Scaling near Mode Locking in a Charge Density Wave Conductor

Mark J. Higgins, A. Alan Middleton, and S. Bhattacharya
NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540
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A striking evolution of the *I-V* curves is observed in a single sample (i.e., for a given realization of quenched disorder) of the charge density wave conductor $NbSe_3$ in the presence of an external rf field of varying amplitude. This yields qualitatively different "depinning transitions" from the pinned and the mode-locked states. In some regimes the transition is apparently continuous and shows scaling behavior in agreement with some recent theoretical results. In other regimes the transition turns first order at both harmonic and subharmonic steps, but with a marked asymmetry.

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The incommensurate charge density wave (CDW) in quasi-one-dimensional conductors [1] has served as a prototype of collective transport in disordered systems [2]. Above a threshold voltage V_T, a measure of the pinning strength due to quenched random impurities, the CDW slides carrying current. Despite many attempts, both theoretical and experimental, a consensus on the sliding state and the depinning transition is still lacking. A recent resurgence in theoretical work [3-7] addressing various aspects of the sliding CDW has led to several new predictions. Progress on the experimental front, however, has been impeded by consistent observation of distinct types of behavior observed in different samples, in most cases without any useful clues to the origin of the differences, but attributed to the presumably different nature of quenched disorder. This has raised the issues of what is intrinsic to these systems and what is the minimal set of experimental observations that needs to be addressed by a successful theory.

In this paper we report new results obtained in the study of mode-locking behavior in the CDW conductor $NbSe_3$. These results show that under controlled conditions, specifically under an external rf field of varying amplitude, a single sample (with a given realization of quenched disorder) displays diverse behavior that is typically observed in different samples and is considered distinct, perhaps requiring distinct explanations.

Mode locking, analogous to Shapiro steps in Josephson junctions, is observed in the CDW systems in the presence of an rf field of frequency ω_{rf}. The washboard frequency, often called the narrow band noise (NBN) frequency ω_{nbn}, of the sliding CDW (given by $\omega_{nbn}=\mathbf{q}_{cdw}\cdot\mathbf{v}$, where $\mathbf{q}_{cdw}$ is the wave vector and $\mathbf{v}$ is the velocity of the CDW), and thereby the CDW velocity, locks to the external frequency when the frequency ratio, called the winding number $W(=\omega_{nbn}/\omega_{rf})$, equals p/q, where p and q are integers. The *I-V* curves for the CDW are given by

$$I=V/R_n+I_{cdw}(V,V_T)\,, \tag{1}$$

where R_n is the normal electron resistance. Thus, locking of the CDW velocity, i.e., I_{cdw}, implies that the differential resistance R_d $(=dV/dI)$ equals R_n on the mode-locked steps, which is the typical experimental signature of mode locking.

The functional form of I_{cdw} in Eq. (1) has received much attention. Fisher's description of depinning as a dynamical critical phenomenon [2] suggests a power-law form:

$$I_{cdw}\sim(V-V_T)^{\beta}\,, \tag{2}$$

where β is the critical exponent. The exact value of β remains uncertain due to the finite rounding of the transition often encountered experimentally which obscures the behavior in the critical regime near threshold. Recently, it has been shown [3] by numerical simulations that the same scaling behavior is obtained near every mode-locked step:

$$|I_{cdw}-I_{p/q}|\sim|V-V_{p/q}^{\pm}|^{\beta}\,, \tag{3}$$

where $I_{p/q}$ is the mode-locked value of the CDW current on the step and $V_{p/q}$ denotes the voltage at which the mode-locked step occurs. The superscripts refer to the upper and lower edges of the step, respectively, i.e., the width of a step $\Delta V_{p/q}\equiv V_{p/q}^{+}-V_{p/q}^{-}$.

Results reported here are obtained in a small sample (4 μm$\times$0.8 μm$\times$1.6 mm) of reasonably high purity (residual resistance ratio, 236) in a two probe configuration below the lower CDW transition. The sample displays a single narrow band noise fundamental and the locked steps are indexed by a single set of p/q values. R_d is obtained by a low frequency modulation technique in a current-biased configuration. Complete locking is observed at harmonic and low order subharmonics where R_d equals R_n.

