Negative Differential Resistance and Instability in $NbSe_3$

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We report the observation of negative differential resistance and dynamic instability in the sliding charge-density-wave state of $NbSe_3$. The instabilities are characterized by an unusually large $1/f$ noise and intermittent chaoticlike response. We interpret our results in terms of domain coupling in the charge-density-wave condensate, resulting in hopping between distinct current-carrying states.

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The linear chain compound $NbSe_3$ displays many unusual properties associated with the formation of independent incommensurate charge-density waves (CDW's) at 144 and 59 K.[1] The first evidence for collective dynamics of the CDW condensate was the observation[2] of nonlinear dc conductivity at very low applied dc electric fields E. Non-Ohmic conduction was later shown to occur only after a well defined threshold electric field E_T is exceeded.[3] A particularly striking transport feature in the nonlinear conductivity region is that of conduction noise, consisting of well defined and stable narrow-band ac frequency components along with the low-level broadband noise.[3]

Although CDW conduction can be accounted for in a general sense by phase excitations of the condensate as first suggested by Frohlich,[4] the microscopic process of CDW depinning and conduction is still unclear. Bardeen[5] has suggested that CDW motion arises from quantum mechanical tunneling of the CDW condensate through an impurity pinning gap. This model predicts for the dc current above E_T

$$J(E)=\sigma_a E+\sigma_b(1-E_T/E)\exp(-E_0/E)E, \quad (1)$$

where σ_a is the Ohmic conductivity, E_0 is a parameter related to the pinning gap, and σ_b is the high-field limit of the excess CDW conductivity. CDW depinning has also been treated classically. Assuming a sinusoidal washboard impurity pinning potential and only a single degree of freedom for the CDW phase, Gruner, Zawadowski, and Chaikin[6] derive for the dc current above threshold

$$J(E)=\sigma_a E+\sigma_b\{1-(E_T/E)^2\}^{1/2}E, \quad (2)$$

appropriate to a voltage-driven system. Solving the same classical model in the current-driven mode yields[7] a field-current relation

$$E(J)=J/\sigma_a-E_T(1-\sigma_a/\sigma_b)\times\{(J/\sigma_a E_T)^2-1\}^{1/2}. \quad (3)$$

Equations (1)-(3) have been compared to experimental results in $NbSe_3$ and TaS_3, with Eq. (1) giving in general the best fit. The differential-resistance predictions of Eqs. (1)-(3) are drastically different. Equations (1) and (2) predict discontinuities in the always positive differential resistance at threshold, while Eq. (3) predicts an infinitely negative differential resistance at threshold.[7]

We here report measurements on the response characteristics of $NbSe_3$ near the threshold E_T in the lower CDW state. At temperatures near 40 K we observe, above threshold, regions of negative differential resistance (NDR) in the dc response. Frequency-domain analysis of the response near the NDR region shows dynamic instabilities associated with anomalously large $1/f$ noise, and temporally intermittent chaoticlike behavior. We interpret our observations in terms of dynamic CDW domain coupling, resulting in an effective hopping between multiple current-carrying states.

We have prepared crystals of $NbSe_3$ by standard vapor transport of the elements in a sealed quartz tube. Typical crystal dimensions were 1 μm $\times 5$ μm$\times 2$ mm, with minimum threshold fields $E_T\simeq 30$ mV/cm at $T=48$ K. Two-probe sample-mounting configurations were employed with conductive silver-paint contacts.

Figure 1 shows a direct current-voltage (I-V) plot for a single $NbSe_3$ crystal at $T=40$ K, obtained by driving the sample from a constant-current source. The position of the first deviation from Ohmic response, i.e., the threshold current I_T, is identified with an arrow. In a well-defined region somewhat above I_T, smoothly increasing the current I results in a smooth *decrease* in the sample voltage. The differential resistance dV/dI is negative in this region. Increasing I further into nonlinear region results in the differential resistance becoming once again positive. We note the presence of a "step" in the NDR region, in which dV/dI is close to zero over a limited current range.

