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Routes to Chaos in Charge Density Wave Systems

The electron–hole condensate, called the charge density wave (CDW) displays a broad variety of nonlinear and frequency dependent response phenomena when subjected to dc and ac electric fields. Various routes to chaos have been identified in various materials where a CDW ground state develops. The experiments may help to clarify issues concerning this novel type of conduction mechanism, and also point to important differences between predictions based on simple equations of motions and observations made on real systems.

1. INTRODUCTION

The deterministic response of driven dissipative systems, where the dynamics are governed by either single or multiple degrees of freedom, has recently been extensively explored by various theoretical methods.[1] For certain ranges of system and drive parameters, the dynamical response may become chaotic, and various "universal" routes to chaos have been established by analytical methods, numerical simulations, and analog computer calculations. Of particular interest are solutions to nonlinear equations which describe the (idealized) behavior of certain real physical systems.

Chaotic behavior has been firmly established experimentally in only a few condensed matter systems. These include Josephson junctions,[2] semiconductor junctions[3] and plasmas,[4] photoconductors,[5] and also systems with nonlinear optical properties.[6] The purpose of this Comment is to point out that various routes to chaos have been clearly established in driven charge density waves.

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The "charge density wave" (CDW) refers to a collective electronic mode, observed in a variety of low dimensional conductors, which can display unique nonlinear and frequency dependent response characteristics when subjected to applied dc and/or ac electric fields.[7] Much of the CDW dynamics can be modelled by retaining only a single degree of freedom (describing the CDW phase relative to the crystal lattice), and a resulting equation of motion is formally identical to that used widely to describe other, well established nonlinear systems, with a rich field of chaotic solutions. Detailed experimental and theoretical work, however, makes it more and more apparent that internal degrees of freedom play an important role in the dynamics of the CDW collective mode. These additional degrees of freedom in fact appear to often dictate fully the development of the chaotic response.

2. DRIVEN CHARGE DENSITY WAVES

In various solids with highly anisotropic band structure, electron–phonon interactions lead to a CDW phase transition. The CDW ground state is characterized by a gap in the single particle excitation spectrum and by a collective mode of carriers condensed below the gap. The CDW transition is driven by electron–hole pairing across opposite sides of the one-dimensional (1D) Fermi surface, resulting in a periodically modulated charge density

$$\rho = \rho_0 + A \cos\varphi(x,t) \quad (1)$$

where A and φ are, respectively, the amplitude and phase of the collective mode. The period of the charge modulation (and similarly the period of the modulation of ions in the underlying lattice) is given by $\lambda = \pi/k_F$. λ may thus be completely unrelated to the original lattice constant of the undistorted lattice, and in this case the CDW is termed incommensurate. As in a superconductor, the phase φ plays an important role in the dynamics of the collective mode. The averaged CDW carrier concentration n and electric current density j carried by the CDW condensate are given by the spatial and time derivatives of the phase,

$$n(x,t) = (1/\pi)\partial\varphi/\partial x; \quad j(x,t) = (-e/\pi)\partial\varphi/\partial t \quad (2)$$

The phase of the CDW may be expressed as

$$\varphi(x,t) = 2k_F x + \phi(x,t) \quad (3)$$

where ϕ describes phase fluctuations of the CDW. If k_F is assumed position and time independent, all CDW dynamics are contained in ϕ. Low-lying phase excitations of the CDW may then be obtained from the Lagrangian, which in one dimension is given by[8]

$$\mathscr{L}_\phi = n\{m^*(\partial\phi/\partial t)^2/8k_F^2 - \kappa(\partial\phi/\partial x)^2/2\} \quad (4)$$

where m^* and κ are the effective mass and elastic constant of the CDW, respectively. Both parameters may be derived using an appropriate electron–phonon Hamiltonian, in terms of the fundamental parameters such as the bandwidth, phonon frequency, and electron–phonon coupling constant. Wave-like excitations of ϕ of the form $e^{i(qx-\omega t)}$ in Eq. (4) are gapless, and satisfy the linear dispersion relation $\omega^2(q) = \kappa(2k_Fq)^2/m^* = m(qv_F)^2/m^*$, where v_F is the Fermi velocity.[9] This has important consequences. The CDW phase can locally distort around various pinning potentials $V(r)$, and such phase fluctuations lead to pinning of the entire collective mode to the underlying lattice. The pinned CDW mode carries no dc current for small applied electric fields, but strong ac polarization effects occur at relatively low frequency.

The existence of charge density waves has been firmly established in a variety of low dimensional solids. Several inorganic "linear chain" compounds such as $NbSe_3$, TaS_3, and $(TaSe_4)_2I$, undergo second-order phase transitions below room temperature. Structural studies indicate the transitions are associated with the development of incommensurate CDW's with period $\lambda = \pi/k_F$. Below the transition, the low-field dc electrical conductivity σ_{dc} of the crystals is governed by the single particle gap Δ, induced at the Fermi energy by CDW formation. However, when measured along the chain direction (along which the CDW's develop), the low field ac conductivity σ_{ac} is strongly frequency dependent, even in the spectral range well below the single-particle gap energy $\hbar\omega_{ex} = \Delta$. In addition, σ_{dc} is strongly nonlinear when the applied dc electric field exceeds a relatively small threshold field E_T, which for pure samples is on the order of tens of mV/cm. In the nonlinear conductivity regime, the excess current carried by the CDW contains an oscillating component, with fundamental frequency f_0 directly proportional to the time averaged CDW current,

$$\langle j(t) \rangle = (\text{const})ef_0 \qquad (5)$$

The above features can all be tentatively ascribed to collective dynamics of the CDW condensate. An incommensurate CDW is translationally invariant only in the absence of impurities, etc. Such defects break the translational invariance and lead to pinning of the CDW phase. The low frequency ac conductivity represents oscillations of the pinned CDW about pinning centers, while the nonlinear dc conductivity and related current oscillations correspond to continuous translation of the entire condensate as originally envisioned by Fröhlich.[10] The "sliding" motion of the CDW is hence often referred to as "Fröhlich conduction."

