Chaotic response of $NbSe_3$: Evidence for a new charge-density-wave phase

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We have measured the charge-density-wave response characteristics of $NbSe_3$ for combined ac and dc driving fields. In the switching regime a clear period-doubling route to chaos is observed, where the bifurcation cascade is periodic in applied dc bias and ac amplitude. These observations provide strong evidence for a new charge-density-wave phase which, in terms of classical dynamics, behaves underdamped.

The linear-chain compound $NbSe_3$ undergoes Peierls distortions to incommensurate charge-density-wave (CDW) states at 144 and 59 K.[1] At temperatures below the transitions, the dc conductivity is highly non-Ohmic if a well-defined threshold field E_T is exceeded.[2] The excess conductivity represents a depinning and subsequent collective motion of the CDW condensate. In $NbSe_3$, CDW dynamics have been extensively studied in the temperature region where E_T attains its minimum value, i.e., near 48 K. In this region the low-field ac conductivity suggests an overdamped response,[3] while the dc conductivity is well described by an empirical expression first suggested by Fleming:[2]

$$\sigma(E)=\sigma_a+\sigma_b \exp[-E_0/(E-E_T)] \ , \qquad (1)$$

where σ_a is the low-field (Ohmic) conductivity and σ_b and E_0 are adjustable parameters. This equation predicts a smooth increase in σ beyond E_T. Equation (1) is, however, not appropriate for all $NbSe_3$ samples. Drastic deviations are realized in crystals which display switching,[4] which occurs predominantly at temperatures below approximately 35 K in $NbSe_3$. Switching refers to a sharp hysteretic jump from the nonconducting to the conducting state, and is observed in virtually all CDW systems which display nonlinear conduction.[5] Despite implications that CDW dynamics may be radically different in the switching regime when compared with the nonswitching regime, the former state has received little attention.

We have explored the CDW response characteristics of $NbSe_3$ in the switching regime. For joint driving fields of the form $E=E_{dc}+E_{ac}\cos\omega t$, we observe dramatic interference phenomena in both the dc and ac response, characterized by hysteretic Shapiro steps and a period-doubling route to chaos. This behavior is strikingly different from that observed in the same crystal outside the switching regime, but still in the CDW state. We are able to account for the majority of our observations by applying a simple classical deterministic equation of motion for the CDW condensate, and assuming a non-negligible inertial contribution.

We have prepared high-purity $NbSe_3$ single crystals by direct reaction of the elements, and have used a two-probe sample mounting configuration with silver paint contacts. The driving source for our experiments employed a high-speed voltage follower constructed in our laboratory, with a 350-MHz bandwidth and an output impedance less than 1 Ω. The response measured was the current through the sample.

Figure 1 shows the dc current-voltage (I-V) characteristics of a $NbSe_3$ crystal at temperature $T=27$ K. With no rf applied, the dc characteristics at E_T indicate switching for this sample. The application of an rf field induces sharp Shapiro steps in the I-V characteristics of the sample. Similar steps have been observed in $NbSe_3$ at higher temperatures.[6,7] The steps represented in Fig. 1 are evenly spaced as a function of excess CDW current, and the slope of each step is equivalent to the low-field differential resistance of the sample. This indicates that the CDW drift velocity is *constant* on each step, and hence on each step the phase of the CDW appears *locked* to a harmonic of the external rf drive signal. The steps commence at the threshold, and persist in the hysteretic nonlinear region as the dc bias voltage is decreased again toward zero.

We have further investigated the stability of phase lock by viewing the current response in the frequency domain with a spectrum analyzer. The response is a complex function of dc bias, rf amplitude, and rf frequency. Figure 2 shows, for a different $NbSe_3$ sample and for fixed rf amplitude and frequency, the typical response in the Shapiro steps region as a function of dc bias voltage. Increasing dc bias leads, on each step, to a period-doubling sequence resulting in chaos, as indicated schematically in Fig. 2(a). The current response spectra for periods 1, 2, 4, and chaos are displayed in Fig. 2(b), where the vertical axis represents a voltage sig-

