at 160 K and $NbSe_3$ at 45 and 120 K, respectively. From a maximum value at 9 GHz, the conductivity decreases at higher frequencies. The solid line represents a best fit of the real part of Eq. (1) to the data with values for the damping parameter $1/2\pi\tau$ as listed in Table I. [This analysis essentially ignores the effects of pinning. However, including deduced (see later) values of ω_0 and using Eq. (2) would change the fit parameters by only a few percent.] Thus from these measurements we obtain $1/2\pi\tau$ and $\sigma_{max} = \sigma_{CDW}(9\text{ GHz}) = ne^2\tau/m^*$.

Another striking feature of our results is that $\mathrm{Im}\sigma_{CDW}(\omega)$ and hence also the dielectric constant $\epsilon(\omega) = \mathrm{Im}\sigma_{CDW}(\omega)/\omega\epsilon_0$ (ϵ_0 is the free-space permittivity) is *negative,* in sharp contrast to the low-frequency (< 10 GHz) behavior where the dielectric constant is positive. This is another essential aspect of the damped inertial response. Figure 1(b) presents our measurements for $\mathrm{Im}\sigma_{CDW}(\omega)$ normalized to σ_{max} for TaS_3 at 160 K. The solid line represents the imaginary part of Eq. (1) using the same damping parameter derived from the fit of Fig. 1(a), and describes the data very well. Thus Eq. (1), obtained by considering the inertial response of the CDW con-

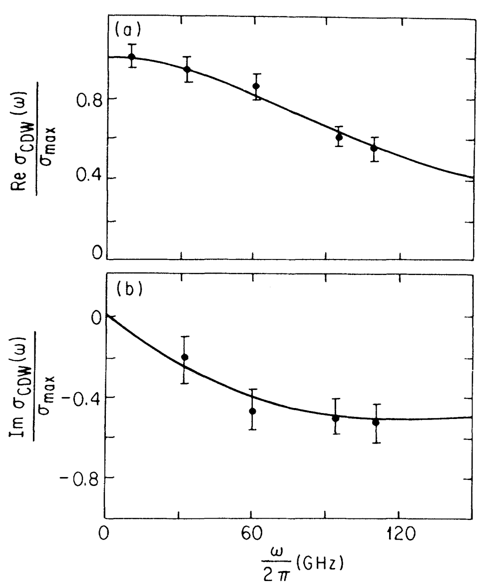

FIG. 1. Frequency dependence of $\sigma_{CDW}(\omega)$ for TaS_3 at 160 K. (a) $\mathrm{Re}\sigma_{CDW}(\omega)$ normalized to $\sigma_{max} = \mathrm{Re}\sigma_{CDW}(9$ GHz). (b) $\mathrm{Im}\sigma_{CDW}(\omega)$ normalized to σ_{max}. The solid lines represent the real and imaginary parts of Eq. (1) with a single fit parameter $1/2\pi\tau = 125$ GHz.

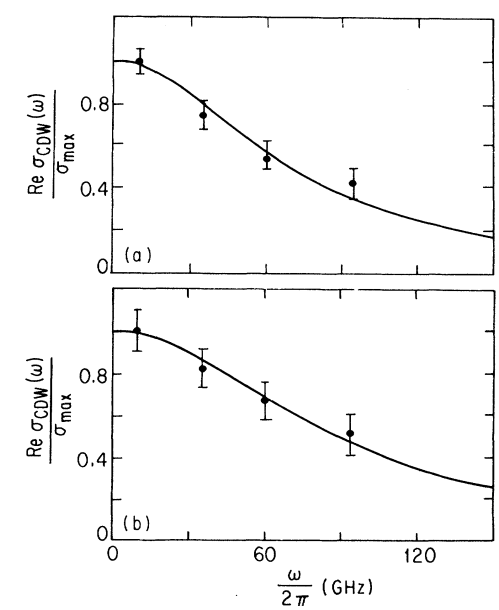

FIG. 2. Frequency dependence of $\mathrm{Re}\sigma_{CDW}(\omega)$ normalized to $\sigma_{max} = \mathrm{Re}\sigma_{CDW}(9$ GHz) for $NbSe_3$ at (a) 45 K and (b) 120 K. The solid lines represent the real part of Eq. (1) with fit parameters $1/2\pi\tau$ of (a) 70 GHz and (b) 90 GHz.

TABLE I. Charge-density-wave parameters for $NbSe_3$ and TaS_3 deduced from the present experiments.

	$\sigma_{max} = \frac{ne^2\tau}{m^*}$ $(\Omega\text{-cm})^{-1}$	$\frac{1}{(2\pi\tau)}$ $(\sec^{-1})$	$\frac{\omega_{co}}{2\pi} = \frac{\omega_0^2\tau}{2\pi}$ $(\sec^{-1})$	$\frac{\omega_0}{2\pi}$ $(\sec^{-1})$	$\frac{m^*}{m_e}$	$\Delta(0)$ (K)	λ	$\frac{m^*}{m_b}$
			Measured				Calculated	
$NbSe_3$ (45 K)	24 000	70×10^9	100×10^6	2.6×10^9	117	106[a]	0.23	78
$NbSe_3$ (120 K)	2000	90×10^9	N/A	N/A	270	253[a]	0.30	340
TaS_3 (160 K)	2500	125×10^9	150×10^6	4.3×10^9	940	390[a]	0.34	715
						700[b]		2303

[a]From mean-field theory, $\Delta(0)=1.74kT_P$.

