Subharmonic Shapiro Steps and Devil's-Staircase Behavior in Driven Charge-Density-Wave Systems

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(Received 14 February 1984)

Subharmonic steps in the dc I-V characteristics are reported in the model compound $NbSe_3$, when subjected to combined dc and ac drives. The steps occur at currents which correspond to frequencies $\omega_{int}=(p/q)\omega_{ext}$, with p and q integers. These steps cannot be explained in terms of the familiar classical equation of motion in a single degree of freedom, because the inertial term is too small. The observed behavior is compared with the devil's staircase, and a fractal dimension of 0.91 ± 0.03 is extracted.

PACS numbers: 72.15.Nj, 03.20.+i, 47.20.+m

The anomalous transport properties observed in certain inorganic linear-chain compounds are suggestive of a novel transport phenomenon carried by charge-density waves (CDW's).[1] The conductivity is highly nonlinear when the applied electric field E exceeds a threshold field E_T. In the nonlinear region current oscillations, with fundamental frequency ω_{int} proportional to the CDW current I_{CDW}, are observed. Although various models (all based on the original idea of Frohlich, that current is carried by moving CDW's) have been advanced to account for the experimental observations, we will use here a simple phenomenological equation of motion,[2] that of a particle moving in a periodic potential. In dimensionless form,

$$\ddot{\theta}+(\omega_0\tau)^{-1}\dot{\theta}+\sin\theta=E/E_T, \tag{1}$$

where $E_T=m\omega_0^2/2k_Fe$ is the threshold field for the onset of nonlinear conduction, τ and ω_0 are the damping constant and resonant frequency, k_F is the Fermi wave vector, and m and e are the mass and charge of the CDW. $\theta=2k_Fx$, with x the position of the CDW, and time is measured in units of ω_0^{-1}. The relation between this description and the tunneling model is discussed by Bardeen.[3] Equation (1) is formally analogous to the resistively shunted Josephson junction (RSJ) model,[4]

$$\ddot{\phi}+G\dot{\phi}+\sin\phi=I/I_J, \tag{2}$$

with $G=(\omega_JRC)^{-1}$, where R and C are the resistance and capacitance of the junction, ω_J the Josephson plasma frequency, and I_J the critical current. The intrinsic CDW oscillation frequency with $\omega_{int}=\text{const}\times I_{CDW}$ corresponds to the ac Josephson effect. This analogy has been extensively used to explore harmonic steps in the response of CDW's to the joint application of ac and dc fields, $E=E_{dc}+E_{ac}\cos\omega t$.[5]

Josephson junctions are regarded as excellent model systems for the study of nonlinear phenomena, including frequency locking and the development of chaos.[6] In driven systems with $I>I_J$ or $E>E_T$ in the presence of an applied ac excitation, a "devil's-staircase" structure has been predicted[7,8] as a consequence of the interplay between the two competing periodicities (that of the intrinsic, Josephson or CDW frequency, and that of the applied frequency). Recently, Bak[9] suggested that such behavior could also occur in driven CDW systems.

In this Letter we report the observation of subharmonic steps in the I-V characteristics of $NbSe_3$. The observed steps are remarkable in two respects. First, their magnitude is much greater than that expected in terms of Eq. (1) within the experimental limit on the magnitude of the mass term. Second, our results resemble the devil's-staircase calculations mentioned above,[7,8] and in fact we can extract a fractal dimension that is the same as that of Ref. 7 within experimental error.

The measurements were performed on $NbSe_3$, which shows all the phenomena characteristic of CDW transport, and in which the intrinsic oscillations are readily observed. Instead of measuring the I-V curve directly in the presence of applied rf fields, the differential resistance, dV/dI, was measured by use of a low-frequency lock-in configuration, with small ac modulation and very slowly sweeping.[10] Peaks in the derivative correspond to steps in the direct I-V curve, with the width of the peak in voltage units corresponding to the height of the step. The dc (plus modulation) was provided by a current source (which, we note, corresponds to neither the voltage-driven nor current-driven boundary condition on the RSJ equation). The rf was provided by a voltage source.

