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ac DYNAMICS OF $NbSe_3$ IN THE SWITCHING REGIME

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We have measured the low-field ac conductivity $\sigma(\omega)$ of switching samples of $NbSe_3$ in the lower charge density wave (CDW) state, over the frequency range 10 Hz to 500 MHz. For zero applied dc bias fields, $\sigma(\omega)$ reflects overdamped response, with no evidence for dominant inertial contributions. For dc bias fields exceeding the threshold E_T, a complicated ac conductivity obtains, whose overall features rule out, as valid descriptions of switching, simple classical overdamped center of mass motion, or overdamped motion with excessive current noise. Our results demonstrate the necessity of additional degrees of freedom in describing switching phenomena.

Several classes of inorganic, quasi-one-dimensional metals undergo Peierls transitions to the charge density wave (CDW) state. In many of these materials, the application of a longitudinal electric field E_{dc} exceeding a well-defined threshold field E_T causes the CDW condensate to depin from the lattice, and to "slide" along the crystal axis[1]. Typically, the velocity of the CDW increases as a smooth function of $E_{dc}-E_T$. In a sizable minority of samples, however, the depinning of the CDW is quite abrupt. This phenomenon, first observed[2] in $NbSe_3$ and known as switching, is manifested as a sharp break in the dc current-voltage (I-V) characteristics of the sample. The onset of CDW conduction in a switching sample is in addition hysteretic, where the onset and extinction of CDW conduction occurs for different values of the applied electric field.

A particularly simple and appealing model of CDW transport is the "rigid particle" description by Grüner, Zawadowski, and Chaikin [3]. In this phenomenological model the CDW is treated as a classical point particle with only a single degree of freedom. The periodicity of the CDW is taken into account by considering the CDW-impurity interaction as a periodic function of the CDW position. If only the first Fourier component of this interaction potential is retained, the center of mass coordinate obeys an equation of motion

$$\frac{d^2x}{dt^2} + \frac{1}{\tau}\frac{dx}{dt} + \frac{\omega_o^2\lambda}{2\pi}\sin(2\pi x/\lambda) = \frac{e\,E(t)}{m^*}. \qquad (1)$$

Here x is the CDW position, $1/\tau = \gamma/m^*$ with γ a damping constant and m^* the effective mass of electrons condensed in the CDW state, $\omega_o^2 = k/m^*$ with k a restoring force constant, and λ is the CDW wavelength.

In the context of switching CDW's, Eq. (1) is attractive in that, without further assumptions, it predicts switching, hysteresis, and chaos, all of which have been observed in switching samples of $NbSe_3$.[2,4] For non-switching samples of $NbSe_3$, the low-field ac conductivity appears overdamped[5], which justifies neglecting the first term on the left (inertial term) in Eq. (1). However, in the framework of Eq. (1), switching, hysteresis, and chaos imply a finite CDW inertia. Our present purpose is to determine to what extent such an implied CDW inertia manifests itself in the dynamics of switching CDW samples, and to what degree Eq. (1) retains integrity by self-consistently describing the switching phenomena.

We have grown crystals of $NbSe_3$ by conventional vapor-transport methods. Several batches of nominally pure material were produced using identical preparation procedures. The ratio of switching samples to nonswitching samples (determined by the dc I-V characteristics at 26 K) varied widely between batches. In some batches, no samples displayed switching, while in other batches the number of switching samples was quite high, of order 10%. Typical threshold fields for switching samples used in this study were E_T = 30 mV/cm at T = 48 K.

The complex low-field ac conductivity of single $NbSe_3$ crystals was measured by two different methods, depending on frequency range. For low frequencies, an HP 4192 computer-controlled impedance analyzer provided continuous frequency sweeps from 10 Hz to 5 MHz, while from 4 MHz to 500 MHz an HP 8754A network analyzer was used. Both systems allowed an external dc bias to be applied to the sample. The ac test signal was typically between 1 mVrms and 2.5 mVrms which corresponded roughly to one percent of E_T in the temperature range of interest. Our experimental set-up also allowed simultaneous measurement of the dc I-V characteristics of the sample, and also simultaneous spectrum analyzer detection of the narrow band noise in the nonlinear conductivity region. A two probe sample mounting configuration with silver paint contacts was used exclusively.

Figure 1 shows both the real and imaginary parts of the low-field complex ac conductivity, $Re\sigma(\omega)$ and $Im\sigma(\omega)$, measured between 4 MHz and 500 MHz, for a $NbSe_3$ crystal at T = 26 K. This data was obtained at zero applied dc bias. The same $NbSe_3$ crystal, however, displayed dramatic switching in the dc I-V characteristics at threshold, as indicated in the inset of Fig. 1. The ac conductivity data of Fig. 1 show that both $Re\sigma(\omega)$ and $Im\sigma(\omega)$ increase smoothly with increasing frequency between 4 MHz and 500 MHz, with no evidence for a threshold frequency for the onset of ac conduction. The crossing of $Re\sigma(\omega)$ and $Im\sigma(\omega)$ indicates a classical "crossover" frequency of 500 MHz.

