Switching and Phase-Slip Centers in Charge-Density-Wave Conductors

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We demonstrate that switching in the charge-density-wave (CDW) conductors Fe_xNbSe_3 and $NbSe_3$ is associated with a division of the CDW condensate into distinct macroscopic regions with independent CDW depinning fields and independent CDW drift velocities. These well-defined regions are separated by localized phase-slip centers. We explicitly determine the location within the crystalline bulk, and the spatial extent, of the phase-slip interface. Switching results from initial phase breaking at either side of the phase-slip center.

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The dramatic nonlinear electrical conductivity observed in the linear-chain compound $NbSe_3$ and related materials is a consequence of depinning and subsequent Fröhlich motion of the collective-mode charge-density-wave (CDW) condensate.[1] The depinning process at the threshold field E_T can be either smooth, or so sharp that an actual (hysteretic) jump or "switching" occurs in the I-V characteristics.[2] Additional switchings may also occur for $E > E_T$. In certain materials switching may be enhanced by chemical doping (e.g.,[3] Fe in $NbSe_3$) or by irradiation damage (e.g.,[4] 2.5-MeV electrons in $K_{0.3}MoO_3$). This feature has suggested that switching may be a strong-impurity–based effect.

In this Letter we demonstrate that switching is associated with a division of the CDW crystal into macroscopic regions, each with an independent depinning field and independent CDW phase velocity. The interface between the regions comprises a spatially localized entity, which we identify as a phase-slip center in analogy to sites of order-parameter collapse in superconducting microbridges.[5] Our experiments allow a precise determination of the size and location of the individually phase-coherent regions within the sample, and consequently also the size and location of the phase-slip centers themselves. Switching is shown to correspond to the onset of phase slip at either side of a given phase-slip interface.

Materials used in these experiments consisted of single crystals of Fe_xNbSe_3 ($x \approx 0.03$) and $NbSe_3$, both produced by conventional vapor-transport methods and of typical dimensions 2.5 mm×2 μm×3 μm. Our experimental setup employed a unique four-terminal probe with which we could nonperturbatively measure dc conductivity in different regions of the sample, and simultaneously observe the narrow-band noise spectrum.[6] Figure 1(a) shows schematically the probe arrangement. Current leads were attached to the ends of the crystal (terminals 1 and 4) with silver paint. Two additional noninvasive voltage-sensing probes (terminals 2 and 3) were formed from fine gold or Constantan wires, which could be independently translated along the length of the crystal by external micrometer screws. Repeatability for the absolute probe positions was generally better than ± 5 μm.

Figure 1(b) shows I-V characteristics of Fe_xNbSe_3 at $T = 42$ K. The top trace represents the I-V trace for the "whole" crystal, measured between terminals 1 and 4. Two switches, S1 and S2, are clearly observed at bias currents $I_{S1} = 130$ μA and $I_{S2} = 160$ μA, respectively. The three lower displaced traces in Fig. 1(b) represent the I-V characteristics of different segments of the crystal, obtained with voltage probe posi-

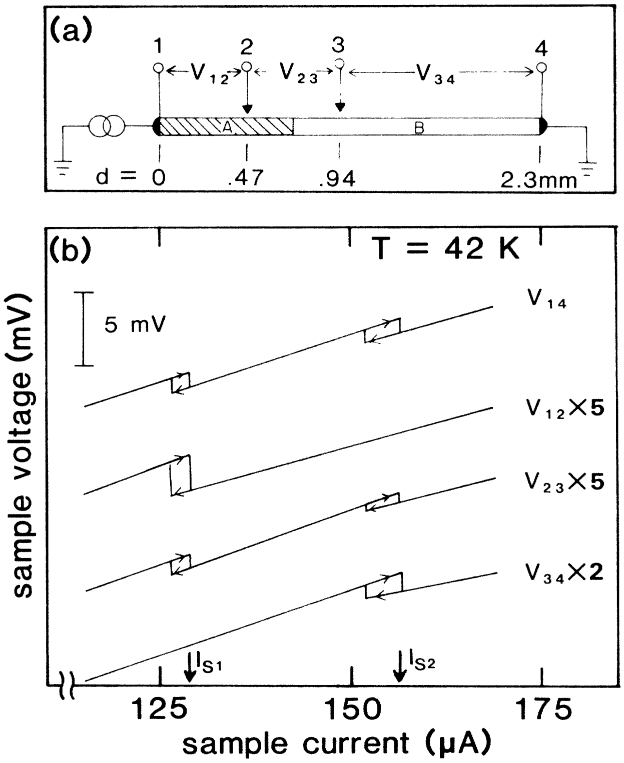

FIG. 1. (a) Voltage probe configuration for I-V traces shown in (b). (b) Simultaneously recorded I-V traces for different segments of a single Fe_xNbSe_3 crystal.

tions as indicated in Fig. 1(a). It is apparent that, at the onset I_{S1} of nonlinear conduction for the entire sample, a uniform I_{CDW} is *not* observed throughout the sample. In particular, segment 3–4 (i.e., that crystal segment lying between probes 3 and 4) remains entirely Ohmic beyond I_{S1} (and hence the CDW in that segment remains pinned), until I_{S2} is exceeded. Furthermore, at I_{S2}, no anomaly is observed in the I-V characteristics of segment 1–2. Only two switches, at I_{S1} and I_{S2}, were observed for this crystal.