A highly oversimplified model,[11] which has been widely used to account for the above experimental findings, assumes that local distortions of the phase around pinning centers lead to an energy minimum configuration, which is identically recovered by translation of the entire CDW by one wavelength λ. The associated "pinning potential" $V(\phi)$ is then a strictly periodic function with period 2π. With additional internal degrees of freedom for the CDW condensate neglected, the equation of motion for the CDW (assuming a sinusoidal pinning potential) is

$$d^2\chi/dt^2 + (1/\tau)d\chi/dt + \omega_0^2\sin(2k_F\chi)/2k_F = eE/m^* \qquad (6)$$

where χ refers to the CDW position, τ is a phenomenological damping constant, ω_0 is the intrinsic oscillation frequency of the CDW, and $E = E_{dc} + E_{ac}\cos(\omega_{ex}t)$ represents the externally applied electric field. For finite but small ac fields, and in the absence of dc fields, Eq. (6) predicts an ac response identical to that for a simple damped harmonic oscillator. A fit of the predicted response to that measured over a wide frequency range for orthorhombic TaS_3 (o-TaS_3) is shown in Fig. 1. The figure illustrates two points, both important to chaotic behavior to be discussed shortly. First, the best fit to the single harmonic oscillator response, shown as a solid line in Fig. 1, indicates an *overdamped* response. From the fit one extracts $\omega_0\tau = 0.03$, much less than one, and detailed

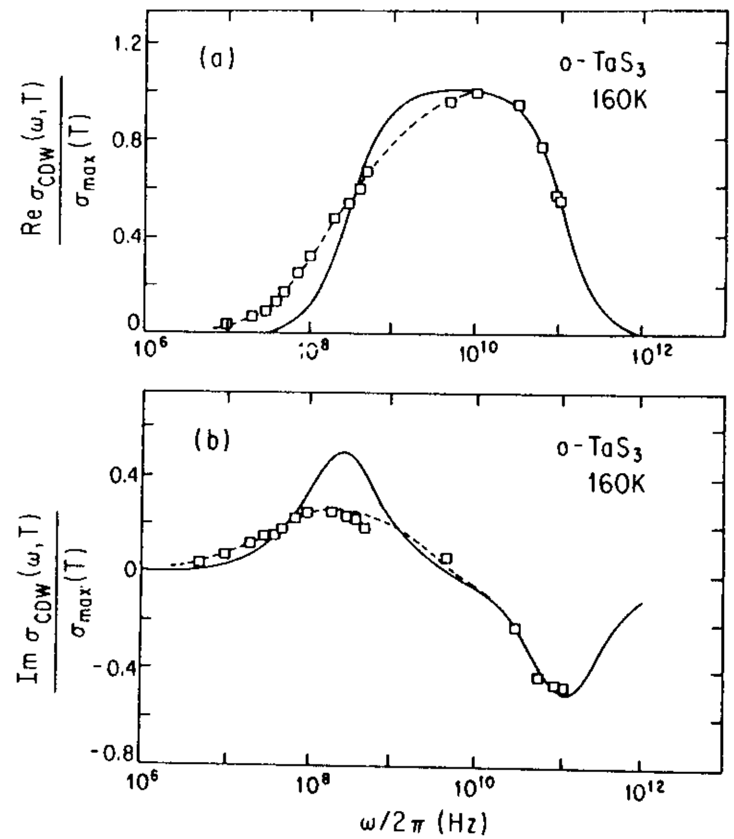

FIGURE 1 Complex frequency dependent conductivity in o-TaS_3. The full line is a fit to a harmonic oscillator expression; the dotted line includes a broad distribution of relaxation times (from Ref. 13).

experiments in the radiofrequency spectral range are again consistent only with overdamped response. Second, substantial deviations from the single harmonic oscillator response are apparent at low frequencies. In this frequency range improved fits are provided by alternative descriptions based on macroscopic quantum tunneling mechanisms,[12] or classical harmonic oscillators with a distribution of pinning frequencies.[13] The dotted line in Fig. 1 has been calculated with a broad distribution in pinning frequencies. Although the assumption of parallel relaxation processes is most probably not entirely appropriate, such a fit does point to the importance of disorder, and additional contributions from the dynamics of the internal deformations of the collective mode. Such internal CDW dynamics have been studied extensively through relaxation effects following the end of electrical current and thermal pulse excitations and remnant polarizations.[14-17] Broadly speaking, the observed phenomena are reminiscent of those observed in glasses and spin glasses.

Equation (6) also leads to a sharp dc threshold field for the onset of dc nonlinear conduction, and a saturation of the dc conductivity in the limit $E_{dc} \to \infty$. These features are in qualitative agreement with experimental results, but the overall nonlinearities observed experimentally are less sharp, in particular near threshold. Again, descriptions which invoke tunneling mechanisms or include internal degrees of freedom[18] give results in improved agreement with experiment.

In dimensionless form, Eq. (6) reads

$$d^2\theta/dt^2 + \Gamma d\theta/dt + \sin\theta = E/E_T \quad (7)$$

where $\theta = 2\pi\chi(t)/\lambda$ may be identified with the time dependent part of $\phi(x,t)$, $\Gamma = (\omega_0\tau)^{-1}$, $E_T = \lambda m^*\omega_0^2/2e$, and time t is measured in units of ω_0^{-1}. This equation of motion is identical to that of a physical pendulum in a gravitational field, or a rigid particle moving in a "washboard" potential. Equation (7) is also well known in the Josephson junction literature,[19] where it describes the dynamics of a Josephson junction in the resistively shunted junction (RSJ) model. In the later case θ represents the superconductor phase difference across the junction, $\Gamma = (\omega_J RC)^{-1}$ with ω_J the Josephson plasma frequency, and R and C, respectively, the resistance and capacitance of the junction. Also, if Eq. (7) is used to describe Josephson junctions, E refers to the current through the junction, and E_T to the critical current of the junction. With this analogy, the current oscillations described by Eq. (5) are analogous to the ac Josephson effect, with the generated frequency linear in time averaged junction voltage. The value of the constant in Eq. (5) then is identically 2. For CDW systems, the constant is evaluated with $\langle j(t)\rangle$ representing the current on each conducting chain, and is found to be between 1 and 2.

