Chaotic ac Conductivity in the Charge-Density-Wave State of $(TaSe_4)_2I$

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We report the observation of chaotic ac response in the charge-density-wave state of $(TaSe_4)_2I$, when coupled to an external inductance and driven by an rf electric field. The chaotic state is achieved, within a well-defined driving frequency window, by increasing rf amplitude. We analyze our results in terms of a simple nonlinear classical equation of motion for the charge-density-wave condensate.

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Many nonlinear dynamical systems are well described by simple deterministic equations of motion. Of great current interest is the study of solutions to these equations which appear to fluctuate randomly and characterize a chaotic state.[1] Chaos has been observed in a variety of systems, including the simple damped pendulum[2] with only a single degree of freedom, and fluid flow[3] with nearly an infinite number of degrees of freedom.

We have studied the onset of chaos in a new solid-state system, the charge-density-wave (CDW) condenstate of $(TaSe_4)_2I$ coupled to an external inductance. When the coupled system is driven by an external radio-frequency (rf) electric field, we find complex system ac current response which depends sensitively on the amplitude and frequency of the rf driving field.

The linear-chain compound $(TaSe_4)_2I$ is semimetallic at room temperature, and undergoes a Peierls transition to a semiconducting CDW state at $T_P = 265$ K.[4,5] The resulting CDW is incommensurate with the underlying lattice along the chain (c) axis of the crystal.[6] Below T_P, nonlinear dc conduction is observed[4,5] for applied dc electric fields E exceeding a highly temperature-dependent threshold field E_T. The minimum value of E_T is approximately 1.5 V/cm at 250 K. Again below T_P, the low-field ac conductivity $\sigma(\omega)$ is strongly frequency dependent with anomalous response observed in the megahertz to gigahertz region.[5,7] The excess dc and ac responses have been interpreted as due to collective motion of the CDW condensate. For low applied dc fields the CDW is assumed to be pinned to the underlying lattice by impurities, and only when the field energy exceeds the pinning potential does the CDW become mobile and contribute to the dc conductivity by the Frölich[8] mechanism. Displacing the CDW by one wavelength λ results in the same energy configuration; hence the pinning potential may be considered a spatially periodic function. In the pinned state, the CDW is unable to contribute to the dc conductivity. However, as was first suggested by Lee, Rice, and Anderson,[9] in the pinned regime the oscillator strength is moved to finite frequency and thus anomalous ac conduction results even for very-low-amplitude ac driving fields. The predicted response is that of a damped harmonic oscillator. Huberman and Crutchfield[10] were the first to suggest that turbulent regimes might be realized in a CDW system by exciting the condensate about its pinning center, with a driving-field amplitude sufficiently large to feel the anharmonicity of the pinning potential. This possibility has also been considered more recently by MacDonald and Plischke.[11]

We have grown crystals of $(TaSe_4)_2I$ by direct reaction of the elements in a gradient furnace at 500 °C, with typical crystal dimensions $1\times1\times10$ mm^3. Actual samples used in this study were significantly smaller, having been easily cleaved from a large single crystal. Electrical contact to the sample was made by evaporated gold pads covered by conductive silver paint. Careful checks indicated negligible contact impedance.

Our experimental system consists of a single $(TaSe_4)_2I$ crystal mounted in series with a miniature inductor L (of negligible resistance) and a resistor R, as shown in the inset of Fig. 1. The combination system (i.e., inductor + sample + resistor) was driven by a sine wave oscillator coupled to a high-speed follower circuit constructed in our laboratory, with an output impedance of approximately 1 Ω. The voltage across the system was thus of the form $V = V_{ac}\cos(2\pi f t)$.