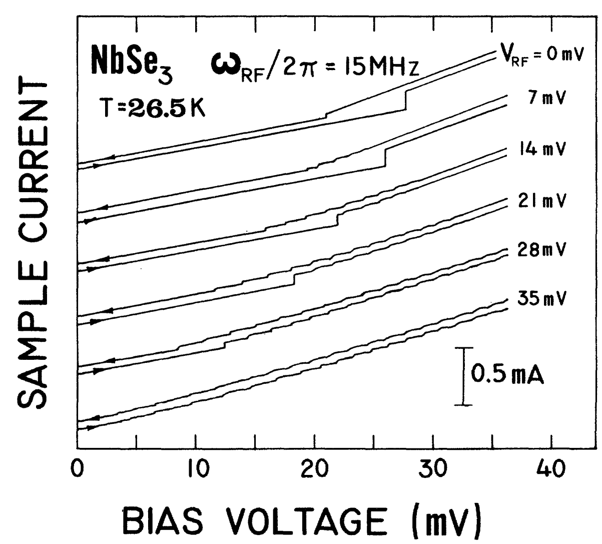

FIG. 1. I-V curves for $NbSe_3$ in the switching regime. An rf field induces hysteretic Shapiro steps.

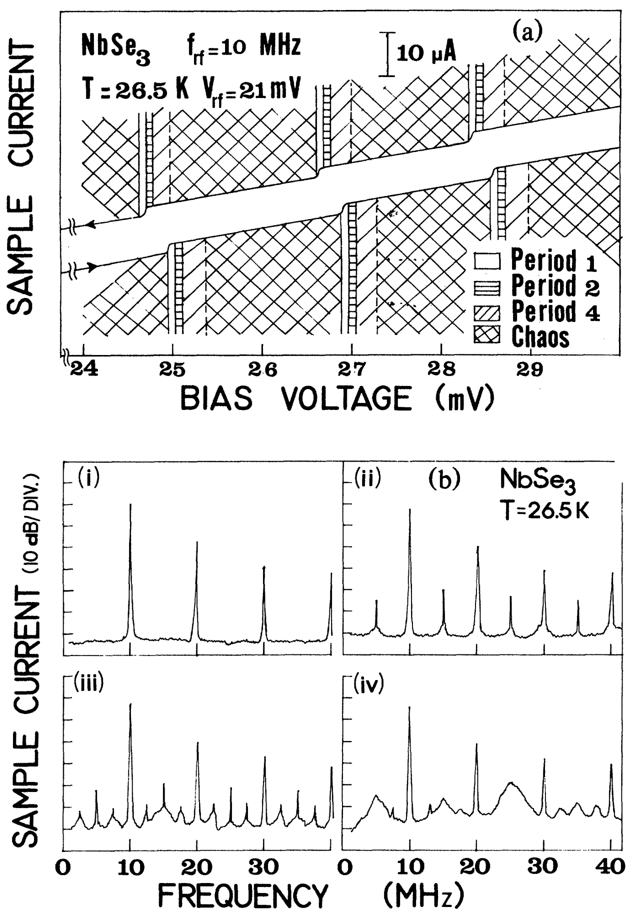

FIG. 2. (a) Schematic representation of current response in Shapiro step region, for forward- and reverse-bias voltage sweeps. (b) Frequency spectrum of current response in Shapiro step region. External rf drive frequency and amplitude as in (a). (i) $V_{dc}=25$ mV, period 1; (ii) $V_{dc}=25.1$ mV, period 2; (iii) $V_{dc}=25.2$ mV, period 4; (iv) $V_{dc}=25.5$ mV, chaos.

nal directly proportional to sample current. Chaotic response always occurs near the end of a particular step, and an increase in dc bias voltage results in a jump to the next highest step, where the bifurcation cascade resumes. The pattern of bifurcations and chaos repeats for many cycles, finally disappearing at high values of dc bias. A similar map is obtained for a decreasing bias voltage sweep. The only difference is a slight shift in the location of each chaotic or periodic region, reflecting the hysteresis[4] of the dc I-V characteristics.