The analogy between the RSJ model and CDW systems has been used extensively to describe (and predict) various types of interference effects in CDW systems when both dc and ac electric fields are applied. Ac field-induced steps (analogous to Shapiro steps in Josephson junctions) have been observed[20] in the dc current voltage (I-V) characteristics of CDW systems, occurring whenever $2\pi f_0 = p\omega_{ex}$, with p an integer. These steps have been analyzed in detail in terms of solutions derived originally for Josephson junctions. Related dc field-induced interference structure in the ac dielectric constant and ac conductivity has also been reported.[21,22] In general, the interference effects in CDW systems are less sharp than those associated with Josephson devices, and it is generally agreed that incoherent effects associated with internal CDW dynamics are responsible for such differences.

Internal degrees of freedom are also believed to be the source of other experimental observations in CDW systems, which have strong implications in terms of chaotic dynamics to be discussed shortly. Lissajous curves,[23] recorded in the presence of large amplitude ac drive fields, have distinctive characteristics in the radiofrequency spectral range, in clear contrast to the overdamped (noninductive) response expected on the basis of the observed small signal ac response. Similarly, the CDW voltage response resulting from large amplitude current pulses is often found to display "ringing" behavior,[17] again suggestive of inertial dynamics.

A most important observation is that, in certain CDW crystals, the onset of the current carrying CDW state (i.e., the dc threshold for nonlinearity) is associated with well-defined switching and hysteresis effects. These phenomena are observed in virtually all materials which display CDW conduction, although not in all crystals of a given material. Crystals which display switching and hysteresis also display a rich spectrum of chaotic response properties. Recent work[24] has shown that switching and hysteresis are related to the dynamics of coupled macroscopic domains within the crystal, where the domains are separated by spatially localized phase slip centers. The location and volume of the respective CDW domains appear to be intimately related to the distribution of (strong) impurity pinning sites, and internal CDW phase (and amplitude) dynamics are expected to play an important role. On the other hand, switching and hysteresis also follow directly from Eq. (7), *if* the CDW response is assumed underdamped and the inertial (acceleration) term in Eq. (7) is retained. Switching and hysteresis then obtains[25] for $\omega_0\tau > 1$, with associated chaotic response.

We thus find evidence that the nonlinear response of the CDW condensate is, for small ac excitations only, best described in terms of overdamped dynamics, while for certain parameter ranges inertial dynamics appear important.

3. ROUTES TO CHAOS

The inherent complexity of many different physical systems is well contained in the often relatively simple nonlinear differential equations which describe, either exactly or approximately, the respective system. It has been appreciated for some time that the simple physical pendulum and resistively shunted Josephson junction display chaotic dynamics, which are reproduced by deterministic solutions of Eq. (7). Such chaotic solutions have been extensively investigated by use of analog computers, numerical simulations, various analytical methods, and by simple iterations of a corresponding "map."[2,26–29] While the overall behavior can be, for an arbitrary set of parameters, exceedingly complicated, several routes to chaos can be clearly identified. Period doubling bifurcations for pure ac drive, devil's staircase behavior in the nonlinear region with applied ac and dc fields, and intermittency associated with transitions between various states are perhaps the most well established. All of these have been searched for, and in some cases established, by studying the response of charge density waves to dc and ac electric fields. The mere observation of chaotic response in overdamped (as suggested by the low field ω_{ex}-dependent conductivity) CDW conductors raises several important questions, since for an overdamped system described by Eq. (7), no chaotic behavior is expected.

Period doubling bifurcations in the presence of ac drive fields alone are the consequence of the anharmonic (sinusoidal) restoring force in Eq. (6). The force is derived from an impurity pinning potential $V(\phi) = 1 - \cos\phi$ which, to leading orders in the displacement, is given by

$$V(\phi) = \phi^2/2 - \phi^4/24. \tag{8}$$

Analog computer solutions[30] of Eq. (7) with the above potential lead to period doubling routes to chaos when the ac drive amplitude is increased. The chaotic response is signalled by a dramatic increase of the low frequency broad band noise; the critical field E_{ac}(crit) where this occurs depends on the drive frequency. Similar behavior has been demonstrated[31] in the CDW conductor $(TaSe_4)_2I$, where the characteristic frequency ω_0 was dramatically reduced (from a few tens of GHz to the MHz range) by applying a series inductance. This also introduces a finite inertia term into the nonlinear equation describing the CDW phase. While period doubling bifurcations have not been detected, a sudden increase of the broad band noise with increased E_{ac} gives clear evidence for a transition to chaos in this system. By using the appearance of broad band noise as the criterion, the periodic–chaotic response boundary can be mapped out. This is shown in Fig. 2, with ω_0 (characteristic of

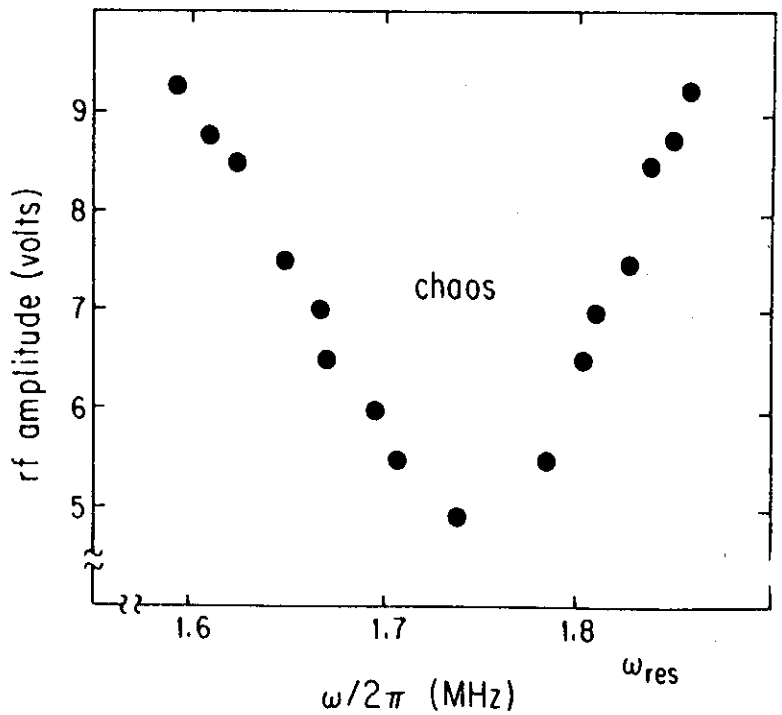

FIGURE 2 Periodic-chaotic response boundary for $(TaSe_4)_2I$ circuit (from Ref. 31).