Figure 1 shows the differential resistance mea-

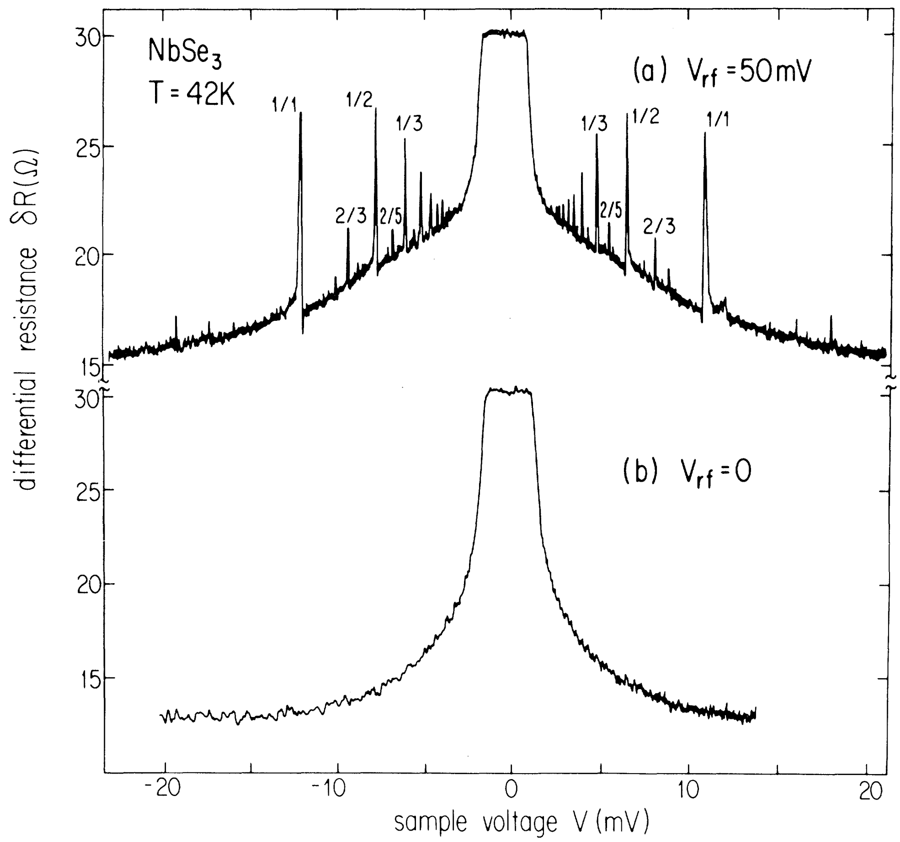

FIG. 1. Differential resistance vs dc sample voltage (a) with and (b) without applied rf voltage at 25 MHz. The peaks in (a) correspond to steps in the direct I-V curve. A few peaks are identified by p/q (see text).

sured with and without externally applied rf voltage at $\omega_{\text{ext}}/2\pi = 25$ MHz. For the rather low rf voltage used here, E_T is not reduced from the value in the absence of rf, but there appears a whole family of peaks at impressed dc current such that

$$p\,\omega_{\text{ext}} = q\,\omega_{\text{int}}, \tag{3}$$

where p and q are integers. The ratio $\omega_{\text{int}}/2\pi I_{\text{CDW}}$ was found to be 30 MHz cm^2/A in agreement with earlier studies.[5,10] A few of the peaks are identified by p/q in the figure. The identifications were confirmed for several p/q by plotting I_{CDW} vs ω_{ext} and checking that the slopes were indeed p/q times that for the fundamental. The features with small p and q are more conspicuous, being both taller and wider. Figure 1 is a typical trace. By varying V_{rf} and ω_{ext}, either more or fewer steps can be observed. While the dependence on these parameters is not strong, substantially increasing or decreasing either V_{rf} or ω_{ext} significantly reduces the number of steps which can be observed.[8]

The steps with $q = 1$, $p \geqslant 1$ have been studied previously[5,11] and are well known to arise from frequency locking between the internal CDW (or Josephson) frequency ω_{int} and all harmonics $p\,\omega_{\text{ext}}$ of the applied rf field. The harmonics of ω_{ext} are induced by the inherent nonlinearity of the system. Direct spectral analysis of the current oscillations[1] in the absence of applied rf reveals rich harmonic current, with intensity of the harmonics $q\,\omega_{\text{int}}$ decaying slowly with q. Hence it is natural to interpret the peaks as regions in which any harmonic of the internal frequency locks to any harmonic of the external field.[12] To the extent that mode locking within such regions is complete, the CDW is unable to respond to changes in the applied dc voltage, and the resistance rises to that of the normal electrons alone. In the present experiment, the steps have lower resistance than the normal electrons, indicating that the locking is not complete. The tendency to lock is weaker for larger p and q.

It is largely agreed that complete mode locking occurs for Eq. (2) only because of the inertial term.[9,13,14] In $NbSe_3$ the inertial term is negligible up to several hundred megahertz.[15] The argument, based on the frequency-dependent conductivity derived from Eq. (1), is detailed by Zettl, Jackson, and Gruner.[16] Therefore the subharmonic steps cannot be explained by use of Eq. (1). We speculate that they are absent because Eq. (1) treats only the center-of-mass motion of the CDW. Evidence of the importance of other degrees of freedom may be found in the volume dependence of the current oscillations[17] and in the frequency dependence of the conductivity at low frequencies.[18] One way in which these additional modes may be modeled is by dividing the sample into several or many regions,[11] each of which obeys Eq. (1) with no inertial term but supplemented by terms coupling the regions. This results in a system of coupled first-order nonlinear equations. Such systems can exhibit bifurcations, mode locking, and chaotic behavior reminiscent of the RSJ model.[19] This picture might also explain why the mode locking is not complete.

