PHYSICAL REVIEW B VOLUME 43, NUMBER 12 15 APRIL 1991-II

Size effects, phase slip, and the origin of $f^{-\alpha}$ noise in NbSe$_3$

M. P. Maher, T. L. Adelman, J. McCarten, D. A. DiCarlo, and R. E. Thorne
Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University, Ithaca, New York 14853

(Received 26 November 1990)

The threshold electric field for charge-density-wave (CDW) depinning in NbSe$_3$ varies inversely with crystal thickness. Since the thickness of nearly all NbSe$_3$ crystals varies in steps across their width, a macroscopic inhomogeneity in CDW pinning results. We show that CDW phase slip occurring along thickness steps provides the dominant source of broadband noise in NbSe$_3$, and is responsible for complicated narrow-band noise spectra and mode-locking behavior. In crystals with no thickness steps, the evidence for scaling behavior of the CDW current near threshold is not convincing.

Voltage or current oscillations are among the most remarkable phenomena associated with motion of charge-density waves (CDW's) in NbSe$_3$ and related materials.[1] For dc excitation above the threshold for CDW sliding, two types of oscillations are observed: (1) coherent oscillations,[2] commonly referred to as narrow-band noise (NBN), whose fundamental oscillation frequency ω_n is proportional to the dc CDW current density J_{CDW}; and (2) incoherent oscillations or broadband noise (BBN),[2–5] whose power spectrum has the form $P(\omega)\sim\omega^{-\alpha}$, with $0.4\leq\alpha\leq 0.8$.

In the absence of CDW phase slip, the time-averaged current density (and CDW velocity) will be spatially uniform, and CDW motion in response to a dc bias is expected to be periodic.[6] The spectral width of the NBN fundamental should then be extremely small, and no steady-state BBN is expected. Experimentally, the NBN peaks are almost always broad ($Q=\omega_n/\Delta\omega_n\approx 1$–$20$) and show substantial structure, and large-amplitude BBN is observed. These effects are extremely sample dependent, but there is a general correlation between the width of the NBN and the amplitude of the BBN.

As in metals and semiconductors, the mechanism of BBN generation is of considerable interest. The noise power $\delta V^2/V^2$, where δV is the noise voltage, has been shown to scale with the sample volume,[3,5] suggesting a bulk origin. BBN has thus been attributed to fluctuations of the CDW phase resulting from interaction with random impurities.[3–5,7]

Here we show that most BBN in NbSe$_3$ does not have a bulk origin, and that CDW phase slip plays a central role in its generation. BBN, complicated NBN spectra, and incomplete mode locking are a consequence of CDW velocity shear which occurs along steps in crystal thickness. This shear drastically affects the form of the I-V characteristics, and thus has important consequences for study of any critical behavior near threshold.

Single crystals of NbSe$_3$ prepared by vapor transport have the form of long ribbonlike whiskers. Crystal lengths can be several centimeters, average thicknesses vary from 0.1 to 10 μm, and widths are typically 10 times larger than the thickness. In undoped NbSe$_3$, the threshold field E_T for CDW depinning varies inversely with thickness t for t less than a few micrometers, and gradually becomes t independent in very thick crystals.[8,9] This size dependence has been attributed[9,10] to a crossover from three-dimensional (3D) to 2D weak pinning, which occurs when t becomes comparable to, or smaller than, the bulk (3D) transverse CDW phase-phase correlation length. Recent high-resolution x-ray measurements[11] indicate transverse correlation lengths of a few micrometers in undoped NbSe$_3$, consistent with this interpretation.

For most NbSe$_3$ crystals, the crystal cross section remains constant in size and shape over lengths of at least several millimeters, much longer than the sample lengths used in transport measurements. However, for essentially all crystals, the cross-section shape is highly irregular. As illustrated in the inset in Fig. 1, the crystal thickness changes in a series of steps running along the whisker axis, with step heights that can be comparable to the average thickness. These thickness variations must result in pin-