Measured R_d with different values of rf amplitudes is shown in Fig. 1. The data with no rf field, curve *a*, show a smooth and somewhat rounded depinning transition and R_d decreases concave downwards. An analysis of data such as these in terms of a scaling form in Eq. (2) yields $\beta\sim1.2$ as reported earlier [8]. Simulations show that although such an exponent obtains over a large range in reduced field [9], this value is not the true critical exponent [4,5]. As the rf field is applied, the depinning sharpens considerably as seen in curve *b*; R_d still decreases with

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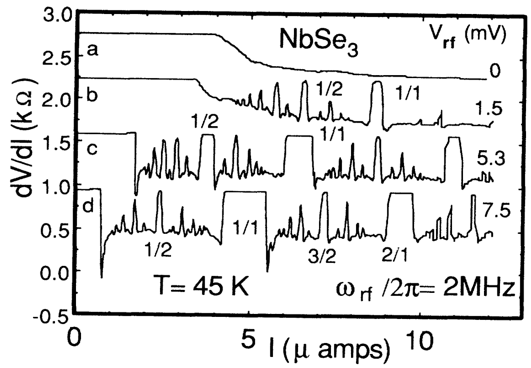

FIG. 1. Current dependence of the differential resistance dV/dI ($=R_d$) at various values of the rf field. The ordinate corresponds to curve d. Other curves are shifted arbitrarily along the ordinate for clarity. Note the negative differential resistance at threshold in curve d.

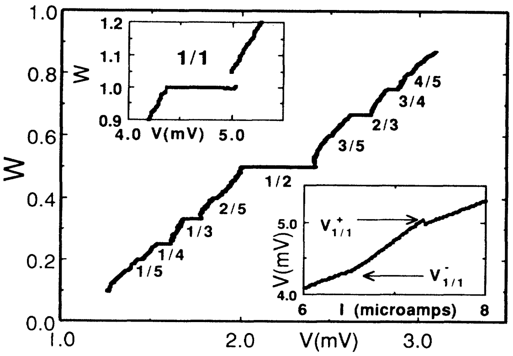

FIG. 2. Voltage dependence of the winding number W obtained from a direct measurement of the narrow band noise frequency. Negative dV/dI occurs at the upper step edges for 1/3 and 1/2 steps and a jump discontinuity on the upper edge of the 2/3 step. The lower inset shows the I-V curve for the 1/1 step; note the negative jump in V at the upper edge. The upper inset shows W vs voltage for the same step obtained from the dc I-V curve.

the same sign of the curvature and mode-locking features appear. In curve c, at a larger rf field, downward wings appear at threshold and at low order mode-locked steps. Such curves have been obtained in many studies [10], but have not been subjected to a scaling analysis, which we will describe below. We note here that the presence of the downward wings implies that Eq. (2) will yield $\beta < 1$. However, even more remarkably, we find that R_d becomes negative at threshold (hereafter referred to as the 0/1 step) and nearly so at the upper edge of the 1/1 step upon further increase in rf amplitude as shown in curve d. Note that a negative R_d in a current-biased measurement is a signature of a "switch," i.e., a discontinuous change in a voltage-biased configuration that is typically observed in the so-called switching samples near threshold without any rf field [11].

A direct study of the time-averaged CDW velocity is obtained through the NBN frequency [12], as shown in Fig. 2, where the winding number W is plotted versus the dc voltage, covering some of the subharmonic steps. The modulation signal is absent in this configuration and the dc current increments are smaller than in Fig. 1 and thus a clearer identification of higher order mode-locked steps is possible. The striking result is that as the dc current is increased monotonically, such a negative R_d appears at subharmonics $1/3^+$, $1/2^+$ in the form of an "overhang" in W and a jump discontinuity occurs at $2/3^+$. The lower inset shows the dc I-V curve with a pronounced negative discontinuity at the upper edge of the 1/1 step. The upper inset shows W near this step. In this case W is obtained from the dc I-V curve [13]. For both the harmonic and subharmonic steps, the switch is strongly asymmetric, observed more markedly at the upper edges.

Thus we observe, as a function of the rf amplitude, different behavior seen *in the absence of rf in different samples:* (1) the common form of a concave downwards R_d at threshold with a small rounding, shown here in curve a in Fig. 1, (2) an R_d with a downward wing in curve c that occurs at threshold, albeit very rarely, in some samples [14], and (3) a switch, i.e., a discontinuous or first order transition, not only at threshold but at both harmonic and subharmonic steps.