Whatever the origin of the subharmonic steps, the experiments show that the I-V curve contains a great array of steps of various sizes. The shape of such a curve has been described as a "devil's staircase."[7,8] If one looks more carefully (e.g., by sweeping more slowly with longer time constant), one sees more steps. It is conceivable that, for any value of current, one is on a step, in which case the staircase is called "complete." Instrumental noise sets a lower limit to the size of the step which can be observed, and so we cannot directly establish completeness. Yet a necessary condition for the staircase to be complete is that it pass the following test[7]:

The test will be performed on an interval of length l in CDW current. Choosing a discrimination level r, one adds up the total length $S(r)$ of steps larger than r.[20] If $N(r)=[l-S(r)]/r$, rN is the fraction of the interval unoccupied by steps larger than r. A complete staircase has $rN \to 0$ as $r \to 0$. Figure 2 shows that on the interval $p/q=(0,\frac{1}{2})$

$$N(r) \propto (r^{-1})^D \tag{4}$$

with a fractal dimension $D=0.91 \pm 0.03$, for two different rf voltages. The same result, within experimental error, was obtained on the intervals $(\frac{1}{2},1)$ and $(\frac{1}{3},\frac{2}{3})$. The result $D=1$ would have indicated an incomplete staircase, although we cannot prove that D will not revert to 1 for smaller r.

It is intriguing that a fractal dimension $D=0.87$ was obtained for devil's-staircase structure derived from circle maps,[7] and $D=0.91 \pm 0.04$ for an RSJ simulator.[21] The relevance of the circle map to the RSJ equation may be argued on the basis that the former illustrates the effect of competing periodicities in a dissipative system[7]; more directly, it has been shown numerically that the return map for the RSJ equation is a circle map.[22] But its relevance to the present experiment is not at all apparent and may be merely fortuitous, for we have argued that Eq. (1) is inadequate to describe the driven CDW system. Furthermore, one expects on the basis of the circle map and the RSJ simulator[21] that the value of D should depend critically on V_{rf}, and on the selected range of p/q, in contrast to what we have found.

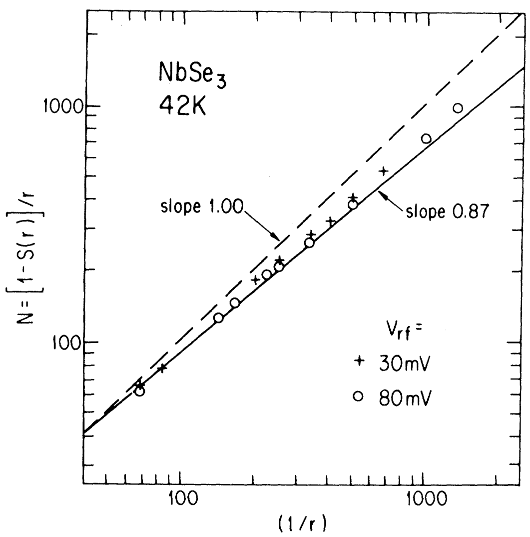

FIG. 2. Log-log plot of N vs $1/r$ for $NbSe_3$ at two rf levels. The slope gives the fractal dimension D. The data are fitted by $D=0.91 \pm 0.03$, while the solid line corresponds to the circle-map calculations ($D=0.87$).

These new results on CDW dynamics have raised several important questions. The first is why we observe subharmonic steps at all in a system which has negligible inertia. Our suggestion that the answer lies in couplings between degrees of freedom needs further investigation. Second, we see no reason why the fractal dimension is the same as the circle-map result, especially since we do not find the expected dependence of D on V_{rf} and on the range of p/q. Can it be shown that a system of coupled equations as described above falls in the same universality class as the circle map? If so, perhaps a distribution of couplings softens the dependence of D on V_{rf} and p/q. Finally we note that the circle map has chaotic solutions,[7] whose possible relevance to the CDW system[9] has not yet been explored.[23]

We thank Per Bak who introduced us to the subject and gave valuable suggestions for the analysis of our experiments. We acknowledge helpful discussions with E. Ben-Jacob, S. Coppersmith, Y. H. Kao, P. Littlewood, J. Rudnick, and W. J. Yeh. This work was supported by National Science Foundation Grant No. DMR 81-21394.

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