[b]From resistivity measurements of Ref. 10.

densate and including a phenomenological damping term, provides an excellent description of our data for both $\mathrm{Re}\sigma_{CDW}(\omega)$ and $\mathrm{Im}\sigma_{CDW}(\omega)$. (For $NbSe_3$, we have established that ϵ is again negative in both CDW phases; however the accuracy of the magnitude is much less as a result of the high conductivity of the sample at all temperatures.)

From the measured σ_{max} and $1/2\pi\tau$, the effective mass m^* can be determined if n is known. At these temperatures, well below the transition temperatures, we assume that all of the electrons are condensed. For TaS_3 we use $n=6.5\times10^{21}$ carriers/cm^3, as determined from the lattice parameters with the assumption of a quarter-filled band.[1] For $NbSe_3$, the total carrier density is similarly estimated[1] to be $5.5\times10^{21}/cm^3$. The Fermi surface does not completely separate at the two transitions; 20% of the Fermi surface is lost at $T_1=149$ K and an additional 60% is lost at $T_2=60$ K.[8] We therefore use $n=0.2(5.5\times10^{21})/cm^3$ at 120 K and $n=0.8(5.5\times10^{21})/cm^3$ at 45 K. The choice of carrier densities below T_2 implicitly assumes that all of the condensed electrons in the lower phase have the same damping and effective mass although they have different pinning frequencies. The effective masses, calculated[9] from the measurements and normalized to m_e (the free electron mass), are listed in Table I and labeled as m^*/m_e measured.

The microscopic mean field theory of Lee, Rice, and Anderson[4] predicts a CDW effective mass of

$$m^*/m_b = 1+4\Delta^2(0)/\lambda(\hbar\omega_{2k_F})^2, \tag{4}$$

where λ is the electron-phonon coupling constant, ω_{2k_F} is the phonon frequency at wave vector $2k_F$, m_b is the band mass, and $\Delta(0)$ is the single-particle gap at zero temperature. To determine the parameters in Eq. (4) we use the mean-field result, $\Delta(0)=1.74k_BT_P$, to calculate the gap and $\Delta=2\epsilon_F\exp(-1/\lambda)$ to calculate λ. We estimate the Fermi energy, using a tight-binding model, to be $2\epsilon_F=0.6$ eV. $\hbar\omega_{2k_F}$ is assumed to be approximately the Debye temperature, $\hbar\omega_{2k_F}/k_B\approx50$ K. The calculated values of λ, $\Delta(0)$, and thus m^*/m_b are also in Table I.

Within the uncertainties in the numerical values for the parameters in Eq. (4), the mean-field theory of Lee, Rice, and Anderson describes our measurements for m^* reasonably well. The greatest uncertainties are in $\Delta(0)$, m_b, and $\hbar\omega_{2k_F}$. In TaS_3, because of strong one-dimensional fluctuations, $\Delta(0)$ can be considerably higher[10] (~700 K), leading to a correspondingly higher m^*/m_b, also listed in Table I. It should also be noted that the measured m^*/m_e value for $NbSe_3$ at 45 K is affected by the estimate of n based upon the assumption of a single CDW.

A microscopic theory for the damping of CDW motion due to interaction with thermal phasons has been initiated by Takada, Wong, and Holstein.[11] The theory is expected to be applicable to TaS_3 and yields an estimate of 6×10^4 $(\Omega\text{-cm})^{-1}$ for the limiting CDW conductivity at 160 K due to this mechanism, larger by an order of magnitude than σ_{max} measured here. With $m^*/m_e=940$, this leads to $1/2\pi\tau\sim5$ GHz. Additional theoretical ingredients, in particular inclusion of long-range Coulomb interactions, are expected to enhance the predicted magnitude of $1/2\pi\tau$, and hence decrease the theoretical maximum conductivity. Detailed analysis including temperature-dependent effects implied by the theory will be reported later.

In terms of Eq. (2), the complex conductivity at radio frequencies has been well documented to have an "overdamped" behavior, with $\omega,\omega_0 \ll 1/\tau$. These low-frequency measurements yield the "crossover" frequency, $\omega_{co}=\omega_0^2\tau$, which is the frequency of the observed peak in $\mathrm{Im}\sigma_{CDW}(\omega)$. We have obtained ω_{co} from a large body of experimental data[10] on $\sigma(\omega)$ for samples with threshold fields comparable to those obtained in the present study. Using ω_{co} and the damping τ measured in the present experiments, we calculate ω_0, the pinning frequency as displayed in Table I. While ω_0 calculated from microscopic theories depends on the (unknown) impurity potentials and concentrations, the values obtained in Table I are in good order-of-magnitude agreement with values estimated from the pinning theory of Lee and Rice and Fukuyama and Lee.[12]

Our measurements on two CDW systems clearly demonstrate that the CDW condensate has inertia, which, in the presence of damping, manifests itself in a Drude-type behavior for the complex conductivity at microwave and millimeter-wave frequencies. These measurements allow a complete characterization of the long-wavelength CDW phason mode since all of the fundamental parameters are now obtained. Our results are in approximate agreement with the microscopic theory of Lee, Rice, and Anderson.

We also note that the tunneling description of $\mathrm{Re}\sigma(\omega)$, which also accounts for the experimental results obtained at radio frequencies,[13] does not lead to a decrease of the conductivity at higher frequencies as observed by us. However, inertia effects, similar to that described by Eq. (2), are expected to occur,[14] with a damping constant related to the high-frequency and high-field limit of CDW conduction.

We believe that these results for the damping and effective mass have important implications for other experiments, such as chaotic CDW behavior and subharmonic response, where inertial dynamics is invoked.

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