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Charge Density Waves in Solids

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CHAOS IN CHARGE DENSITY WAVE SYSTEMS

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We investigate chaotic response in the charge density wave (CDW) condensates of $(\text{TaSe}_4)_2\text{I}$ and NbSe_3 . In $(\text{TaSe}_4)_2\text{I}$, non bifurcative routes to chaos occur when the pinned CDW is excited by an external ac electric field. The behavior is interpreted as that of a driven anharmonic oscillator. In NbSe_3 in the switching regime, a period doubling route to chaos occurs for combined ac + dc fields. The route to chaos is characteristic of instabilities in phase lock for systems of competing periodicities. Intermittent chaos is also observed in dc biased NbSe_3 with negative differential resistance. We interpret the chaotic behavior in terms of simple models with restricted numbers of degrees of freedom, and return maps appropriate to these models.

Introduction

There has been much study on turbulent or chaotic behavior in systems which have macroscopic numbers of degrees of freedom. Of particular interest is the existence of universality classes describing the onset of chaos, which provides a direct connection between highly complex real systems, and simplified models representing only a small number of degrees of freedom. Well known examples of universality are the period doubling route to chaos¹ and the onset of intermittent chaos².

We shall here be interested in the association of chaos with the dynamics of charge density wave (CDW) condensates. A number of phenomena are investigated in $(\text{TaSe}_4)_2\text{I}$ and NbSe_3 which can, to a surprising degree, be well explained in terms of simple deterministic equations of motion possessing one dimensional return maps. Our purpose at present is to gain insight into CDW dynamics by analyzing the particular route to chaos involved.

The anharmonic oscillator: application to $(\text{TaSe}_4)_2\text{I}$

As was first discussed by Lee, Rice, and Anderson³, the low field ac response of a pinned CDW condensate might be expected to follow a damped harmonic oscillator behavior. For large ac drive fields, nonlinear anharmonic potential effects are inevitable. Under appropriate conditions, such anharmonic terms may lead to chaotic structure. Huberman and Crutchfield⁴ were the first to demonstrate for a simple anharmonic oscillator a bifurcation cascade to the chaotic state, with a response characterized by a strange attractor in phase space. A signature of the chaos is a dramatic rise in broadband noise in the

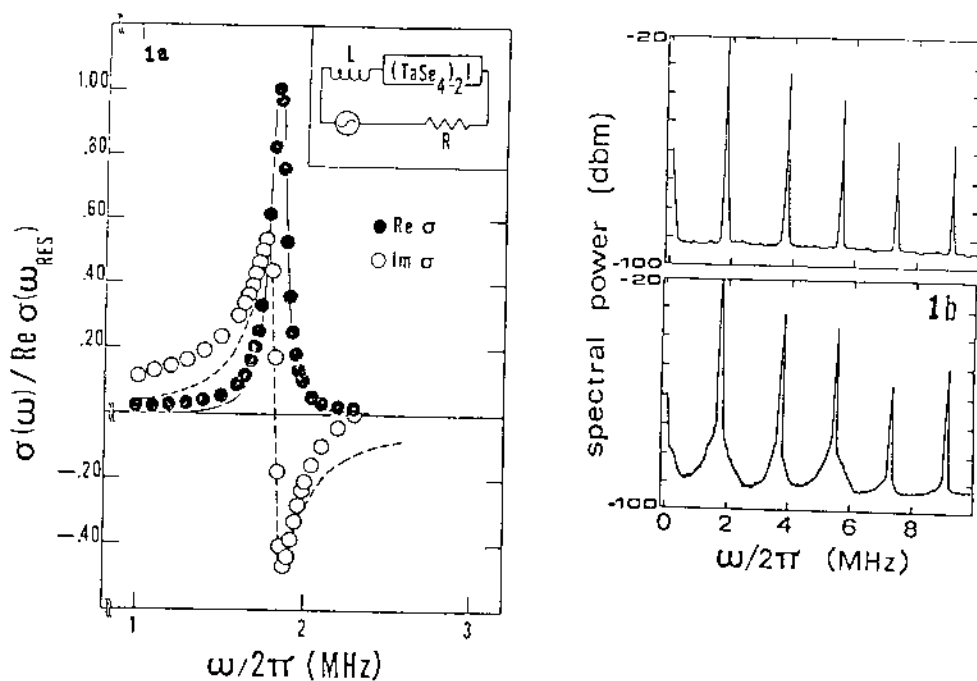
response spectral density. Subsequent studies of the damped pendulum⁵ have again demonstrated routes to chaos for sufficiently large ac drive amplitude. In dimensionless form, the damped pendulum equation of motion reads

$$\beta \frac{d^2\theta}{dt^2} + \frac{d\theta}{dt} + \sin\theta = c_{dc} + e_{ac} \sin \Omega t \quad (1)$$

Eq.(1) also describes a resistively shunted Josephson junction⁶, and it has been suggested to describe CDW dynamics in an approximate classical limit⁷. A critical parameter in Eq.(1) is β , which reflects system inertia. In the limit $\beta \rightarrow 0$, Eq.(1) does not predict dynamical chaos.