the $(TaSe_4)_2I$-inductor system) also indicated by the arrow. The periodic–chaotic boundary observed here is qualitatively very similar to that established by analog computer solutions of Eq. (7).

While the arrangement used in the $(TaSe_4)_2I$ study mimics an underdamped response—and consequently the chaotic behavior found is not entirely surprising—other consequences of the anharmonic potential as given by Eq. (7) have not been established by experiment. In particular, such a potential should lead to an increased low frequency dielectric constant ϵ in the presence of applied dc fields, since for a sinusoidal restoring force $\epsilon(\omega_{ex} \rightarrow 0)$ diverges as $E_{dc} \rightarrow E_T$ from below. The measured ϵ in $(TaSe_4)_2I$, however, has not borne out this divergence, at least for ω_{ex} near 1 MHz. The most likely explanation for this is that an applied dc field leads to transitions between various metastable configurations which are due to random pinning forces.[32] While such behavior could leave the very low field ac response unaffected for $E_{dc} < E_T$, the broad distribution of relevant energies would suggest a highly nonlinear response as ac field energy (i.e., ac amplitude) is increased. A nonsinusoidal response to an excitation of the form $E_{ac}\cos(\omega_{ex}t)$ is the natural consequence of this "glassy" behavior. This also leads to higher harmonics in the frequency response spectrum.[33] Whether period doubling bifurcations can be the consequence remains to be seen; no such mechanism has been suggested to date.

A different route to chaos for CDW's is expected in the current carrying, nonlinear conductivity region, when both dc and ac electric fields are applied. This can be viewed as a consequence of two competing periodicities in the driven system, one given by the current oscillations, with frequency described by Eq. (5), the other given by the frequency of the applied ac field ω_{ex}. Because the spectrum of current oscillations contains many harmonics, and also because of the intrinsic nonlinearity, interference structure in the dc response spectrum may occur not only for $2\pi f_0 = \omega_{ex}$, but in general whenever

$$2\pi f_0 = (p/s)\omega_{ex}, \qquad (9)$$

with p and s integers.[29] Such interference structure is commonly observed[19,34] in dc biased Josephson junctions in the presence of

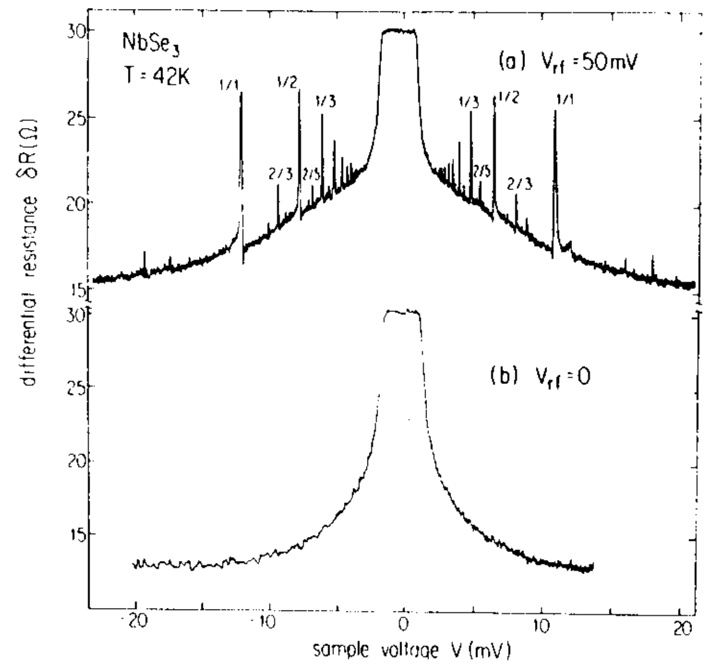

FIGURE 3 Differential resistance vs. sample voltage in $NbSe_3$ (a) with and (b) without applied rf voltage at 25 MHz. The peaks correspond to steps in the direct I-V curve. A few peaks are identified by p/s (from Ref. 36).

microwave radiation, and analogous "Shapiro step" behavior has been observed[20,35–37] in $NbSe_3$ and o-TaS_3 by examining in detail the differential resistance dV/dI in the presence of radiofrequency radiation. Steps in the direct I-V curves correspond to peaks in the differential resistance. Figure 3 shows the differential resistance observed in $NbSe_3$ both in the absence and in the presence of an applied ac field. A spectacular array of peaks is observed in the presence of an ac drive; the peak corresponding to $p = 1$ and $s = 1$, labeled as 1/1, is the usual (fundamental) Shapiro step; peaks closer to the origin ($V = 0$) labeled according to p/s are called the subharmonic steps.

Modelling ac-dc interference structure as arising from interaction of resonances, the Shapiro step behavior can be studied in detail by constructing a return map which describes the time evolution of the CDW phase, sampled at discrete times $t_m = 2\pi m/\omega_{ex}$. Assuming that $\dot{\theta}_m$ is determined only by θ_m, due to dissipation, a "circle map"

$$\theta_{m+1} = \theta_m + \Omega + K\sin(2\pi\theta_m)/2\pi \qquad (10)$$

results, where $\Omega = 2\pi f_0/\omega_{ex}$, and K describes the strength of the coupling between the system and the external perturbation—and is consequently assumed proportional to E_{ac}. The circle map is the return map appropriate to Eq. (8), as demonstrated by detailed numerical integrations.[29,38] For finite coupling K, mode locking occurs for certain values of ω_{ex} and f_0, i.e., f_0 remains fixed at $(p/s)\omega_{ex}$ over a finite range of E_{dc}. From Eq. (4), the time averaged CDW current (and hence $\langle\partial\phi/\partial t\rangle$) is then dictated strictly by ω_{ex}. The mode locked regions are separated by regions corresponding to free running solutions of the phase.