For fixed rf frequency and dc bias above threshold, the repetitive period-doubling route to chaos can also be achieved by smoothly increasing rf amplitude. The resulting sequence is qualitatively similar to that observed for increasing dc bias, demonstrated in Fig. 2(a). The bifurcation cascade was also found to occur over a large rf frequency range, from approximately 0.5 to over 50 MHz. However, with increasing rf frequency, larger rf amplitudes are required to achieve bifurcations and chaos. Increasing temperature in the switching regime leads to a decrease in the hysteresis loop and a smearing of the sharp switch. Associated with these effects are a smearing of the leading and trailing edges of the Shapiro steps, and a gradual disappearance of the period-doubling and chaotic response. Above approximately 40 K period doubling and chaos were not observed for any $NbSe_3$ sample, and in this temperature region the Shapiro step structure was consistent with that reported previously for $NbSe_3$ near 42 K.[7]

In analyzing these results, we first note that our observations do not reflect the trivial response of driving the sample through the dc switch by application of the rf field. This would not lead to the observed dependence on dc bias voltage. We interpret our results as representing an example of universal instability in phase locking for a driven nonlinear system characterized by an intrinsic limit cycle. If a system with intrinsic frequency ν_1 is externally perturbed at frequency ν_2, mode locking may occur for $\nu_1/\nu_2=m/p$, with m and p integers. Instability in phase lock and the transition to chaos in such systems has been investigated in terms of the circle map by various authors.[8] The map of the circle onto itself may be represented as

$$\theta_{i+1}=\theta_i+\Gamma+C\sin 2\pi\theta_i \ , \qquad (2)$$

where $\Gamma=\nu_1/\nu_2$ in the absence of locking, and C describes the strength of the coupling between the external perturbation and the system. In the parameter range $C>1/2\pi$, the circle map displays stable mode-locked solutions, which bifurcate successively to chaos as Γ is smoothly increased. Since θ is a modulo 1 variable, changing Γ to $\Gamma+n$, with n an integer, will not change Eq. (2). Thus the pattern of bifurcations to chaos will repeat itself as Γ is increased monotonically.

We associate the steps in Fig. 1 with locked states of the circle map. The parameter ν_1 is associated here with the CDW narrow-band noise frequency, and hence Γ increases with increasing dc bias. The periodicity of the bifurcation sequence in dc bias is thus consistent with the periodicity of the behavior predicted by the circle map. We note that the circle map has recently been applied to investigate the devil's staircase in $NbSe_3$ outside the switching regime.[9]

To consider explicitly the parameters associated with the chaotic response, we assume for the CDW an equation of motion[10]

$$\frac{d^2x}{dt^2}+\frac{1}{\tau}\frac{dx}{dt}+\frac{\omega_0^2}{2\pi/\lambda}\sin(2\pi x/\lambda)=\frac{e}{m^*}E \ , \qquad (3)$$

where x is the CDW displacement, $1/\tau=\gamma/m^*$ with γ a damping constant and m^* the effective mass of electrons condensed in the CDW state, $\omega_0^2=k/m^*$ with k a restoring force constant, and λ is the CDW wavelength. The connection between the return map of Eq. (3) and the circle map has recently been demonstrated by numerical integration.[11] In dimensionless form Eq. (3) becomes

$$\beta\frac{d^2\theta}{dt^2}+\frac{d\theta}{dt}+\sin\theta=e_{dc}+e_{ac}\sin\Omega t \ , \qquad (4)$$