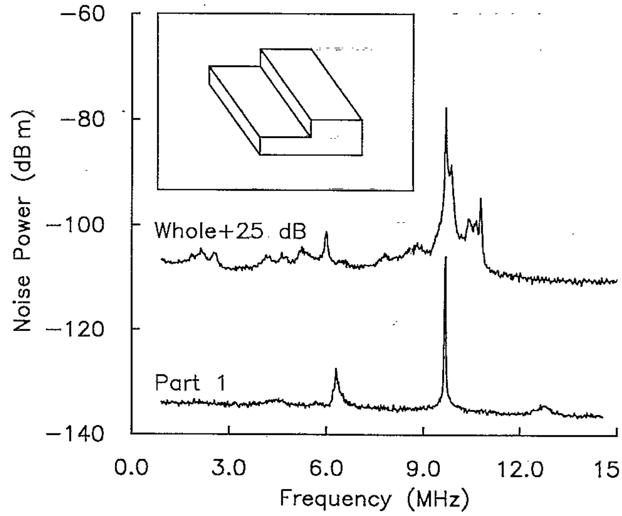

FIG. 1. Narrow-band noise fundamental spectral peaks for the whole and for part 1 of sample A. The spectral width in part 1, which had a nearly rectangular cross section, is much smaller. The inset schematically illustrates the crystal studied, whose cross section had one large step in its thickness running parallel to the whisker axis. The crystal was split along the step to produce parts 1 and 2.

ning-force variations within the crystal cross section. When an electric field is applied, these macroscopic pinning-force variations will in turn result in strain along the thickness steps. If this strain is large enough, the CDW will tear by phase slip along the steps, and CDW velocity shear will occur.

To investigate the effects of thickness steps on CDW transport in $NbSe_3$, we have performed a very simple experiment. A high-purity $NbSe_3$ crystal (residual resistance ratio of 200) having one large step is selected and cut in half lengthwise (i.e., the cut is made perpendicular to whisker axis). One of these halves is then carefully split along the step. This procedure yields one crystal with an irregular cross section (referred to as the "whole"), and two crystals with more nearly rectangular cross sections (referred to as parts 1 and 2). Electrical contacts were applied far from the cut ends, to avoid any damaged regions. Measurements of the NBN, BBN, and differential resistance were made at $T=125$ K on several samples prepared in this way, with similar results in each case. Data presented here are for two representative samples whose parts had the following lengths, maximum widths, and average thicknesses: sample A: 0.96 mm×4.4 μm×0.95 μm for part 1; 1.2 mm×3.4 μm×1.12 μm for part 2; and 0.9 mm×7.8 μm×1.0 μm for the whole; sample B: 0.7 mm×33 μm×1.5 μm for part 1; 0.8 mm×25 μm×2.2 μm for part 2; and 1.4 mm×57 μm×2.0 μm for the whole.

Figure 1 shows the fundamental NBN spectral peaks for part 1, whose cross section was nearly perfectly rectangular, and for the whole of sample A. The whole's fundamental is very broad and shows substantial structure. Part 1's fundamental has one extremely sharp peak ($Q \approx 350$) and a second much smaller peak at lower frequency. In samples with large thickness differences between parts 1 and 2, the peaks in the whole's fundamental are well separated and can be unambiguously identified as arising from part 1 or 2. Since ω_n is proportional to the CDW velocity and current density, this implies that the CDW moves with different time-averaged velocities in different parts of the whole's cross section. CDW phase slip must then occur along the thickness step to allow for this velocity shear.

Figure 2 shows the normalized broadband noise power per unit volume versus frequency for sample B. The NBN fundamental frequency was set at 50 MHz, to ensure that the current densities in each crystal were approximately the same, and that no NBN was within the BBN measurement window. The BBN per unit volume in the whole is more than 2 orders-of-magnitude larger than in either of the parts; similar behavior was observed for sample A. This clearly indicates that the boundary between the whole's parts—the thickness discontinuity—is entirely responsible for the difference in BBN amplitudes. BBN in $NbSe_3$ is thus associated primarily with phase slip along thickness discontinuities, and is not a bulk effect.

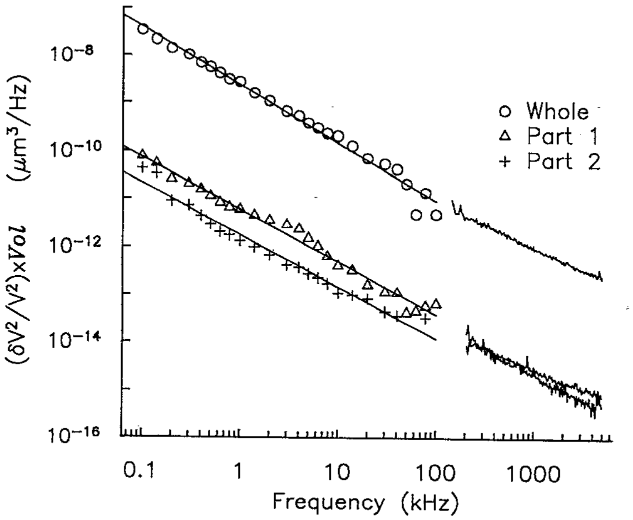

FIG. 2. Normalized broadband noise power per unit volume vs frequency for sample B. The more than 2 orders-of-magnitude difference between the whole and parts 1 and 2 is due to the thickness step in the whole's cross section. Data below and above 100 kHz were obtained using a lock-in amplifier and spectrum analyzer, respectively.