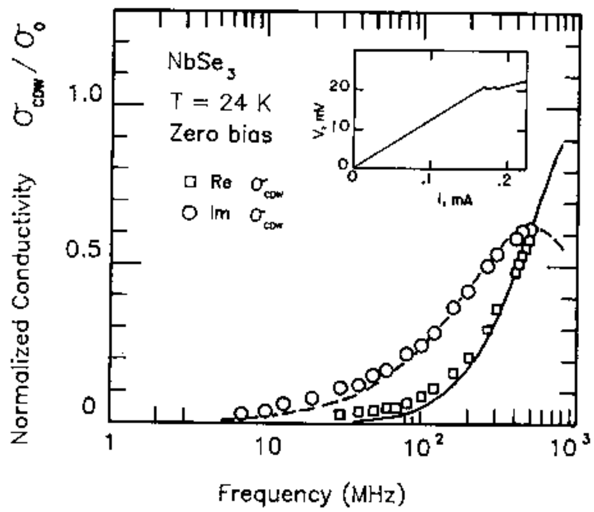

Fig. 1 Low-field ac conductivity $\sigma_{CDW}(\omega)$ of switching $NbSe_3$, in the pinned CDW regime. The inset shows the dc I-V characteristics for the same crystal.

Figure 2 shows $\sigma(\omega)$ for the same $NbSe_3$ sample as was used for Fig. 1, except now a dc bias field exceeding threshold has been applied. The fundamental noise frequency for this value of dc bias was 25 MHz. Figure 2 indicates that, for low frequencies, both $Re\sigma(\omega)$ and $Im\sigma(\omega)$ initially *decrease* with increasing frequency. Between 4 MHz and 20 MHz, $Re\sigma(\omega)$ decreases nearly 70% while $Im\sigma(\omega)$ increases in magnitude, by approximately 25%, while remaining negative in sign. Sharp "dip" structure appears in $Re\sigma(\omega)$ and $Im\sigma(\omega)$ at $\omega/2\pi$ = 25 MHz, 50 MHz, and 75 MHz, the result of interference resonances between the ac test frequency, and the fundamental narrow band noise frequency and its harmonics. In the frequency range between the interference resonances, both $Re\sigma(\omega)$ and $Im\sigma(\omega)$ display similar behavior, first increasing and then decreasing smoothly with increasing frequency. For frequencies above 25 MHz, the general trend for both $Re\sigma(\omega)$ and $Im\sigma(\omega)$ is an increase with increasing frequency, with no apparent saturation in $Re\sigma(\omega)$ or turnover in $Im\sigma(\omega)$, up to 500 MHz.

From Fig. 1, it is apparent that the low-field ac conductivity of switching CDWs *in the pinned regime* is qualitatively similar to that determined previously for nonswitching samples. The general form of $\sigma(\omega)$ indicates overdamped behavior, with a characteristic crossover frequency near 500 MHz. We note that there is no apparent scaling relation between the ac and dc conductivities for $NbSe_3$ in the switching regime. Figure 1 shows a smooth rise in $Re\sigma(\omega)$ with increasing frequency, while earlier studies[6] have demonstrated a differential dc conductivity independent of dc bias above threshold.

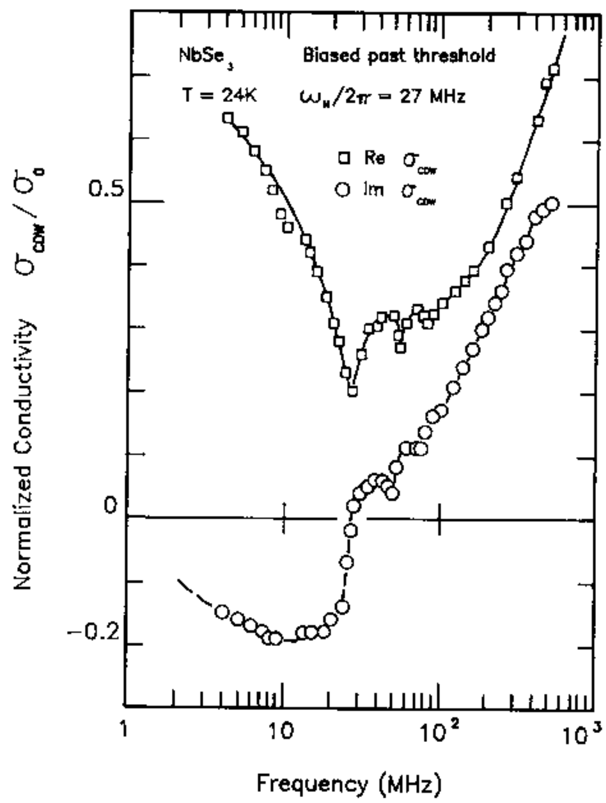

Fig. 2 Low field ac conductivity $\sigma_{CDW}(\omega)$ of switching $NbSe_3$, in the sliding CDW regime. Resonances correspond to interference with narrow-band noise.