where $\theta=2\pi x/\lambda$, $e_{dc}=E_{dc}/E_T$ with $E_T=m^*\omega_0^2\lambda/2\pi e$, $e_{ac}=E_{ac}/E_T$, $\Omega=\omega/\omega_0^2\tau$, $\beta=\omega_0^2\tau^2$, and time t is measured in units of $(\omega_0^2\tau)^{-1}$. We also define $\omega_c=\omega_0^2\tau$ and $Q=\beta^{1/2}$. Equation (4) is well known to display chaotic solutions, *if* the inertial term is retained.[12] Since the CDW response in $NbSe_3$ outside the switching regime is, in general, assumed overdamped, chaotic behavior is not expected unless the damping is in some manner reduced, for example, by the addition of an external inductance.[13] However, a switching behavior at threshold is in itself suggestive of an intrinsic finite inertial contribution to the CDW response. In the context of Eq. (4), we may determine the effective inertia from the magnitude of the hysteresis loop in the dc I-V characteristics. We have solved Eq. (4) by numerical integration on a digital computer.[14] For $Q < 2\pi/10$ the CDW motion is strongly damped and no switching occurs, while for $Q > 2\pi/10$ switching appears. The onset of hysteresis occurs at approximately $Q = 1$. We define a hysteresis parameter $H = (V_{c1} - V_{c2})/V_{c1}$, where V_{c1} and V_{c2} are the respective dc thresholds for increasing and decreasing voltage drives. Figure 3 displays the calulated results of H vs Q relevant to Eq. (4). From the top trace in Fig. 1 we find an experimental value $H = 0.24$, which implies $Q = 1.5$ for the $NbSe_3$ system in the switching regime. ω_c may be determined directly from the magnitudes of the Shapiro steps. The first Shapiro step magnitude is given by[7] $\delta V = V_T|J_1 \times (e_{ac}/\Omega)|$, where $J_1(z)$ is the first-order Bessel function, for which the first maximum occurs at $z = 1.84$. Extending the data of Fig. 1 to higher values of rf amplitude, we find that the maximum in the Shapiro step occurs at $V_{ac} = 35$ mV. Using $V_T = 28$ mV and $\omega/2\pi = 15$ MHz, we find $\omega_c/2\pi = 22$ MHz. The above values of Q and ω_c reduce to $\tau^{-1} = 61$ MHz and $\omega_0/2\pi = 15$ MHz. These values of ω_0 and τ^{-1} are orders of magnitude smaller than those determined for the same crystal at slightly higher temperature, outside the switching regime. In other words, in the switching regime, and for dc bias values exceeding threshold, the CDW response appears *underdamped.*

A detailed analysis[15] of Eq. (4) in the underdamped limit with $\beta = 25$ has indeed demonstrated Shapiro steps with a period-doubling route to chaos on particular steps. The results are not critically dependent on the value of β, as long as the response remains underdamped. However, Eq. (4) predicts that instabilities in phase lock occur predominantly for driving frequencies in the range $\beta^{-1} < \Omega < \beta^{-1/2}$, which would imply a frequency range of approximately 1 MHz for the $NbSe_3$ system. As noted earlier, however, we observe in $NbSe_3$ phase-lock instabilities over a much wider frequency range.

It is nevertheless clear that Eq. (3), with inertial terms included, gives to first order an excellent account of our experimental findings. It is interesting to speculate on the origin of the inertial term. This could arise if the CDW damping parameter were velocity dependent at lower temperatures, or if the coupling of various CDW domains resulted in an effective retarded phase feedback mechanism.[4] These possibilities could be explored through detailed studies of the ac conductivity in the switching regime. Our results presented here demonstrate that the CDW response in the switching regime is dramatically different from that obtained at higher temperatures. These findings, together with earlier demonstrations[16] that the dc threshold field in the switching region is temperature independent, suggest an entirely new CDW phase in the switching regime. Finally, we remark that similar period-doubling routes to chaos might be expected in the switching regimes of other sliding CDW systems.

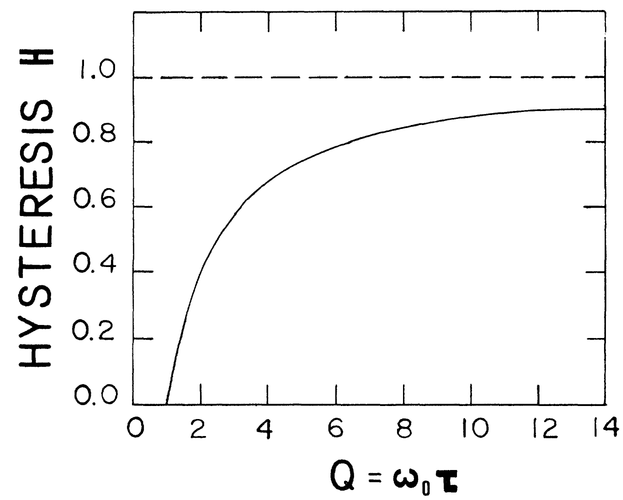

FIG. 3. Hysteresis parameter H vs Q calculated for Eq. (4) (see text).

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