We have constructed a mode-locking phase diagram in Fig. 3, obtained from current-biased dc I-V curves in the presence of an rf field. It shows the Arnol'd tongues for the 0/1, 1/2, 1/1, and 2/1 steps. We compare the variation of the 0/1 step with that obtained from numerical simulations [15] of the elastic medium model for both 2D and 3D cases and find the latter to be a better agreement. The locked-unlocked phase boundary indicates continuous transitions for most of the phase space for all four steps. But a first order transition occurs for an intermediate range of rf amplitudes [16]. For both 1/1 and 1/2 steps, the first order transition occurs when the mode-locked region is the widest. Moreover, there is a large

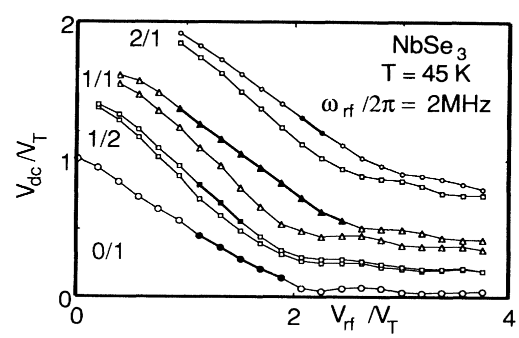

FIG. 3. Mode-locking phase diagram. The open and closed symbols, respectively, correspond to continuous and discontinuous transitions across the locked-unlocked phase boundary.

asymmetry between the upper and lower edges of the steps [17]. We emphasize that while the overall variation of shape and width of the various Arnol'd tongues is described well by the available models [3,15], they account for neither the first order transition nor the remarkable asymmetry.

Finally, we return to the scaling behavior near the mode-locked steps. Figure 4 shows scaling plots of the form in Eq. (3) for a few steps, obtained from the dc I-V curves. The horizontal segments in the figure represent the various mode-locked steps. Recent renormalization group analysis [6] has yielded a value of the exponent $\beta = 1 - \varepsilon/6$ where ε is $4-d$, yielding $\beta = 0.83$ in 3D and 0.67 in 2D, in agreement with numerical simulations [4]. Comparison with lines drawn corresponding to these two values shows that the 3D exponent is a better fit, also consistent with the agreement with a 3D simulation of an automaton model [15], shown in Fig. 4. Note also the large increase in the scaling range over previous studies [8], a mark of the dramatic decrease in the rounding of the transition. Thus we conclude that *in regimes where downward wings are observed in R_d, but R_d itself does not become negative, the data are consistent with theoretical results for a 3D system* [18]. The single-particle regime, where $\beta = \frac{1}{2}$, presumably remains outside the resolution of these studies. We note an interesting hierarchy of the various scaling regimes for mode-locked steps with different order. Consider, e.g., the $1/2^{\pm}$ steps. The apparent scaling regime includes all higher order steps,

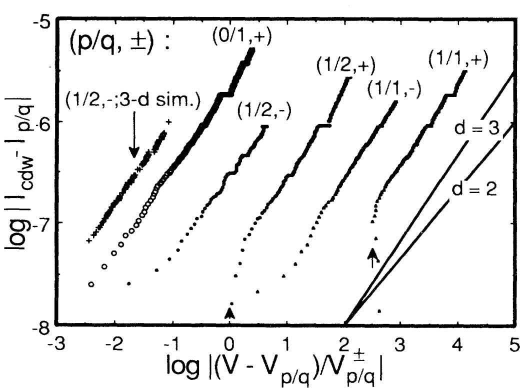

FIG. 4. Scaling plots for low order mode-locked steps for an rf frequency of 2 MHz. The abscissa corresponds to the 0/1 step. V_{rf}/V_T is 2.05 for the $0/1^+$ and $1/1^{\pm}$ steps and 1.12 for the $1/2^{\pm}$ steps. Each curve is shifted up from the previous one by one decade each in reduced field. Results of a simulation of a 3D automaton model are shown for comparison; here the abscissa is shifted down by two decades in the reduced field and the ordinate is in arbitrary units. The upward arrows mark the locations of (weakly) first order transitions. The horizontal sections in each curve correspond to various mode-locked states. The straight lines mark the results of Narayan and Fisher (Ref. [5]), which are the same as the numerical results of Tang and Bak (Ref. [19]) for the "sandpile" automaton model.

i.e., larger q values, with only local departures from the power-law fits. However, the scaling plot clearly breaks down near a lower order, i.e., the 1/1 or 0/1 step. Similarly, for the $1/1^-$ step, scaling extends over a larger regime until the 0/1 step is approached. Numerical simulations show very similar results [3,15].