Simultaneously with the dc I-V characteristics we

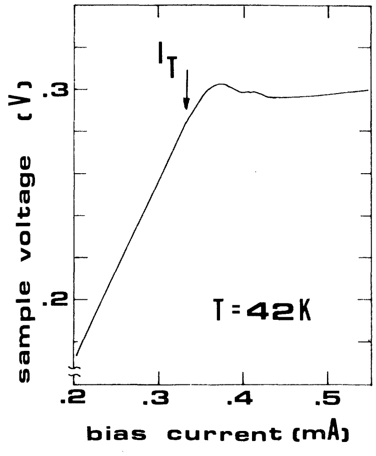

FIG. 1. I-V characteristics of NbSe$_3$. The arrow indicates the threshold current. A negative differential resistance is clearly observed near threshold.

have measured the ac voltage response spectrum with a spectrum analyzer. Immediately starting at I_T we have observed narrow-band and broadband noise, consistent with that observed in previous studies.[3] Just before the NDR region, however, a sudden onset of high-level broadband noise occurs, as illustrated in Fig. 2. The frequency distribution of the noise voltage in Fig. 2 follows a $1/f^{\alpha}$ behavior, with $\alpha = 0.5$. Hence the power response follows a $1/f$ distribution. At 1 MHz the absolute power level of the noise is approximately four orders of magnitude larger than the conventional broadband noise, and approximately two orders of magnitude larger than the narrow-band noise, observed immediately before the NDR region. Hence the noise usually associated with CDW conduction[3] is totally dominated by the $1/f$ noise, and is not observable in Fig. 2. Although the onset of high-level broadband noise corresponds to the beginning of the NDR region, the noise persists even after the current has exceeded the NDR region. However, beyond the NDR region the frequency spectrum becomes distorted and no longer follows a $1/f^{\alpha}$ behavior.

A second remarkable feature of the spectral response in the NDR region is that of temporal instability. Intermittent sharp frequency structure appears spontaneously in addition to the broadband $1/f$ noise, with a variable duration and frequency of appearance. This additional intermittent structure appears only for bias currents in the NDR region, and it occurs predominantly on the step $dV/dI \simeq 0$ observed within this region. In the step the duration of the additional frequency structure was typically 0.1 to 0.5 sec, while the frequency of appearance ranged from several hertz to approximatley 0.1 Hz. This apparently random behavior is suggestive of chaotic response. Figure 3 shows the intermittent chaoticlike voltage response in the detection frequency range 0–25 kHz, with the sample current biased to the step in the NDR region shown in Fig. 1. Figures 3(a) through 3(f) represent fast Fourier transforms of the response wave forms taken sequentially and approximately 1 sec apart in real time. All other experimental conditions for the plots are identical. The vertical scale in Fig. 3 is such that the $1/f$ noise discussed previously is largely suppressed, and thus the intermittent structure dominates the $1/f$ noise. The same dominance of the intermittent structure to the $1/f$ noise was observed in the detection frequency range 1 to 10 MHz.

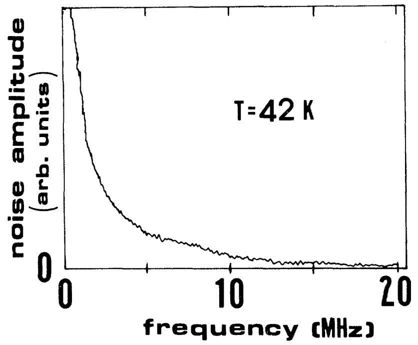

FIG. 2. Voltage response spectrum for NbSe$_3$, current biased into the NDR region. The power spectrum is $1/f$.

The features of NDR and instability described above are temperature dependent . With increasing temperature above 40 K the NDR region becomes progressively broader, and above approximately 47 K only a smooth decrease in the always positive differential resistance is observed with increasing current above I_T. $1/f$ noise and intermittency are not observed in this temperature region. With decreasing temperature below 40 K, the NDR region decreases and moves closer to I_T. At temperatures somewhat below 30 K (depending on the sample) a gradual transition of the NDR behavior into hysteretic switching, as reported previously for NbSe$_3$,[8] is observed.

