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 $E$ exceeding a well-defined threshold field $E_T$ causes the CDW condensate to depin from the lattice, and to "slide" along the crystal axis. Typically, the velocity of the CDW increases as a smooth function of $E>E_T$. In a sizable minority of samples, however, the depinning of the CDW is quite abrupt. This phenomenon, first observed 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 extincion 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 Orhan, Zawadowski, and Chakravarty. 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

$$\frac{d^2 x}{dt^2} + \frac{2k}{m}\cos(2\pi x) = \frac{E(t)}{E_L}.$$  (1)

Here $x$ is the CDW position, $1/\gamma = \gamma/m^2$ with $\gamma$ a damping constant and $m$ the effective mass of electrons condensed in the CDW state, $2k/m^2$ with $k$ a restoring force constant, and $E_T$ 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$_3$. For non-switching samples of NbSe$_3$, the low-field ac conductivity appears overdamped, 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 non-switching 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 for frequencies 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, $\Re(\sigma)$ and $\Im(\sigma)$, 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 $\Re(\sigma)$ and $\Im(\sigma)$ 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 $\Re(\sigma)$ and $\Im(\sigma)$ indicates a classical "crossover" frequency of 500 MHz.

![Normalized Conductivity vs Frequency](image)

**Fig. 1** Low-field ac conductivity $\sigma_{ac}(\omega)$ 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 $\Re(\sigma)$ and $\Im(\sigma)$ initially decrease with increasing frequency. Between 4 MHz and 20 MHz, $\Re(\sigma)$ decreases nearly 70% while $\Im(\sigma)$ increases in magnitude, by approximately 25%, while remaining negative in sign. Sharp "dy" structure appears in $\Re(\sigma)$ and $\Im(\sigma)$ 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 $\Re(\sigma)$ and $\Im(\sigma)$ display similar behavior, first increasing and then decreasing smoothly with increasing frequency. For frequencies above 25 MHz, the general trend for both $\Re(\sigma)$ and $\Im(\sigma)$ is an increase with increasing frequency, with no apparent saturation in $\Re(\sigma)$ or turnover in $\Im(\sigma)$, up to 500 MHz.

![Normalized Conductivity vs Frequency](image)

**Fig. 2** Low field ac conductivity $\sigma_{ac}(\omega)$ of switching NbSe$_3$, in the sliding CDW regime. Resonances correspond to interference with narrow-band noise.

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 $\Re(\sigma)$ with increasing frequency, while earlier studies have demonstrated a differential dc conductivity independent of dc bias above threshold.

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

$$\frac{\alpha^2}{\omega} - \frac{d\theta}{dt} + \sin \theta = \epsilon_{dc} + \epsilon_{ac} \sin \omega t$$

where $\theta = (\omega_d \tau)^2$, time is measured in units of $(\omega_d \tau)^{-1}$, $\omega_d = 2\pi/\tau$, $\epsilon_{dc} = \sigma_{dc}/\tau$, $\epsilon_{ac} = \sigma_{ac}/\tau$, and $\omega = \omega/\omega_d$. We also define $\omega_d \tau = \omega_c$. In the case of zero applied dc bias and small $\epsilon$, Eq. (2) is easily solved to yield a CDW conductivity

$$\Re(\sigma) = \frac{\ne^2 \omega_c^2}{\hbar^2} \frac{\alpha^2}{(1-\beta^2)^2 + \alpha^2}$$

$$\Im(\sigma) = \frac{\ne^2 \tau}{\hbar^2} \frac{\alpha(1-\beta^2)}{(1-\beta^2)^2 + \alpha^2}$$

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 $\omega_0/2\pi = 500$ MHz, and $B = (\omega_0)^2 \times 10^{-4}$. This value of $B$ 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 the Shapiro steps observed in switching NbSe$_3$, there have been analyzed 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 = $\omega_0/2\pi$. Additional analysis of the degree of hysteresis in the dc T=0 characteristic has implied substantial CDW inertia, with $B = (\omega_0)^2 = 2.2$. These values of crossover frequency and inertial coefficient determined experimentally for the damped 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.

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. The chosen parameters are $\omega_0/2\pi = 22$ MHz and $B = 2.1$, 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 Re$\omega$ and Im$\omega$. Notable are the relative frequency independence of both Re$\omega$ and Im$\omega$ at very low frequencies, and the exceptionally strong interference effects whenever the ac test frequency matches a narrowband noise frequency (or a higher harmonic).

Figure 3 also indicates that the high frequency conductivity becomes again independent of frequency, and both Re$\omega$ and Im$\omega$ asymptotically approach zero at high frequencies. Figure 3 shows that Re$\omega$ at high frequencies attains a value substantially lower than its low frequency, $\omega \to 0$, limit.

The analog computer predictions of Fig. 3 should be compared to Fig. 2 which shows the experimental results for $\sigma$ 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 effect. Experimentally, relatively small anomalies are observed in Re$\omega$ and Im$\omega$ 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 discrepancy 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 Re$\omega$ measured at low and high frequencies. Experimentally, Re$\omega$ at high frequencies increases with increasing frequency, 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 Re$\omega$ 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 Im$\omega$ at high frequencies. Experimentally, Im$\omega$ 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 at high frequency by 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 Re$\omega$ at high frequency. We note that in this overdamped parameter range, our analog computer results agree well with perturbation theory results for Eq. (2). We also remark that our $\sigma$ results of Fig. 2 appear to rule out a recent model by von Hevesy-ger"{a}10 where switching in induced by current noise, in an otherwise overdamped classical system. In this model, Im$\omega$ 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 conductivity 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."
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 Fig. 2)\textsuperscript{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\textsuperscript{2}, in which coupled domains play a crucial role, and a model of CDW depinning\textsuperscript{10} 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 l/f noise recently observed\textsuperscript{17} in switching samples of NbSe\textsubscript{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 tunnelling model does not predict switching, an increasing conductivity with increasing frequency (even for a pinned CDW condensate) is in some ways suggestive of tunnelling processes. Hence, the possibility of macroscopic quantum tunneling here warrants further investigation.

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REFERENCES

7. To facilitate a meaningful comparison between parameters extracted from different experiments, we have used for the present measurements above 10 MHz NbSe\textsubscript{3} samples all cleaved from a larger single NbSe\textsubscript{3} crystal. The data of ref. 4 was obtained for a NbSe\textsubscript{3} sample cleaved from the same parent crystal.
8. The analog computer used is a Philipp-Gillette JA-100.
14. We remark that pseudo-inductive "ringing" has been obtained from a classical zero mass model with internal degrees of freedom, S. R. Coupersmith and P. B. Littlewood, in proceedings of ref. 1, p. 236.