Figure 3 shows the differential resistance dV/dI of sample A measured in the presence of a 10 MHz ac voltage. Mode locking of the NBN frequency ω_n to the applied ac frequency ω_{ac} when $\omega_n/\omega_{ac}=p/q$ results in Shapiro steps on the dc I_{CDW}-V characteristics and corresponding peaks in dV/dI.[1] The mode locking is said to be complete when I_{CDW} is constant for some range of dc bias, so that dV/dI is equal to the Ohmic (single-particle) resistance measured below threshold. In Fig. 3, mode locking of the whole's 1/1 peak is only 65% complete, and few subharmonic peaks are visible. In contrast, the 1/2 and 1/1 peaks of part 1 lock completely, and subharmonics down to $p/q=1/7$ can be resolved. Since an incompletely locking crystal can be made to lock completely by making its cross section more rectangular, incomplete mode locking

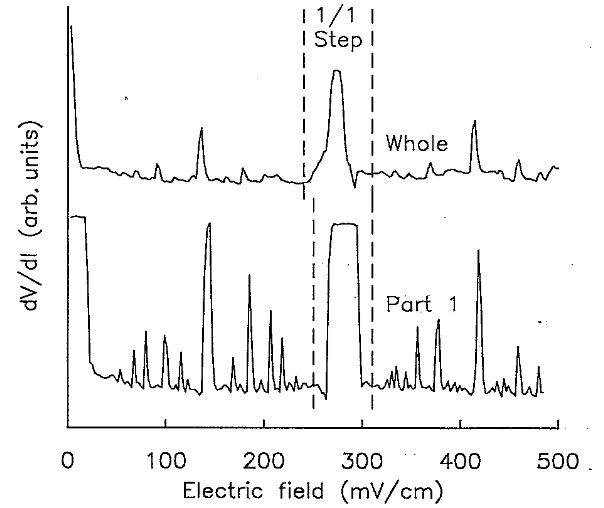

FIG. 3. Differential resistance for sample A vs dc bias, measured in the presence of a 10 MHz ac field. The ac amplitude was chosen to obtain the maximum mode-locked width of the 1/1 peak. The completeness of the mode-locking correlates with the uniformity of the crystal cross section.

can no longer be viewed[12] as basic to the CDW response in $NbSe_3$.

The ubiquity of large-amplitude BBN, complicated NBN, and incomplete mode locking in previous work on $NbSe_3$ is a consequence of the extreme rarity of crystals with rectangular cross sections. In growths prepared by standard methods, no such crystals can be found. With improved growth conditions, roughly one crystal in 10000 will have a rectangular cross section. Measurements on such a crystal were reported previously.[13] The NBN fundamental had a spectral Q of 30000 for $\omega_n = 200$ MHz; nearly 150 subharmonic steps with $p/q < 1/1$ and complete mode locking at frequencies up to 300 MHz were observed; and no BBN was measured. Based on these and other results, it was suggested[13] that the BBN, complicated NBN spectra and incomplete mode locking were a consequence of CDW velocity shear, and did not reflect the intrinsic CDW dynamics. The present results confirm this controversial interpretation and establish the origin of the velocity shear.

Our observation that most BBN in $NbSe_3$ does not have a bulk origin is not necessarily inconsistent with previous measurements of the volume dependence of the BBN. The BBN amplitude from a single thickness step should roughly scale with the cross-sectional area beneath the step. The number of steps scales roughly with the crystal width, so that the BBN power should be proportional to the crystal volume.

Large-amplitude BBN has been observed in TaS_3, $K_{0.3}MoO_3$, and other CDW conductors.[1] TaS_3 crystals have even more irregular cross sections than $NbSe_3$ crystals and exhibit a similar size dependence of E_T, so that a similar BBN generation mechanism is expected. More generally, BBN should result whenever there is CDW velocity shear. In $K_{0.3}MoO_3$, where size-dependent pinning may be less important, such shear arises from the large electrical anisotropy, unfavorable crystal geometries, high contact resistances, and/or inclusions of other crystalline phases.