A NDR region is not found in all NbSe$_3$ samples. Although we have observed NDR and the related instabilities in many different samples from different preparation batches, we have not been able to identify any clear correlation with regard to sample impurity (as determined by E_T) or sample geometry. However, all samples which display NDR at relatively high temperatures show dramatic switching at low temperatures. We also remark that

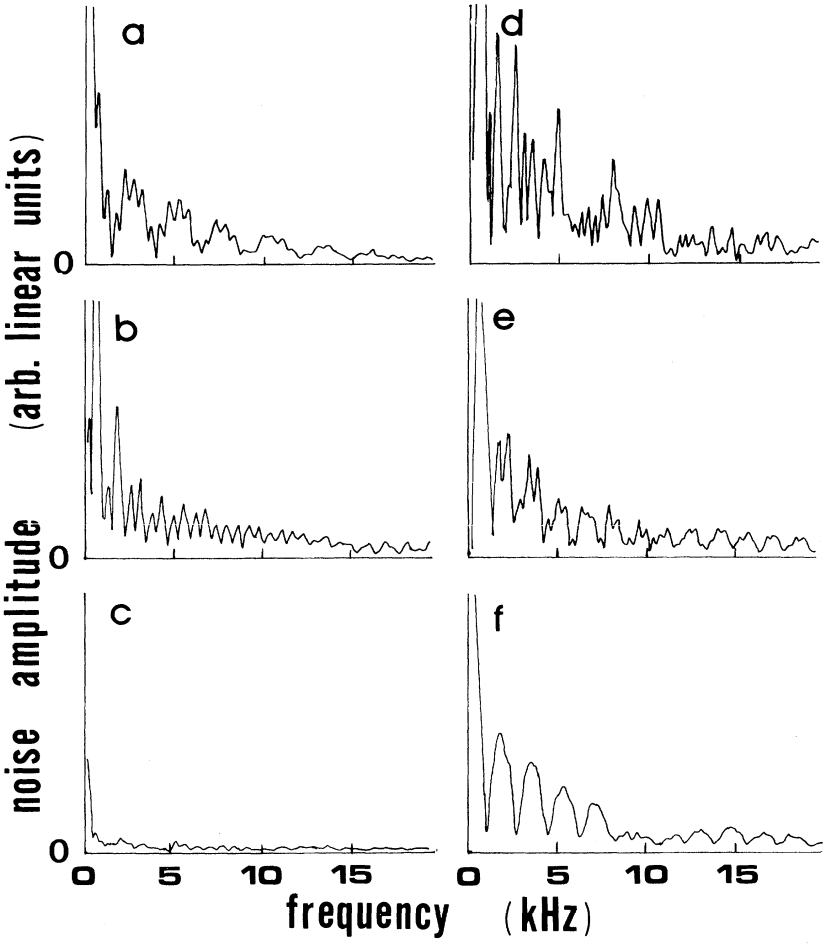

FIG. 3. Voltage response spectra for $NbSe_3$ at $T = 42$ K, current biased to the "step" in the NDR region (see text). The experimental conditions for (a)–(f) are identical; differences reflect temporal intermittency.

studies of $NbSe_3$ by Monceau, Richard, and Renard[7] have found large rms noise associated with samples which display a "knee" in I-V characteristics at threshold. However, NDR and intermittency were not observed by these authors.