Figure 4 shows mode locked and free running solutions for the circle map in K-Ω parameter space. With increasing K, the mode

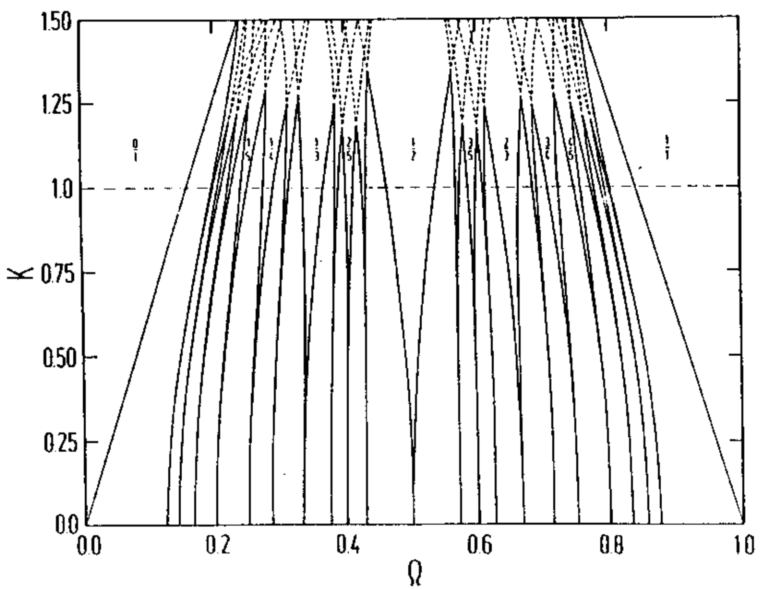

FIGURE 4 Mode locked and free running solutions of the sine circle map. Some of the locked regions are identified by the corresponding p/s value. Above the critical line at $K = 1$, resonances overlap (dotted lines) (from Ref. 29).

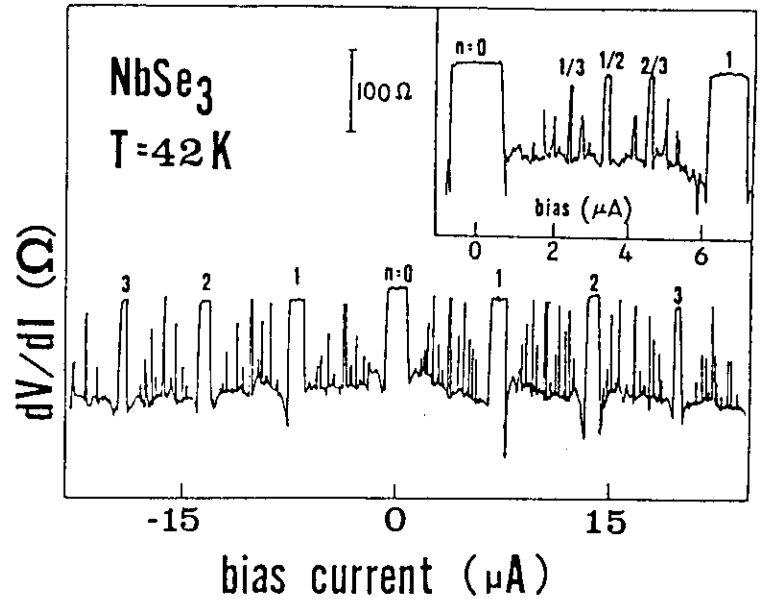

FIGURE 5 Mode locked Shapiro steps in $NbSe_3$. Over the mode locked region, dV/dI is independent of dc bias. The inset shows the subharmonic structure in detail, with corresponding p/s values (from Ref. 35).

locked regions grow in size, and at $K = 1$ a critical line forms where mode locking occurs for every f_0; i.e., free running solutions no longer exist (for clarity, not all mode locked solutions are displayed in Fig. 4). At $K = 1$, the "devil's staircase" formed by the mode locked solutions is complete. The complementary Cantor set has dimension $d = 1$ for $K < 1$, and fractal dimension $d = 0.87$ at $K = 1$.[39] For $K > 1$, overlapping of resonances occurs, and chaos is possible.

The interference structure displayed in Fig. 3 for $NbSe_3$ can be associated with a precursor to system mode locking. Indeed, for substantially greater E_{ac}, mode locking over well-defined ranges of E_{dc} does occur, as shown in Fig. 5. The subharmonic structure, as detailed in the inset to Fig. 5, indicates a smaller interval of mode locking for higher order subharmonics, in agreement with Eq. (10).

The extent of mode lock, i.e., the completeness of the devil's staircase, can in principal be determined directly from the subharmonic interference structure. Instrumental noise sets a lower limit to the size of the step which can be observed experimentally, and consequently the completeness of the staircase cannot be confirmed directly. The completeness of the staircase can, however, be examined by the following test as well: choosing a discrimination lever r, one adds up the total width $S(r)$ of steps wider than r. If $N(r) = [1 - S(r)]/r$ in an interval of unit length, then rN is the fraction of interval unoccupied by steps larger than r. For a complete staircase, $rN \rightarrow 0$ as $r \rightarrow 0$, and

$$N(r) = r^{-d} \qquad (11)$$

with d defining the fractal dimension of the complementary Cantor set. As mentioned previously, the circle map, Eq. (10), leads to $d = 0.87$ for $K = 1$. This universal behavior has been confirmed by analog computer simulations and by studying a two-dimensional dissipative map.[29,40] An analysis as described above, for experiments performed on $NbSe_3$,[36] is shown in Fig. 6. Here a fractal dimension $d = 0.91$ is obtained, in striking agreement with theory.