The precise mechanism by which velocity shear along steps in thickness produces BBN is unclear. However, many qualitative features are reproduced by a simple model for two coupled overdamped CDW's moving in periodic potentials of slightly different strength, as given by

$$d\phi_i/dt = F - V_i \sin(\phi_i) + J \sin[(\phi_j - \phi_i)/\phi_0], \quad i = 1,2, \qquad (1)$$

where ϕ_i and ϕ_j are the phases of the two CDW's, F is the applied field, V_i are the pinning strengths, J is the coupling strength, and ϕ_0 determines the minimum phase difference for phase slip. As the coupling J between the CDW's is increased (with F, V_1, V_2, and ϕ_0 fixed), the spectrum of the response progresses from two distinct NBN peaks with no BBN to two broadened peaks with BBN to a single NBN peak with no BBN. A more complete model which includes the effects of many thickness steps, of thermal processes in phase slip, and of relaxation of internal CDW modes following phase slip seems likely to reproduce much of the complex time, temperature, and frequency dependence of the NBN and BBN reported[3–5,12,14] for nonrectangular crystals.

The present results have important consequences for study of CDW transport near threshold. Fisher has suggested[15] that CDW depinning is a dynamical critical phenomenon, and has predicted that the CDW current (velocity) should vary with applied field E near threshold as

$$J_{CDW} \sim (E - E_T)^{\zeta}. \qquad (2)$$

A mean-field calculation gives $\zeta = 1.5$, while recent numerical simulations[16] give $\zeta \approx 1.16$ in 3D and 0.95 in 2D.

Previous measurements[1,17] of the J_{CDW}-E relation in $NbSe_3$ have yielded ζ values between 1.1 and 1.5, varying strongly with temperature and from sample to sample. The crystals used in these measurements were not screened for the quality of their NBN or mode locking, and almost certainly did not have rectangular cross sections. As indicated in Fig. 4 for sample A, velocity shear associated with thickness steps rounds the onset of CDW conduction and increases the value of ζ. For $NbSe_3$ crystals with nearly perfectly rectangular cross sections and with thicknesses such that the static pinning is believed to be 2D, the onset of CDW conduction at threshold is abrupt[13] (dV/dI exhibits a discontinuous drop at E_T, reminiscent of behavior expected for a single-coordinate CDW) and the data above $(E-E_T)/E_T \approx 0.1$ are well described by Eq. (2) with $\zeta \approx 1.09$, in between the 3D and 2D predictions. However, it is far from clear that this power-law behavior has anything to do with critical phenomena: The ζ value is very close to 1, the value which must be obtained at high fields. At fields closer to threshold, power-law fits with a single exponent cannot be made over any significant field range.

Recently, it has been suggested[18] that fluctuations of the CDW pinning strength resulting from random impurity distributions will lead to unbounded local strains in sufficiently large systems when electric fields are applied. These strains will result in phase slip, destroying the critical behavior near threshold and producing BBN. However, simple estimates suggest that phase slip by this mecha-

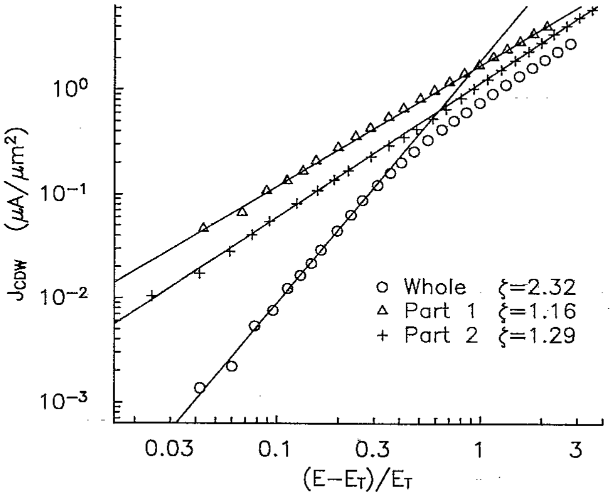

FIG. 4. CDW current density vs reduced electric field for sample A. The solid lines indicate fits to Eq. (2). The power-law exponent ζ is larger in crystals with irregular cross sections.

nism is unlikely to be important in NbSe$_3$. Since the dynamical CDW correlation length is expected to be much longer than the static correlation length in the critical regime, and since typical NbSe$_3$ crystals are only 10–100 static correlation lengths long, we suggest instead that any scaling regime is eliminated by finite-size effects.

