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CHAOTIC RESPONSE OF DRIVEN CHARGE DENSITY WAVE SYSTEMS

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<u>Abstract</u> We investigate the response parameters of the charge density wave condensates of  $(TaSe_4)_2I$  and  $NbSe_3$  for ac, dc, or ac+dc combined electric drive fields. Chaotic conductivity obtains for well-defined ranges of temperature, driving field amplitude, and ac drive frequency. We discuss these results in terms of simple equations of motion for CDW transport.

Many non-linear dynamical systems exhibit chaotic response, even when driven by purely sinusoidal forces. A classic example is the simple damped pendulum, which, when driven sufficiently hard, undergoes transitions to chaotic response characteristic of a strange attractor in phase space 1,2. The intrinsic dc nonlinearity and strong ac polarization associated with charge density wave (CDW) condensates is in many ways analogous to response parameters of a damped rigid pendulum, and thus one might expect chaotic response for driven CDW systems 1.

We have investigated the possibility of chaotic conductivity in the CDW materials  $({\rm TaSe}_4)_2{\rm I}$  and NbSe $_3$ . Both systems exhibit chaotic response, but only under restricted conditions.

## (TaSe<sub>4</sub>) <sub>2</sub>I

In the CDW state, the Peierls semiconductor  $(TaSe_4)_2I$  shows anomalous ac conduction associated with excitations of the CDW condensate about pinning centers. In terms of a harmonic oscillator-type behavior, the response is overdamped, with characteristic pinning frequency in the far infrared<sup>3</sup>. Due to the strong damping, chaos is not expected, and is not observed, in the

presence of applied ac drive fields. However, an inductor placed in series with the crystal effectively introduces artificial CDW inertia. As shown in Fig. 1a, the response for such a hybrid circuit is underdamped; the high Q circuit resonates at 1.8 MHz, and is well described by a harmonic oscillator solution (solid and dashed lines in the figure). If driven near resonance, the circuit displays chaotic current response, observable as an increase in broad-band noise as shown in Fig. 1b. Chaos results for sufficiently high drive field, and within a well-defined drive frequency range, as mapped out in Fig. 1c. The chaotic conductivity is strictly associated with the pinned CDW, and the broad-band noise which results is orders of magnitude larger than conventional broad-band CDW sliding conduction noise.

The most simple analysis of our experimental results is to assume the model of Gruner, Zawadowski, and Chaikin for the CDW response. Although this model has many serious drawbacks, it often gives good qualitative, if not quantitative, accounts of overall CDW transport features. Within this model framework, we have for the voltage across the  $(TaSe_A)_DI$  hybrid circuit  $^5$ 

$$V(t) = \alpha \frac{d^2 \phi}{dt^2} + \beta \frac{d\phi}{dt} + \sin \gamma \phi, \quad \alpha, \beta, \gamma \text{ constants, } \phi \text{ CDW phase.}$$
 (1)

Eq.(1) has been analyzed in detail by many authors. Indeed, chaotic current response is predicted at sufficiently high ac drive field, and again within a well-defined drive frequency range<sup>2</sup>. To a first approximation, Eq.(1) describes extremely well the chaotic response observed in  $(TaSe_4)_2I$ . A notable discrepancy is the possiblity of period-doubling routes to chaos, which we have not observed experimentally.

The chaotic response observed in  $(TaSe_4)_2$  I is thus well described by simple determinisic equations of motion. Here the chaos results from driving the always pinned condensate about an anharmonic potential well.

## NbSe<sub>2</sub>

In the lower CDW state, the low-field ac conductivity of NbSe<sub>3</sub> again displays an overdamped response. Hence simply driving the condensate about its pinning center will not lead to chaos, even for large ac drive amplitude. Unfortunately, the "trick" of using a series inductance to introduce inertia into the CDW system does not work for NbSe<sub>3</sub>, due to the large conductivity contribution of remaining normal electrons. We have followed a related but different approach to investigate possible chaos in NbSe<sub>3</sub>. Some samples of NbSe<sub>3</sub> display dramatic switching and hysteresis at low temperatures, a phenomenon in itself suggestive of CDW "inertia". Hence, such samples might be expected to exhibit chaotic response. Indeed this is the case<sup>7</sup>.

Fig. 2a shows the dc I-V characteristics for a single NbSe<sub>3</sub> sample at T=26K. Switching and hysteresis are clearly observed, and the application of an ac field induces clear Shapiro steps. On each step, the CDW phase is locked to a harmonic of the external ac drive. A simultaneous observation of the ac response of the sample, performed with a spectrum analyzer, indicates that, on each Shapiro step, a period doubling route to chaos occurs. This is shown in Figs. 2b and 2c. Similar period doubling routes to chaos are found if dc bias is fixed on a particular step, and the ac amplitude is smoothly increased.

Obviously, a clear understanding of the observed period doubling route to chaos in NbSe<sub>3</sub> requires a fundamental understanding of switching and hysteresis phenomena. Although several models have recently been proposed to describe switching<sup>8</sup>, we shall here interpret our results as representing an example of universal instability in phase locking for a driven non linear system characterized by an intrinsic limit cycle. We assume a return map of the form

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

i.e. the map of the circle onto itself .  $\Gamma$  represents the ratio of

intrinsic to external frequency (in this case the narrow-band noise frequency to the external ac drive frequency), and C represents the strength of the coupling between the external perturbation and the system. In the parameter range C>1/2m, the circle map displays stable mode-locked solutions, which bifurcate successively to chaos as r is smoothly increased. Since  $\theta$  is a modulo 1 variable, the pattern of bifurcations to chaos will repeat itself as  $\Gamma$  is increased monotonically. The observed behavior in NbSe, is thus consistent with that predicted by the circle map. This map may be directly related to the equation describing a simple damped  $\mathsf{pendulum}^{\,9}$  . In the context of such a description, our experimental results suggest  $\tau^{-1}$  = 61 MHz for the CDW relaxation time, and  $\omega_0/2$ = 15 MHz for the intrinsic classical pinning frequency. These values are orders of magnitude smaller than those determined for the same crystal outside the switching regime. In the switching regime, the depinned CDW condensate in NbSe 3 displays an underdamped response.

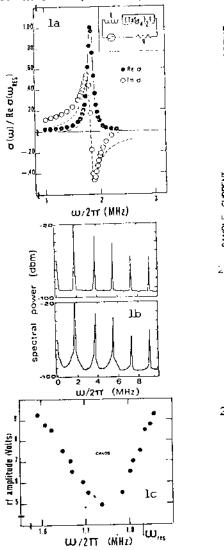
In conclusion, dynamic chaos has been observed in  $({\rm TaSe}_4)_2{\rm I}$  and  ${\rm Nbse}_3$ . The response is well described by simple deterministic equations of motion, although more detailed studies are required to, for example, clearly establish the attractor dimension, and the number of degrees of freedom necessary to describe CDW dynamics. These studies are currently underway.

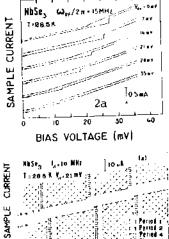
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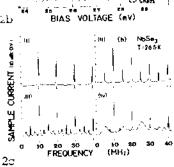


Fig.1 Chaos in  $(TaSe_4)_2T$ Fig. 2 Chaos in NbSe<sub>3</sub>