There are, however, several serious problems with such an interpretation. First, one expects, on the basis of the circle map and corresponding simulations of Eq. (7), that the value of d depends critically on E_{ac} and on the selected range of p/s. Such behavior has not been observed in $NbSe_3$, and indeed a critical line cannot be clearly identified. Equally important, subharmonic steps are predicted by Eq. (7) only in the underdamped limit, and therefore should not exist in an overdamped system as determined from the frequency dependence of the low field ac response. Furthermore, in the underdamped limit where Eq. (7) does predict subharmonic interference steps, chaos is also predicted on mode locked steps. In contrast to this, for the samples displayed in Figs. 3 and 5, chaos is *not* observed for any combination of ac and dc drive parameters. In the language of the circle map, the fractal dimension $d \approx 0.91$ indicates that the system is at criticality ($K = 1$), yet the absence of chaos suggests $K = 1$ cannot be exceeded, irrespective of the magnitude of E_{ac}.

It has been suggested that subharmonic structure in CDW systems is the consequence of the dynamics of internal modes. These

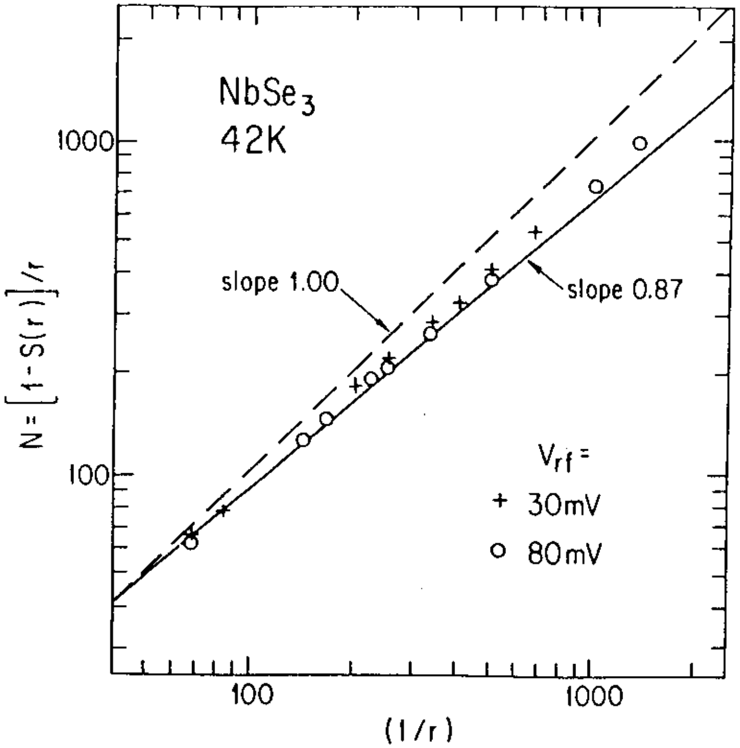

FIGURE 6 $N(r)$ vs. $(1/r)$ for Shapiro step subharmonic structure in $NbSe_3$. The fractal dimension is $d = 0.91$, close to $d = 0.87$ predicted by the circle map at $K = 1$ (from Ref. 36).

can be modelled by assuming that the system is broken, under the influence of pinning, into domains. Computer simulations[41] of a set of coupled domains leads to subharmonic mode locking structure with striking resemblance to the devil's staircase behavior, suggesting that, even if inertial terms are neglected, internal degrees of freedom may lead to frequency locking given by Eq. (8). Alternatively, local distortions around random impurity pinning centers can be considered by including a gradient term $\nabla\phi(x,t)$, as suggested by the Lagrangian, Eq. (4). Perturbation treatment of these local deformations leads to anomalies in the differential resistance, corresponding to Eq. (9) in the $2s$ order of perturbation theory.[42] While these results appear to point in the right direction, their direct relevance to observations of a fractal dimension remains to be seen. Also, theoretically the absence of subharmonics is confirmed only for a sinusoidal potential; for a nonsinusoidal potential a rich subharmonic structure can be obtained.[43] Whether models which assume particular forms for the pinning potential lead to fractal dimensions, however, remains to be seen.

As mentioned before, in certain CDW specimens the onset of the current carrying state is not smooth, and the current–voltage characteristics display well-defined switching and hysteresis behavior. When subjected to combined ac and dc electric drive fields, such specimens again display dramatic mode locking, with hysteretic structure.[25] The dc response is like that predicted by the circle map, Eq. (10), and similar to that displayed in Fig. 5. However, the range of mode lock for the harmonic Shapiro steps, i.e., for $p = 1,2,3, \ldots ; s = 1$, is substantially larger, and the harmonic steps virtually fill all of Ω space, as shown in Fig. 7(a). The simultaneously measured ac response is a complicated function of ac amplitude E_{ac} and frequency ω_{ex}, and also dc bias E_{dc}. With fixed E_{ac} and ω_{ex}, increasing E_{dc} leads, on each mode locked step, to a period doubling route to chaos. This is displayed schematically in Fig. 7(a), which shows vertical windows, each corresponding to a particular period (or chaos) for the response. The corresponding frequency response spectra are displayed in Fig. 7(b). Period doubling bifurcations and a chaotic behavior, distinguished by large broad band noise, are clearly observed. A period doubling route to chaos can also be observed by keeping the dc bias constant and smoothly changing E_{ac} or ω_{ex}. By variation of all three parameters, i.e., E_{dc}, E_{ac}, and ω_{ex} systematically, the response boundary between period 1, 2, 4, . . . , and chaotic, solutions may be mapped out. In general, repetitive (hysteretic) period doubling routes to chaos can be induced by varying all three parameters monotonically through a limited range of phase space.