The low field ac conductivities of NbSe_3 , TaS_3 , and $(\text{TaSe}_4)_2\text{I}$ all appear to represent "overdamped" response, for which β is vanishingly small. Indeed, experiments aimed at achieving chaos in the pinned CDW states of these materials, by simply driving the condensate with an ac field, have not been successful.

We introduce finite inertia into the CDW system by addition of a real inductor in series with the CDW crystal. By appropriate choice of circuit parameters and sample, a high Q resonance circuit can be created, as demonstrated in Fig. 1a. The low field response of this hybrid $(\text{TaSe}_4)_2\text{I}$ circuit is underdamped, i.e. β is substantially greater than zero. For this circuit, as the ac drive amplitude is increased, chaotic response results for the pinned CDW condensate, yielding high-level broadband noise in the current response spectrum.



The onset of chaos is sudden and non-bifurcative. Such non-bifurcative transitions are quite possible in terms of Eq.(1), as has been discussed by various authors^{5,8}. Eq.(1) is able to account well for the chaotic response observed in this hybrid CDW circuit.

Phase lock and Shapiro steps: period doubling routes to chaos in NbSe₃.

In NbSe₃, dramatic Shapiro steps⁹ result for combined ac and dc drive fields, i.e. for finite e_{dc} and e_{ac} in Eq.(1). The steps are a manifestation of phase lock between the CDW condensate and the externally applied ac field. In the limit $\beta = 0$, subharmonic steps are not predicted. However, in NbSe₃ at $T=42K$, substantial subharmonic structure has been observed¹⁰, as demonstrated in Fig. 2a. This behavior would indicate a finite β . A detailed analysis¹¹ of such subharmonic structure in terms of the Devil's staircase has suggested that at this temperature NbSe₃ is close to, but not at, chaos.

At temperatures below 42K, NbSe₃ may show switching behavior¹². In a phenomenological sense switching drastically enhances β , leading to hysteresis

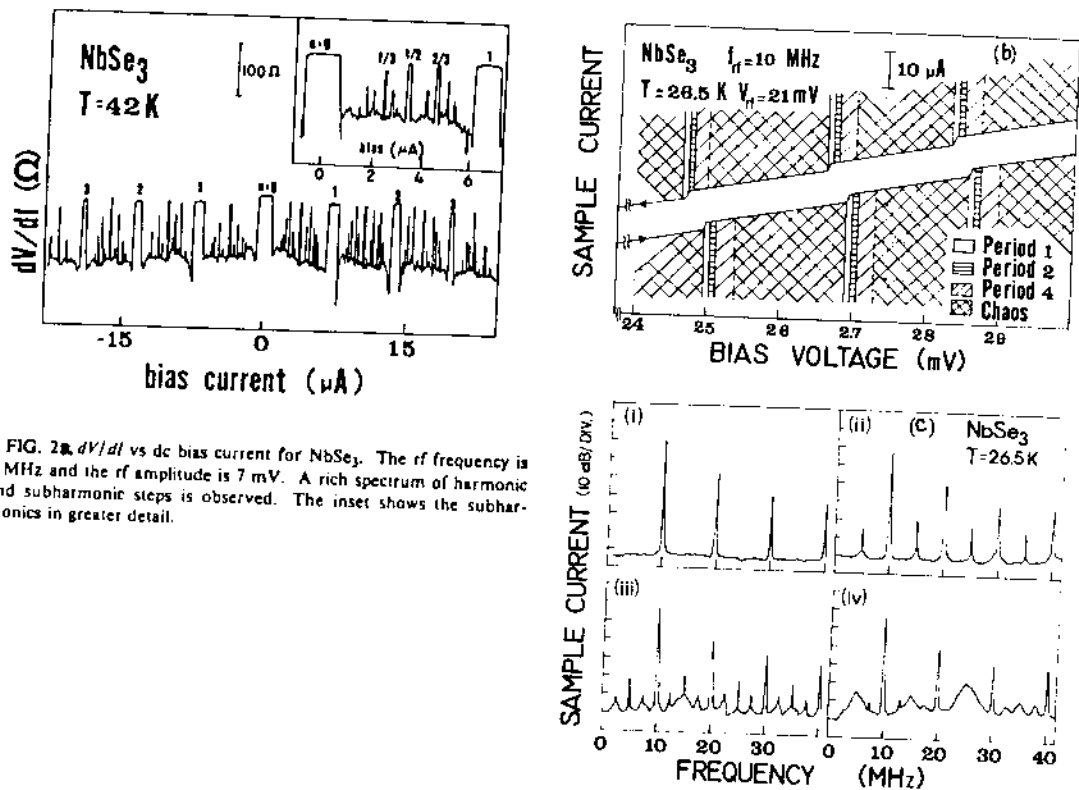


FIG. 2a. dV/dI vs dc bias current for NbSe₃. The rf frequency is 5 MHz and the rf amplitude is 7 mV. A rich spectrum of harmonic and subharmonic steps is observed. The inset shows the subharmonics in greater detail.

FIG. 2. (b) Schematic representation of current response in Shapiro step region, for forward- and reverse-bias voltage sweeps. (c) 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.