In conclusion, we have shown (1) that BBN in NbSe$_3$ arises primarily[19] from macroscopic pinning-force variations and the resulting CDW velocity shear, and can be viewed as a finite-size effect; and (2) that crystals with rectangular cross sections must be used to evaluate the intrinsic dynamics of sliding CDW's. Crystals with a single thickness step should provide convenient systems for quantitative study of $f^{-\alpha}$ noise and CDW phase slip.

The authors appreciate fruitful discussions with C. Myers, J. P. Sethna, and A. Middleton. Two of us (M.P.M. and D.D.) gratefully acknowledge support provided by IBM and by the National Science Foundation (NSF), respectively. This work was supported by the Alfred P. Sloan Foundation, by the AT&T Foundation, and by NSF Grant No. DMR-89-58515.

---

[1] For comprehensive reviews of CDW's, see P. Monceau, *Electronic Properties of Quasi-One-Dimensional Materials* (Reidel, Dordrecht, 1985), part II, p. 139; G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988).

[2] R. M. Fleming and C. C. Grimes, Phys. Rev. Lett. **42**, 1423 (1979).

[3] J. Richard, P. Monceau, M. Papoular, and M. Renard, J. Phys. C **15**, 7157 (1982).

[4] A. Zettl and G. Grüner, Solid State Commun. **46**, 29 (1983).

[5] S. Bhattacharya, J. P. Stokes, M. O. Robbins, and R. A. Klemm, Phys. Rev. Lett. **54**, 2453 (1985).

[6] P. B. Littlewood, Phys. Rev. B **33**, 6694 (1986); J. Bardeen (private communication.)

[7] J. Richard, P. Monceau, and M. Renard, Phys. Rev. B **25**, 948 (1982); J. R. Tucker, *ibid.* **40**, 5447 (1989).

[8] P. J. Yetman and J. C. Gill, Solid State Commun. **62**, 201 (1987). Size dependence of E_T is also observed in TaS$_3$; see D. V. Borodin, F. Ya. Nad', S. Savitskaya, and S. V. Zaitsev-Zotov, Physica **143B**, 73 (1986).

[9] J. McCarten, M. Maher, T. L. Adelman, and R. Thorne, Phys. Rev. Lett. **63**, 2841 (1989).

[10] For a discussion of alternative origins of the size dependence of E_T, see J. C. Gill, Phys. Rev. Lett. **65**, 271 (1990); R. E. Thorne and J. McCarten, *ibid.* **65**, 272 (1990).

[11] E. Sweetland, C.-Y. Tsai, B. A. Wintner, J. D. Brock, and R. E. Thorne, Phys. Rev. Lett. **65**, 3165 (1990).

[12] R. P. Hall, M. F. Hundley, and A. Zettl, Synth. Met. **19**, 813 (1987); S. Bhattacharya, M. J. Higgins, and J. P. Stokes, Phys. Rev. B **38**, 7177; 10093 (1988).

[13] R. E. Thorne, J. S. Hubacek, W. G. Lyons, J. W. Lyding, and J. R. Tucker, Phys. Rev. B **37**, 10055 (1988); R. E. Thorne *et al.*, *ibid.* **35**, 6348; **35**, 6360 (1987).

[14] S. E. Brown and L. Mihaly, Phys. Rev. Lett. **55**, 742 (1985); S. Bhattacharya, J. P. Stokes, M. J. Higgins, and R. A. Klemm, *ibid.* **59**, 1849 (1987); Phys. Rev. B **40**, 5826 (1989).

[15] D. S. Fisher, Phys. Rev. B **31**, 1396 (1985).

[16] P. Sibani and P. B. Littlewood, Phys. Rev. Lett. **64**, 1305 (1990). Also see H. Matsukawa, J. Phys. Soc. Jpn. **57**, 3463 (1988).

[17] P. Monceau, M. Renard, J. Richard, M. C. Saint-Lager, and Z. Z. Wang, in *Charge Density Waves in Solids,* edited by Gy. Hutiray and J. Solyom (Springer-Verlag, Berlin, 1985), p. 279; J. C. Gill, *ibid.*, p. 377; S. Bhattacharya, M. J. Higgins, and J. P. Stokes, Phys. Rev. Lett. **63**, 1503 (1989).

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

[19] Most BBN studies of the q_1 CDW in NbSe$_3$ have been performed at temperatures between 90 and 135 K. The BBN generation mechanism outlined here dominates in this temperature range. However, between 90 and 60 K, the NBN spectrum broadens dramatically and the BBN increases, even for those crystals with rectangular cross sections and extremely coherent NBN at higher temperatures. This increase may be associated with the rapid increase in strains required for phase slip at current contacts with decreasing temperature.