A repetitive period doubling route to chaos on each mode locked Shapiro step suggests a modulo 1 variable for the CDW phase, such as θ_m in Eq. (10). This equation is identically recovered by changing Ω to $\Omega + n$, with n an integer. The periodicity of the bifurcation sequence in dc bias is thus consistent with the periodicity of the behavior predicted by the circle map. Similarly, period doubling routes to chaos on mode locked Shapiro steps are predicted by Eq. (7), if the inertial term is retained.[27]

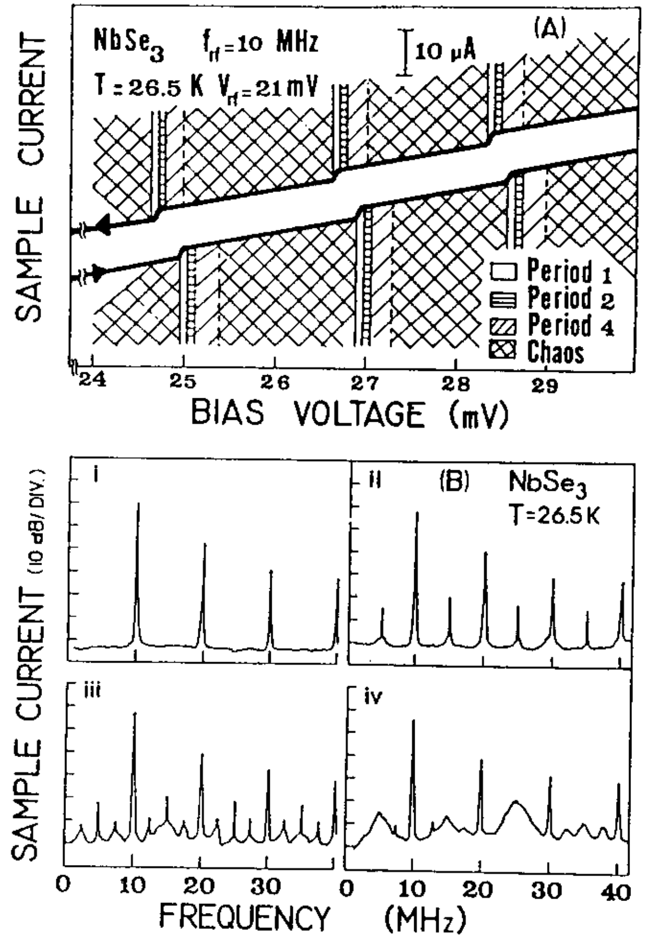

FIGURE 7 (a) Schematic representation of current response in Shapiro step region of $NbSe_3$, for forward and reverse bias voltage sweeps. The heavy lines are the direct I-V traces, offset vertically for clarity. (b) Frequency spectrum of current response in Shapiro step region. External rf drive frequency and amplitudes as in (a). (i) V_{dc} = 25.0 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 (from Ref. 25).

The assumption of an effective inertial term in Eq. (7) to describe the chaotic behavior can be justified by additional experiments. For example, as mentioned previously, the low field ac conductivity is a sensitive probe of CDW inertia. Experiments[44] performed on $NbSe_3$ crystals which display chaos show, for zero dc bias, overdamped (noninertial) response. However, for applied bias fields $E_{dc} > E_T$ (where chaos is observed), the low field ac conductivity shows[44] substantial inductive (inertial) response, i.e., lim $\sigma(\omega_{ex}) < 0$, precisely in the frequency range where chaos is observed. Figure 8 shows clear inductive frequency response (i.e., lim $\sigma < 0$) at low frequencies, measured for a $NbSe_3$ crystal which displayed switching, hysteresis, and chaos; the response is in qualitative

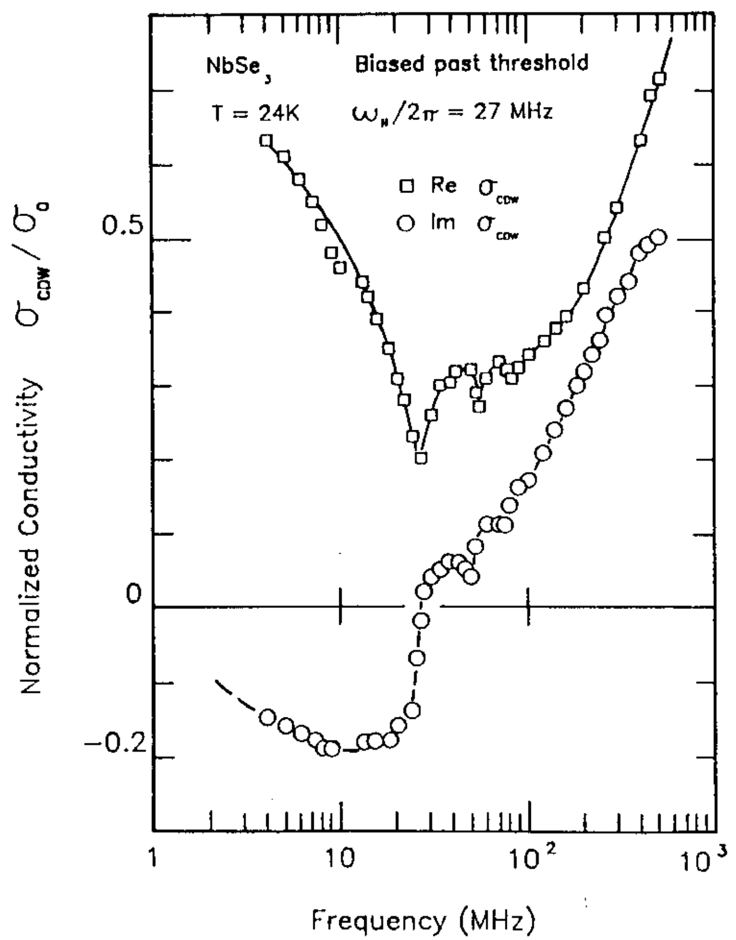

FIGURE 8 Ac response spectrum for $NbSe_3$ in sliding CDW state. This switching sample shows inductive response at low frequency (from Ref. 44).

agreement with analog computer solutions[44] of Eq. (7) for similar drive parameters, assuming underdamped behavior. Samples of $NbSe_3$ which display no chaos also show no extended inductive response in the ac conductivity, even for $E_{dc} > E_T$. In a sense, these nonswitching and nonhysteretic crystals are more fully overdamped. In the context of Eq. (7), the lack of chaotic behavior here is then not surprising.