The purpose of the inductor was to scale down the $(TaSe_4)_2I$ response frequency from the microwave[5] to the rf, and to improve the overall quality factor of the resonant L-sample-R circuit. Figure 1 shows both the real and the imaginary parts of the low-field ac conductivity of the system for typical component parameters $L = 10$ mH, $R = 10$ Ω, and a sample temperature $T = 77$ K. The conductivity

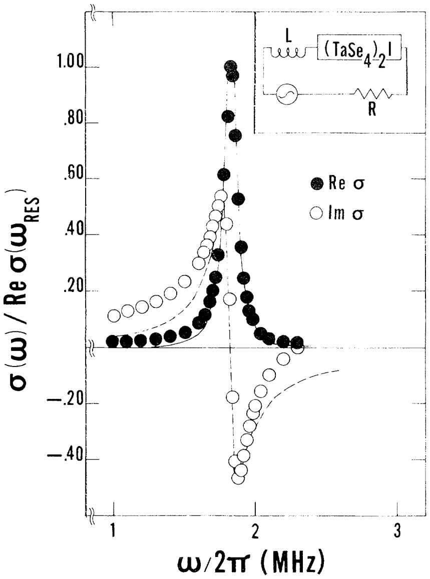

FIG. 1. Low-field ac conductivity of circuit displayed in inset. Solid and dashed lines are the predictions of Eq. (2).

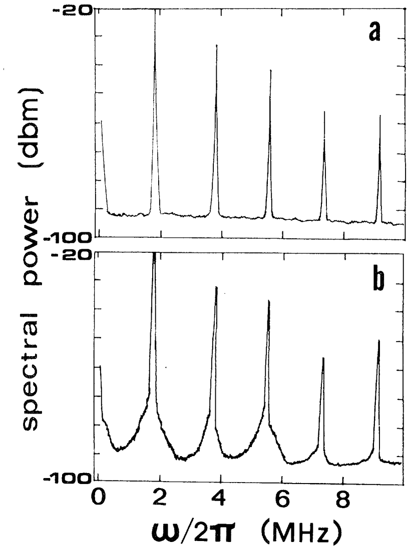

FIG. 2. (a) Response spectrum for $(TaSe_4)_2I$ circuit (see inset, Fig. 1), with $f = 1.8$ MHz and $V_{ac} = 5$ V. (b) Same as (a), except V_{ac} increased to 6 V. Detector gain is 20 dB; bandwidth 10 kHz.

was determined by fixing V_{ac} at a small but finite value and measuring the magnitude and phase of the current through the resistance R, for various frequencies f. The conductivity observed is suggestive of an underdamped–harmonic-oscillator response. From the resonance frequency $f_{res} = 1.830$ MHz in Fig. 1, and from the relation $2\pi f_{res} = (LC)^{-1/2}$, we obtain an effective sample capacitance $C = 0.75$ pF. With the measured sample length $l = 0.7$ mm and cross-sectional area $A = 2.5 \times 10^{-5}$ cm^2 this corresponds to a dielectric constant $\epsilon = 10^4$, consistent with that obtained from previous ac conductivity studies[7] of $(TaSe_4)_2I$ at low temperatures.

To test for chaotic ac response, L and R were fixed, and V_{ac} and f were independently varied. The detected quantity was the rf voltage across R, observed in both the time and the frequency domains. Figure 2 shows, for two values of V_{ac}, the frequency power spectrum for the current response with a driving frequency near resonace and a sample temperature $T = 77$ K. L and R are again 10 mH and 10 Ω. Because of the nonlinear ac response of the crystal, Fig. 2(a) shows strong harmonics of f in the current response. However, no other outstanding features are observed. As the ac amplitude is increased across the circuit, a sudden onset of chaotic response occurs, as evidenced by the appearance of high-level broadband noise in the response power spectrum. This is illustrated in Fig. 2(b). The noise spectrum in Fig. 2(b) is complicated and we shall not attempt to analyze it in detail. We note, however, that much of the noise power lies on the flanks of the fundamental driving frequency and its harmonics, and the distribution about these narrow-band frequencies is asymmetric. Using the relation $\langle V^2 \rangle = 4Rk_BT\Delta f$, where Δf is the detection bandwidth, we find an effective noise temperature range for the chaotic response of 10^4–10^6 K.