Even for weakly first order transitions, an apparent scaling behavior obtains, as shown in Fig. 4 for the $1/1^+$ and $1/2^+$ steps. Allowing the transition voltage to vary, we obtain a similar scaling plot but with a transition voltage different from the step edge. A preemptive first order transition, shown by the arrow, occurs close to, but distinctly away from, this continuous transition point; this is similar to weakly first order transitions in conventional phase transitions. The difference ΔV between the actual first order point and the incipient second order point is a measure of the strength of the first order transition; e.g., for the data in Fig. 4 this difference in reduced voltage $(\Delta V/V^+_{p/q})$ is 3×10^{-2} and 1×10^{-2} for the $1/1^+$ and $1/2^+$ steps, respectively.

These results have many implications for the available theory. On several issues, the theory based on a deformable elastic medium approach agrees with our data. (1) In some regimes we observe a critical exponent close to what is predicted by the renormalization group analysis and the numerical simulations [3–5]. The same apparent exponent obtains even for weakly first order transitions. (2) The apparent scaling regime with $\beta = 1.2$ disappears in both theory [3,15] and experiment. We also find, in agreement with numerical simulations [3], that the scaling regime with a single exponent is greatly expanded when an rf field is applied. (3) Our results also confirm recent simulations [3] which show that a single exponent describes the approach to other mode-locked steps, i.e., Eq. (3) is valid in some parts of the phase diagram. It is particularly interesting to note that the same exponent is obtained [19] in the original "sandpile" automaton model proposed in the context of self-organized criticality. This study may thus also represent an experimental observation of that critical point.

There remain several serious differences with the standard elastic medium model, nevertheless. (1) The physical origin of the initial sharpening of the depinning transition and the consequent change in the apparent exponent in the presence of the rf field remain a mystery. Varying the rf amplitude changes the scaling behavior as well as the scaling regime. Thermal effects may cause the rounding seen experimentally [20]. Whether this is altered in the presence of an rf field needs to be addressed by both theory and simulations. (2) Within the elastic medium model, first order transitions do occur when phase slips are allowed, an issue emphasized by Coppersmith [6]. The nonmonotonicity of the first order transitions makes it difficult to understand why this occurs only for intermediate rf amplitudes and does not occur (or at least occurs more weakly) at both larger and smaller amplitudes. (3) In addition, the remarkable asymmetry between the upper and lower edges of the mode-locked steps is an issue that theory needs to address. (4) A switch, i.e., a first order transition has recently been addressed by a modification of the standard model by Levy *et al.* [7] who postulate a global coupling between the uncondensed electrons and the CDW condensate. It remains to be seen if this model could explain these results.

To conclude, we have shown that upon the application of an rf field, dramatic changes in the CDW transport characteristics occur. We find continuous depinning of one kind changing to another, with different qualitative behavior and also, most surprisingly, to a first order transition, a behavior commonly called a switch. Since different types of behavior are seen in a single sample at a fixed temperature with a fixed realization of quenched disorder, a successful theory ought to be able to unify them under a common conceptual framework.

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[1] *Charge Density Waves in Solids*, edited by L. P. Gorkov and G. Gruner (Elsevier, New York, 1989).

[2] D. S. Fisher, in *Nonlinearity in Condensed Matter*, edited by A. R. Bishop, D. K. Campbell, P. Kumar, and S. E. Trullinger (Springer-Verlag, Berlin, 1987); Phys. Rev. Lett. **50**, 1486 (1983); Phys. Rev. B **31**, 1396 (1985). The exponent β in Eq. (2) has been denoted as ζ in earlier work.

[3] A. A. Middleton, O. Biham, P. Sibani, and P. B. Littlewood, Phys. Rev. Lett. **68**, 1586 (1992).