To simplify analysis in terms of rigid-particle motion, Eq. (1) may be written in the dimensionless form

$$\beta \frac{d^2\theta}{dt^2} + \frac{d\theta}{dt} + \sin\theta = e_{dc} + e_{ac} \sin\Omega t \qquad (2)$$

where $\beta = (\omega_0\tau)^2$, time is measured in units of $(\omega_0^2\tau)^{-1}$, $\theta = 2\pi x/\lambda$, $e_{dc}=E_{dc}/E_T$, $e_{ac}=E_{ac}/E_T$, and $\Omega = \omega/(\omega_0^2\tau)$. We also define $\omega_0^2\tau = \omega_c$. In the case of zero applied dc bias and small θ, Eq. (2) is easily solved to yield a CDW conductivity

$$Re\sigma(\Omega) = \frac{ne^2\tau}{m^*} \frac{\Omega^2}{[(1-\beta\Omega^2)^2 + \Omega^2]} \qquad (3a)$$

$$Im\sigma(\Omega) = \frac{ne^2\tau}{m^*} \frac{\Omega(1-\beta\Omega^2)}{(1-\beta\Omega^2)^2 + \Omega^2} \qquad (3b)$$

with n the concentration of electrons condensed in the CDW state. The solid and dashed lines in Fig. 1 are Eq. (3), with fitting parameters $\omega_c/2\pi$ = 500 MHz, and $\beta = (\omega_o\tau)^2 = 10^{-3}$. This value of β is appropriate to overdamped response. The parameter values extracted from the fit in Fig. 1 may be compared to those determined from other experiments on switching samples of $NbSe_3$. For example, Shapiro steps observed for switching $NbSe_3$ have been analyzed[4] in terms of Eq. (2). There the variation of Shapiro step magnitude with applied rf amplitude has indicated a crossover frequency of only 22 MHz = $\omega_c/2\pi$. Additional analysis[4] of the degree of hysteresis in the dc I-V characteristics has implied substantial CDW inertia, with $\beta = (\omega_o\tau)^2 = 2.2$. These values of crossover frequency and inertial coefficient determined experimentally for the *depinned* CDW condensate are orders of magnitude different from those extracted from the low field ac conductivity of the *pinned* CDW condensate, as in Fig. 1.[7]

We investigate the possibility that the parameters characterizing CDW dynamics for switching samples are different for pinned and depinned CDW condensates.

Figure 3 shows the results of an analog computer solution of Eq. (2) for the ac CDW conductivity as a function of frequency, with an applied dc bias exceeding threshold.[8] The chosen parameters are $\omega_c/2\pi$ = 22 MHz and β =2.1, close to the values determined from Shapiro step and hysteresis experiments on $NbSe_3$. The applied dc bias field was chosen to generate a narrow band noise frequency at 18 MHz. The computer results of Fig. 3 show a complicated behavior for both $\text{Re}\sigma(\omega)$ and $\text{Im}\sigma(\omega)$. Notable are the relative frequency independence of both $\text{Re}\sigma(\omega)$ and $\text{Im}\sigma(\omega)$ at very low frequencies, and the exceptionally strong interference effects whenever the ac test frequency matches a narrow-band noise frequency (or a higher harmonic). Figure 3 also indicates that the high frequency conductivity becomes again independent of frequency, and both $\text{Re}\sigma(\omega)$ and $\text{Im}\sigma(\omega)$ asymptotically approach zero at very high frequencies. Figure 3 shows that $\text{Re}\sigma(\omega)$ at high frequencies attains a value substantially *lower* than its low frequency, $\omega \to 0$, limit.

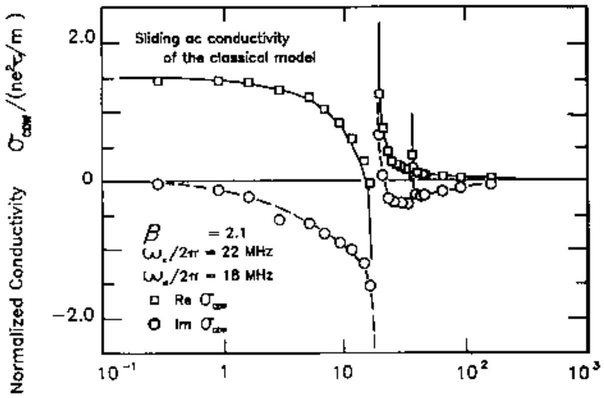

Fig. 3 Low-field ac conductivity $\sigma_{CDW}(\omega)$ as calculated from Eq. (2), in the "sliding" regime. The parameters on the figure are appropriate to underdamped response.