For a given large ac amplitude V_{ac}, chaotic response is observed only within a well-defined driving frequency window. This is illustrated in Fig. 3, where the chaotic state is defined within V_{ac}-f phase space. These data were generated by fixing

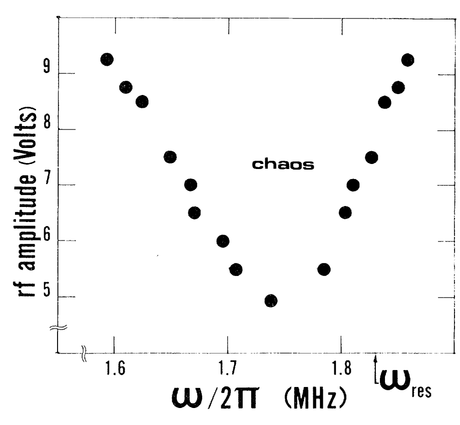

FIG. 3. Periodic-chaotic response boundary for $(TaSe_4)_2I$ circuit (see inset, Fig. 1).

V_{ac}, and sweeping f into and out of the chaotic regime. We note that the minimum ac amplitude needed for chaos occurs at a frequency somewhat below the resonance frequency of the driven system.

In Fig. 3, the critical amplitude V_c necessary for chaotic response is approximately 5 V at 1.75 MHz. We are not able to determine with certainty the ratio V_c/V_T, where V_T is the threshold voltage for dc CDW depinning. The threshold field in $(TaSe_4)_2I$ increases rapidly with decreasing temperature below 220 K, and is approximately 10 V/cm at 120 K. In our samples at 77 K slight non-Ohmicity was observed even for applied dc voltages $V_{dc} \rightarrow 0$; hence a sharp threshold voltage for the onset of dc CDW conduction could not be clearly established.

It is, however, very important to realize that the observed chaotic response in $(TaSe_4)_2I$ is fundamentally different from the usual conduction noise associated with a sliding CDW state.[4,12] In sliding CDW materials [including $(TaSe_4)_2I$ (Ref. 5)], conduction noise appears spontaneously when a dc bias exceeding the threshold field is applied. In the $(TaSe_4)_2I$ system at 77 K, such conduction noise was not observed for applied dc fields sufficiently large to induce substantial CDW depinning, yielding a sample current density nearly double the Ohmic value. Hence the chaotic response described here is strictly associated with the ac conductivity of the CDW condensate.

We remark that the onset of chaotic conductivity in our system is quite temperature dependent. With increasing temperature above 77 K, it becomes increasingly difficult to achieve a chaotic state (i.e., the frequency window decreases and the critical rf amplitude increases). Above approximately 90 K we were not able to observe chaotic response for any available rf drive amplitude and frequency.

We now discuss these results. As described previously, $(TaSe_4)_2I$ in the CDW state is an intrinsic nonlinear device. Numerous models, both classical and quantum mechanical, have been proposed to describe CDW dynamics in detail. A very simple phenomenological model developed by Gruner, Zawadowski, and Chaikin[12] treats the CDW as a rigid object in a periodic potential, obeying classical dynamics with only a single degree of freedom. This model has been successful in describing many overall features of electronic transport in the CDW materials $NbSe_3$ and TaS_3. The model also provides excellent quantitative fits to experiments performed in the presence of combined ac and dc fields, for example, ac-induced dc conductivity[13] and Shapiro-steps phenomena.[14] The CDW equation of motion in this classical model is

$$\frac{d^2x}{dt^2}+\frac{1}{\tau}\frac{dx}{dt}+\frac{\omega_0^2}{Q}\sin Qx=\frac{eE}{m^*}, \tag{1}$$

where x is the CDW position, $1/\tau=\gamma/m^*$ with γ the damping coefficient and m^* the effective mass of electrons in the CDW state, $\omega_0^2=k/m^*$ with k the restoring force constant, and $Q=2\pi/\lambda$.