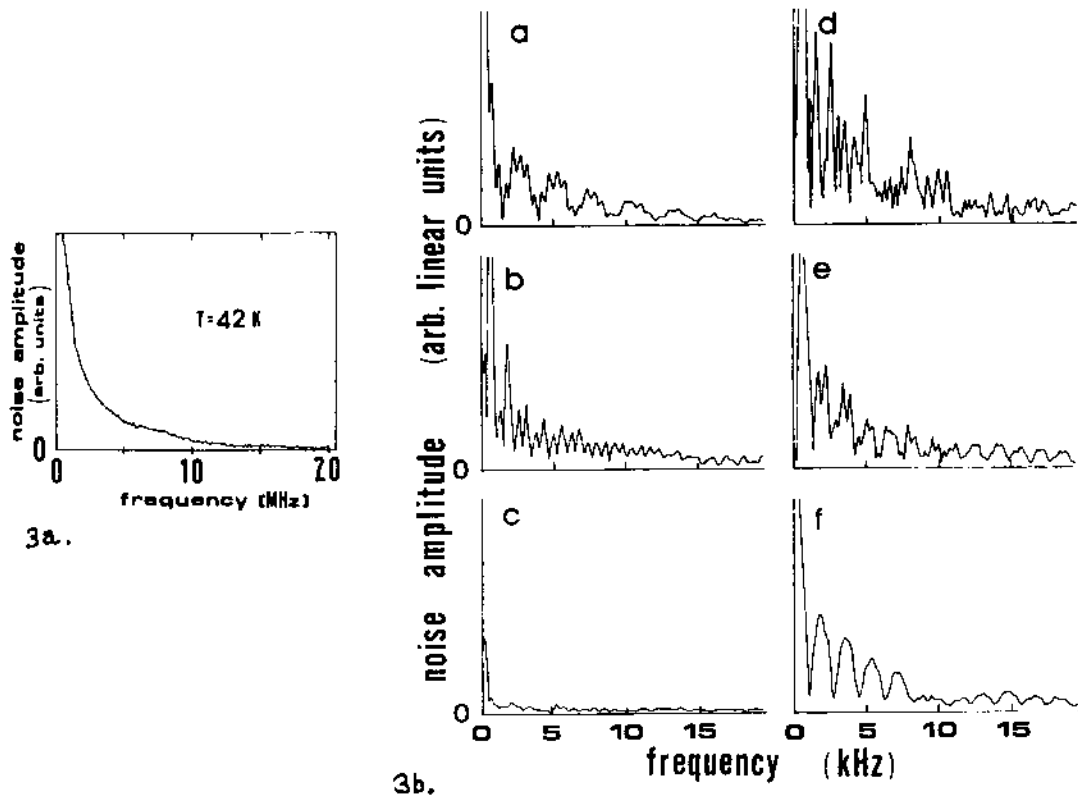
effects in the dc I-V characteristics, and also to modified Shapiro step structure. On each Shapiro step, an increase in dc bias field can lead to a well-defined period doubling route to chaos, as illustrated in Figs. 2b and 2c. The repetitive nature of the response is most easily interpreted in terms of the one dimensional return map of Eq.(1), i.e. the circle map¹³

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

where Γ represents the ratio of external drive frequency to internal (narrow-band noise) frequency. In the parameter range $C \gg 1/2\pi$, 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 patterns of bifurcations to chaos will repeat as Γ is increased monotonically. The bifurcation sequence in dc bias observed in NbSe₃ is consistent with the periodicity of the behavior predicted by the return map of Eq.(1).

System bistability: intermittency and 1/f noise

The initial onset of CDW dc conduction is in general quite smooth with no evidence for divergent behavior. In switching samples, however, CDW depinning is dramatically sharp. "Intermediate" switching may result in negative differential resistance (NDR) just beyond the depinning threshold, as has been observed¹⁴ in NbSe₃. The NDR region is associated with dramatic broadband noise response and additional random structure in the frequency spectrum, indicative of temporally intermittent chaos. Fig. 3a shows the voltage response spectrum of a NbSe₃ sample biased into the NDR region. The 1/f noise level is approximately 4 orders of magnitude larger than that associated with conventional CDW motion. Fig. 3b indicates the additional intermittent structure. Here data represent fast Fourier transforms of the voltage response, taken sequentially under identical experimental conditions. This type of chaotic behavior could arise from system bistability, as might occur for fluctuating current paths in the NbSe₃ sample (reflecting possibly macroscopic CDW domain structure). In the context of a domain model by Joos and Murray¹⁵, there can exist an instability between having n and $n+1$ channels open and conducting CDW current. The system may then hop between the $n+1$ and n open channel states, effectively representing hopping between valleys of a bistable system. The model is equivalent to that considered by Ben-Jacob et al¹⁶ for intermittent chaos in Josephson junctions. There the random-like hopping between states leads to intermittent chaos, with response



power spectra not unlike those of Fig. 3b. In NbSe_3 , the $1/f$ noise and intermittent structure could well represent quite similar processes, with of course different characteristic attempt frequencies.

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