The analog computer predictions of Fig.3 should be compared to Fig.2 which shows the experimental results for $\sigma(\omega)$ in the presence of a dc bias exceeding threshold. An obvious difference between the predictions of Eq. (2) and the experimental results are the magnitudes of the interference effects. Experimentally, relatively small anomalies are observed in $\text{Re}\sigma(\omega)$ and $\text{Im}\sigma(\omega)$ when the ac test frequency matches a harmonic of the narrow-band noise frequency. Eq. (2), on the other hand, predicts nearly divergent behavior in the interference region. This discrepency may not be profound, however, as thermal or broadband noise could smear the resonances and reduce the interference effect in the real CDW crystal. A striking discrepancy between theory and experiment is of course the relative magnitude of $\text{Re}\sigma(\omega)$ measured at low and high frequencies. Experimentally, $\text{Re}\sigma(\omega)$ at high frequencies increases with increasing frequencies, and exceeds in magnitude the low frequency, $\omega \to 0$ limit. Eq. (2), on the other hand, predicts exactly the opposite effect, with the $\omega \to 0$ limit of $\text{Re}\sigma(\omega)$ exceeding the high frequency limit by a factor of five. This particular failure of Eq. (2) however is a direct consequence of the divergent differential conductance predicted near threshold, as the dc bias field approaches threshold from above. Such a divergence problem at threshold is endemic to any single degree of freedom classical description.

The more serious difference between Figs. 2 and 3 is the form of $\text{Im}\sigma(\omega)$ at high frequencies. Experimentally, $\text{Im}\sigma(\omega)$ rises dramatically at high frequencies, while Eq. (2) predicts no out-of-phase component at sufficiently high frequencies. Hence the experimentally observed ac conductivity at both high and low frequencies appears inconsistent with simple center of mass motion of the CDW condensate.

We recall that Fig. 3 is appropriate to classical CDW response in the *underdamped* regime. We have also solved Eq. (2) by analog computer, using parameters appropriate to *overdamped* response, as might be suggested by the results of Fig. 1. The results are not dramatically different from the underdamped case, and the only significant difference appears to be the rate of decay of $\text{Re}\sigma(\omega)$ at high frequency. We note that in this overdamped parameter range, our analog computer results agree well with perturbation theory results for Eq. (2)[9]. We also remark that our $\sigma(\omega)$ results of Fig. 2 appear to rule out a recent model by Wonneberger[10] where switching is induced by current noise, in an otherwise overdamped classical system. In this model, $\text{Im}\sigma(\omega)$ is again predicted to be zero at moderately high frequency in the sliding regime, in sharp contrast to the experimental observations.

Our results thus show that dc switching and the high frequency ac conduction of the CDW condensate cannot be fully described by simple center of mass motion, even with a substantial inertial contribution or current noise. An additional conductivity mechanism is required. The most natural extension to a rigid-particle description is one where additional degrees of freedom are introduced by taking into account internal CDW deformations[11-13]. Although switching has not been explicitly demonstrated for such models, the rich spectrum of metastable states obtainable suggests the possibility of switching phenomena. The high frequency CDW conductivity in the sliding regime could then be totally dominated by internal excitations of the CDW condensate itself. It is not, however, obvious if such a description could in addition be consistent with the *inductive* low frequency response of switching CDWs (see Im$\sigma(\omega)$ in Fig. 2)[14].

We also note that a number of models have recently been suggested to account for switching phenomena. These include a statistical "kinetic Ising" model by Joos and Murray[15], in which coupled domains play a crucial role, and a model of CDW depinning[16] in which macroscopic deformations of the CDW lead to switching. Unfortunately, none of these models makes predictions about the ac response, either in the pinned or depinned regime. However, an essential feature of such models is that they introduce additional degrees of freedom, and hence the possibility exists for an enhanced conductivity at high frequencies, even for a sliding CDW condensate. The domain-coupling model of Joos and Murray is attractive in that it predicts the correct turn-on statistics for CDW conduction at the switching threshold, and is consistent with negative differential resistance and 1/f noise recently observed[17] in switching samples of $NbSe_3$. An incomplete coupling of domains in the sliding CDW region could lead to an enhanced polarization at high frequencies, as observed. We also remark that although Bardeen's tunneling model[18] does not predict switching, an increasing conductivity with increasing frequency (even for a depinned CDW condensate) is in some ways suggestive of tunneling processes. Hence, the possibility of macroscopic quantum tunneling here warrants further investigation.

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