We model our system as the circuit shown in the inset of Fig. 1, with the sample consisting of a CDW condensate described by Eq. (1) in parallel with a resistance R_N representing normal carriers thermally excited across the Peierls gap. The total voltage across the circuit then becomes

$$V(t)=\frac{Llm^*}{eR_N}\frac{d^3x}{dt^3}+\left[\frac{Ll}{eR_N}\frac{m^*}{\tau}+Ln_ceA+\left(1+\frac{R}{R_N}\right)\frac{l}{e}m^*\right]\frac{d^2x}{dt^2}$$
$$+\left[\frac{L\omega_0^2m^*l}{eR_N}\cos Qx+\left(1+\frac{R}{R_N}\right)\frac{l}{e}\frac{m^*}{\tau}+Rn_ceA\right]\frac{dx}{dt}+\left(1+\frac{R}{R_N}\right)\frac{\omega_0^2m^*l}{Qe}\sin Qx, \tag{2}$$

where l is the length of the crystal and A its cross-sectional area, $R_N=\rho_Nl/A$ with ρ_N the normal low-field resistivity of the crystal, and n_c is the concentration of carriers condensed in the CDW state. For ρ_N large, as is appropriate for $(TaSe_4)_2I$ at low temperatures, Eq. (2) may be approximated by

$$V(t)\simeq\left(LAn_ce+\frac{lm^*}{e}\right)\frac{d^2x}{dt^2}+\left(\frac{l}{e}\frac{m^*}{\tau}+Rn_ceA\right)\frac{dx}{dt}+\frac{l\omega_0^2m^*}{Qe}\sin Qx. \tag{3}$$

The easily derived low-field ac conductivity solution of Eq. (3) has been plotted in Fig. 1 with chosen parameters $\omega_1/2\pi=1.83$ MHz and $(\omega_1/\omega_0^2\tau+R/\omega_1L)^{-1}=18$, where $\omega_1^2=\omega_0^2/(1+LAn_ce^2/m^*l)$. We note that the effective system inertia is nonnegligible.

Equation (3) is formally equivalent to the damped-pedulum problem or the Steward-McCumber equation describing a resistively shunted Josephson junction with capacitance.[2] These systems have been studied extensively, and chaotic states are observed for a wide parameter range. Digital and analog computer solutions[2] of Eq. (3) with $(\omega_1/\omega_0^2\tau + R/\omega_1 L)^{-1} = 4$ show an onset of chaos near $\omega/\omega_1 = 0.7$, in reasonable agreement with the experimentally determined value $\omega/\omega_1 = 0.95$ for the onset of chaos in the $(TaSe_4)_2I$ system. The general form of the chaotic-periodic response boundary in Fig. 3 is also in qualitative agreement with that predicted by Eq. (3). We note, however, that Eq. (3) suggests, under certain conditions, a period-doubling route to chaos. We have not observed a bifurcation cascade in our experimental system, although we remark that this is expected to occur only over a limited V_{ac}-f parameter range. Indeed, calculations[15] by Ben-Jacob *et al.* and Kautz have demonstrated that bifurcative processes are not necessary to achieve chaotic response. Our experimental results are thus a new example of such nonbifurcative routes to chaos. The temperature dependence of the onset of chaos in the $(TaSe_4)_2I$ system is a direct result of the relation between system parameters in Eq. (2). A critical parameter is R_N, the low-field (normal) resistance of the CDW crystal. Because of the semiconducting nature of $(TaSe_4)_2I$, R_N decreases strongly with increasing temperature, and for sufficiently high temperatures the approximations made in reducing Eq. (2) to Eq. (3) are no longer valid. For the particular component values $L = 10$ mH and $R = 10\ \Omega$, we find the approximations to remain valid as long as $R_N > 2\times 10^6\ \Omega$. The measured low-field dc resistivity of the $(TaSe_4)_2I$ crystal indicates that $T = 89$ K at $R_N = 2\times 10^6\ \Omega$, in excellent agreement with the observed temperature for loss of chaotic response of the coupled system, $T = 90$ K. It should perhaps be emphasized that the temperature at which chaotic response is lost is not intrinsic to $(TaSe_4)_2I$ itself, but is a function of all system parameters.

The success of Eq. (3) in describing at least qualitatively our experimental findings indicates that, under certain conditions, the CDW condensate obeys simple deterministic equations of motion. Whether our experimental observations are consistent with other models of CDW conduction, for example, descriptions[16] where the internal dynamics of the CDW critically determine the response characteristics, or models[17] where the CDW undergoes quantum tunneling through a pinning gap, remains to be seen.

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