The hysteretic behavior observed in the dc conductivity properties of CDW materials has other important consequences. Defining E_{T1} as the threshold field for the onset of CDW conduction as E_{dc} is increased from zero, and E_{T2} ($< E_{T1}$) as the critical field for switching back from the conducting to the pinned regime, it has been observed[45] that for $E_{T2} < E_{dc} < E_{T1}$, i.e., within the hysteresis loop, there exist a number of distinct current carrying states. Each state has a unique I-V characteristic, and both field induced and apparently random transitions between these states are observed. While the precise origin of the hysteretic behavior is not clear, it appears to reflect different numbers of macroscopic domains in the crystal conducting CDW current. In the hysteretic regime, different states are accessible to the system, each corresponding to a different current carrying configuration. In the hysteresis region, the system is then a prototype example of a bistable (or multistable, depending on the number of states available) configuration, with the possibility of intermittent chaotic behavior. This has been studied in detail by numerical simulations of Eq. (7), and the general picture which emerges is the following: outside the hysteresis region, the solutions correspond to periodic solutions, with $d\phi/dt = 0$ below E_{T2} and $d\phi/dt$ = (const.) above E_{T1}. In the regime $E_{T2} < E_{dc} < E_{T1}$, no periodic solution is stable, and the solution may hop between two periodic solutions. This leads to a large low frequency broad band noise, with the spectrum decaying algebraically at high frequency.[46] Enormous broad band noise associated with such instabilities has been observed[47] in $NbSe_3$ (with noise level corresponding to an effective temperature $T_{eff} = 10^6$ K), with strongly time dependent power spectra, representing both broad band and narrow band frequency structure. Figure 9 shows an example of frequency response spectra obtained for $NbSe_3$ in such an unstable, multistate configuration. This structure is observed in *addition* to the $T_{eff} = 10^6$ broad band noise. The obser-

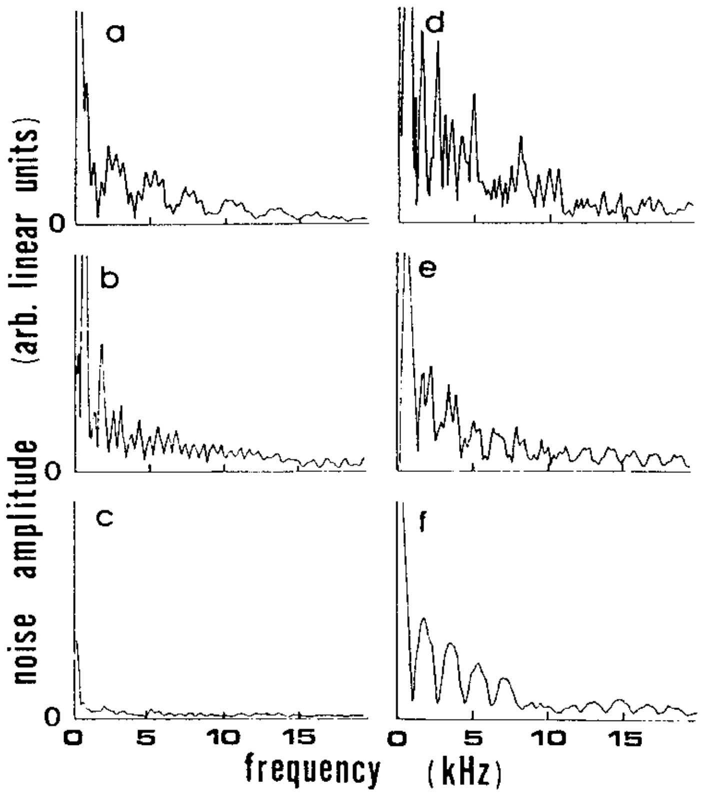

FIGURE 9 Successive fast Fourier transforms of voltage response in dc-biased $NbSe_3$ crystal in negative differential resistance region. Traces (a)–(f) were taken successively under identical experimental conditions. Frequency emissions correspond to intermittent chaos (from Ref. 47).

vations are qualitatively similar to those obtained by computer simulations of Eq. (7), and are suggestive of intermittent chaotic response in the hysteretic region. Similar results follow from other systems which exibit bistability.

4. CONCLUSION

Initial progress in the area of chaos in CDW systems raises several important and yet unresolved questions. While detailed comparisons between theory and experiment have not been performed in

the majority of cases, several routes to chaos have been qualitatively well established. A simple description in terms of single degree of freedom dynamics often requires important inertia effects, although this is often in clear contrast to the behavior of the small amplitude ac response.

For certain observations, such as intermittency, all that is required is a bistable configuration, and whatever the origin of switching and hysteresis in CDW's, its appearance also leads to intermittent routes to chaos. This, however, is most probably not valid for other observations such as period doubling routes to chaos or mode locking with subharmonic structure. There the dynamics of the internal deformations of the collective mode are most probably important, and they may lead to overdamped response for small amplitude ac and dc excitations, but may conspire to lead to apparent inertia-like effects for large amplitude ac and dc drives. While several calculations point in this direction, this "glassy" nonlinear dynamics aspect of the problem has not been explored in detail.

The experiments discussed here suggest that driven charge density waves are more than simply more complicated examples of nonlinear circuit analogs. They reflect the interplay between disorder and coherence in nonlinear dynamics. Some recent experiments also demonstrate that the nonlinear coupling between the intrinsic oscillations and externally applied ac fields leads, through phase synchronization, to enhanced coherence in the presence of large amplitude ac drives, where the dynamics of internal modes are strongly suppressed.[48] These experiments imply a scenario where the system evolves from a pinned glassy state to a fully coherent state, which then may display chaotic behavior. Together with more detailed investigations which differentiate clearly between the underdamped-like behavior of subharmonic interference and switching and hysteresis structure, and the overdamped low field ac response, these phenomena deserve further experimental and theoretical study.

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