As mentioned previously, Eq. (3) predicts a NDR at threshold. The associated classical single-particle model[6] also predicts switching[8] for a finite CDW inertia. However, a finite inertia is inconsistent with low-field ac conductivity studies[9] which indicate an overdamped response. The model also cannot account for broadband noise behavior unless a multidomain structure is invoked.[7] In light of these discrepancies and the strong correlation between the NDR and switching phenomena in $NbSe_3$, we proceed by applying recent concepts from the theory of switching. Joos and Murray[10] have studied the onset of CDW conduction by considering an array of CDW "domains," where conduction occurs only after a continuous channel of domains is open and conducting. This model predicts correctly the time dependences and associated probability distributions for the switching process observed in $NbSe_3$ at low temperatures.[8] We suggest that at higher temperatures the transverse correlation between CDW domains is weaker, allowing open and conducting channels to be adjacent to domains which remain pinned and nonconducting. The model must also be generalized to include a finite probability for a conducting domain to return to the pinned state.

In a current-driven experiment, the domain coupling model could lead to a NDR: Since open channels have a higher conductivity than do closed channels, an increase in current could increase the number of open channels, resulting in a decrease in the voltage across the specimen, as observed experimentally. The same model may be used to predict both the $1/f$ noise and the intermittent chaoticlike behavior. For a given current bias in the NDR region, an instability exists between having n and $n+1$ channels open. If n channels are open, the voltage across the specimen is $V_n + V_{\rm NBN} + V_{\rm BBN}$, where V_n is the dc voltage associated with a current I through n channels, and $V_{\rm NBN}$ and $V_{\rm BBN}$ represent respectively the conventional narrow-band and broadband noise associated with CDW conduction. We suppose that, at the end of an "oscillation," there exists a probability that the system hops to a state with a different number of channels open, and a complementary probability that it remains in the same state. This situation is analogous to hopping between valleys of a bistable system. Our model is, in fact, entirely equivalent to that considered by Ben-Jacob *et al.*[11] for intermittent chaos in Josephson junctions. These authors consider a junction biased to an unstable region between two stable regions where $\dot{\theta}=y_1(t)$ or $\dot{\theta}=y_2(t)$, θ being the phase difference across the junction. $\dot{\theta}$ then may hop back and forth between $y_1(t)$ and $y_2(t)$. The randomlike hopping between states leads to intermittent chaotic behavior, with a response spectrum strikingly similar to that displayed in Figs. 2 and 3. A related study of bistable systems by Arecchi and Lisi[12] again finds dramatic increases in the low-frequency noise level, consistent with a $1/f$ power law. We thus find that a domain-channel model which considers "switching" between different current-carrying states can describe at least qualitatively all our observations; namely negative differential resistance, $1/f$ noise, and intermittency. Our simultaneous observations of $1/f$ noise and intermittent response, however, suggest two distinct time scales for the characteristic attempt frequency in hopping between metastable states. The $1/f$ behavior displayed in Fig. 2 appears to be dictated by a relatively high frequency, possibly the intrinsic narrow-band noise frequency, while the intermittent behavior displayed in Fig. 3, suggests a longer time scale, of the order of 10^{-2} sec. The origin of such a long time scale is not clear. We note, however, that long time scales of similar magnitude have been reported in transient-response experiments in $\rm NbSe_3$.[13] We also remark that low-frequency oscillations, independent of the narrow-band noise, have recently been observed in the sliding CDW state of the blue bronze $\rm K_{0.3}MoO_3$.[14]

In summary, we have demonstrated negative differential resistance with associated chaoticlike instabilities in the sliding CDW state of $\rm NbSe_3$. These results are inconsistent with single-domain tunneling or single-domain rigid-particle classical models, which cannot account for negative differential resistance, switching, and overdamped ac response in a self-consistent fashion. We are able to account for our observations by considering a CDW domain structure and hopping between states of a bistable (or multistable) system. This approach is relatively independent of the actual CDW transport mechanism, and thus an adequate description of our observations should be possible within either a quantum tunneling or classical depinning framework, if domain coupling is taken into account. Finally, we note that classical theories of CDW conduction have recently been extended to include internal degrees of freedom of the CDW condensate.[15] Although the types of instabilities discussed here have not been explicitly considered, the additional degrees of freedom in such models can in principle lead to metastable states. Switching between these states, resulting in $1/f$ noise and intermittency, is then a distinct possibility.

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