[4] A. A. Middleton, Ph.D. thesis, Princeton University, 1990; C. R. Myers, Ph.D. thesis, Cornell University, 1991; A. A. Middleton and D. S. Fisher, Phys. Rev. Lett. **66**, 92 (1991).

[5] O. Narayan and D. S. Fisher, Phys. Rev. Lett. **68**, 3615 (1992); Phys. Rev. B **46**, 11 520 (1992).

[6] S. Coppersmith, Phys. Rev. Lett. **65**, 1044 (1990).

[7] J. Levy, M. S. Sherwin, F. F. Abraham, and K. Wiesenfeld, Phys. Rev. Lett. **68**, 2968 (1992). Also see P. B. Littlewood, Solid State Commun. **65**, 1347 (1988).

[8] S. Bhattacharya, M. J. Higgins, and J. P. Stokes, Phys. Rev. Lett. **63**, 1503 (1989); M. P. Maher, T. L. Adelman, J. McCarten, D. A. DiCarlo, and R. E. Thorne, Phys. Rev. B **43**, 9968 (1991).

[9] P. Sibani and P. B. Littlewood, Phys. Rev. Lett. **64**, 1305 (1990).

[10] See, for example, S. E. Brown and A. Zettl in Ref. [1]. For an earlier study of the phase diagram, see S. Bhattacharya, M. J. Higgins, and J. P. Stokes, Phys. Rev. B **38**, 7177 (1988). Also see R. E. Thorne, Ph.D. thesis, University of Illinois, Urbana-Champaign, 1987; J. Levy and M. Sherwin, Phys. Rev. Lett. **67**, 1846 (1991).

[11] Typical experiments on switching samples show a hysteretic jump in current in a voltage-biased experiment. In a current-biased configuration, discontinuities in voltage and/or negative differential resistance are seen. We do not distinguish between them (which may depend on details such as the amplitude of modulation in an ac dV/dI measurement) and refer to both as switching. See M. S. Sherwin, A. Zettl, and R. P. Hall, Phys. Rev. B **38**, 13028 (1988), and references therein. For theoretical work on switching, see M. Inui and S. Doniach, Phys. Rev. B **35**, 6244 (1987); S. Strogatz, C. M. Marcus, R. Westervelt, and R. E. Mirollo, Phys. Rev. Lett. **61**, 2380 (1988).

[12] S. Bhattacharya, M. J. Higgins, J. P. Stokes, and R. A. Klemm, Phys. Rev. Lett. **59**, 1849 (1987).

[13] Since the sample mode locks completely, simultaneous measurement of the NBN frequency determines the proportionality constant relating $\omega_{\rm nbn}$ to $I_{\rm cdw}$ and thus W can be obtained from the dc I-V curve. This is useful near the harmonic steps where the direct rf signal and its harmonics dominate the spectrum analyzer response.

[14] R. E. Thorne, J. R. Tucker, and J. Bardeen, Phys. Rev. Lett. **58**, 828 (1987).

[15] A. A. Middleton (unpublished).

[16] We find that with varying rf amplitude the switch varies in strength and thus we set an experimental criterion for a switch as either an overhang in $I_{\rm cdw}$ or a measurable discontinuous drop in V at the step edge upon increasing I.

[17] Note that we cannot rule out the occurrence of first order transitions at the lower step edges in other samples. But the asymmetry between the two step edges may persist.

[18] A comparison with a study of finite-size effects [J. McCarten, D. A. DiCarlo, M. P. Maher, T. L. Adelman, and R. E. Thorne, Phys. Rev. B **46**, 4456 (1992)] shows agreement in the threshold field for a sample of comparable R residual resistivity ratio. Our sample also shows sharp thresholds typical of 3D samples. But the thickness (0.8 μm) would belong to the 2D regime in this study which estimates the transverse correlation length to exceed 20 μm. The reason for this apparent discrepancy is not known.

[19] C. Tang and P. Bak, Phys. Rev. Lett. **60**, 2347 (1988).

[20] A. A. Middleton, Phys. Rev. B **45**, 9465 (1992); M. O. Robbins, J. P. Stokes, and S. Bhattacharya, Phys. Rev. Lett. **55**, 2823 (1985).