It is apparent that the sample used for Fig. 1 is at 42 K divided into different regions with different depinning fields E_T, and that switching corresponds to initial CDW conduction in only a particular region of the crystal. By repositioning probes 2 and 3, we have determined that the crystal used for Fig. 1 contains only two independent regions, A and B, with a common interface located in the original segment 2–3. The two regions are identified in Fig. 1(a). The interface between regions A and B corresponds to a discontinuity in the advancement of the CDW phase ϕ, and we identify the interface zone as comprising a phase-slip center. To locate precisely the phase-slip center and to determine its spatial extent, the following I-V measurements were employed. Probe 3 was fixed in region B, and probe 2 was successively advanced from terminal 1 to points along region A, eventually traversing the phase-slip center. At each point an I-V trace was recorded for the segment between probes 2 and 3, with terminals 1 and 4 still serving as current leads. Figure 2 shows the voltage switch height h of S1 extracted from such I-V traces, as a function of position of probe 2 along the crystal. h decreases linearly with probe position, and a least-squares fit to the data places the boundary of region A at a position 776 ± 20 μm from terminal 1. The boundary of region B was similarly determined by fixing probe 2 in region A, moving probe 3 to different points in region B, and measuring the resulting switch height of S2. Results of this experiment are indicated as triangles in Fig. 2, and again a linear dependence on probe position is obtained. To within experimental accuracy, regions A and B abut along a common interface near 776 μm.

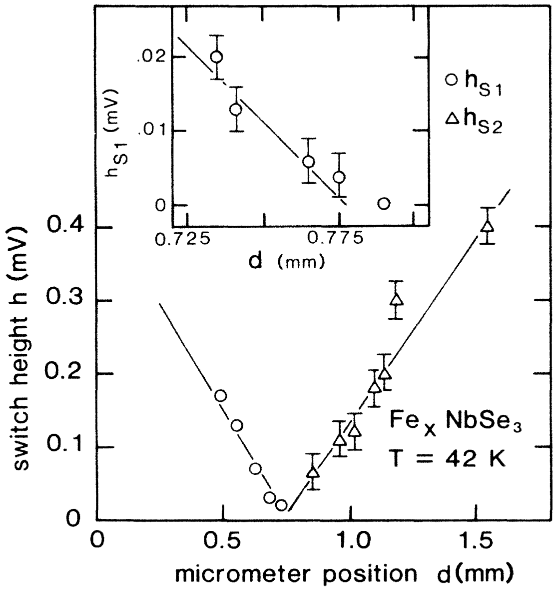

FIG. 2. Switch height h vs probe position for Fe$_x$NbSe$_3$. Inset: In greater detail, h_{S1} near the phase-slip region.

The actual width of the phase-slip interface is determined most accurately by investigation of the height of S1 versus small displacements of probe 2 near the phase-slip center, with probe 3 fixed in region B. The inset of Fig. 2 shows the results of this experiment. No deviation from linear behavior is apparent to the last nonzero datum point at 775 μm, while at 787 μm region A has clearly been overstepped. An upper bound for the full width of the phase-slip center is thus established; approximately 25 ± 20 μm.

In addition to the dc conductivity measurements discussed above, we have performed simultaneous measurements of the narrow-band noise spectrum. This method provides an independent determination of the CDW velocity distribution within the crystal, since the noise frequency is directly proportional to CDW velocity.[1] For $I < I_{S1}$ no noise was present, for $I_{S1} < I < I_{S2}$ a single frequency (with harmonics) was present, while for $I > I_{S2}$ two independent frequencies were present and identified as originating from regions A and B, respectively. These results again demonstrate that the interface between regions A and B, which corresponds to a zone of CDW velocity discontinuity, remains intact even after *both* regions A and B have depinned.

As might be inferred from samples which display numerous switches, more than one phase-slip center can exist within a given crystal. Figure 3(a) shows the total voltage-driven I-V characteristics (measured between terminals 1 and 4) for a NbSe$_3$ sample at $T = 29$ K. Ten distinct switches are observed. By placement of nonperturbative voltage probes on the sample, it was verified that the switches again correspond to different regions of the sample depinning independently. The corresponding narrow-band current noise spectrum for this NbSe$_3$ sample is shown in Fig. 3(b). Each successive switching corresponds to a new fundamental frequency entering (at finite frequency) the total spectrum. Of particular importance in Fig. 3(b) is that all observed noise fundamentals are truly independent, i.e., no peaks are related by a constant multiplier, and no low-frequency peaks correspond to difference (mixing) frequencies of higher-frequency peaks. In addition, we observed no tendency for noise peaks to lock together, even for those noise peaks originating from adjacent regions.

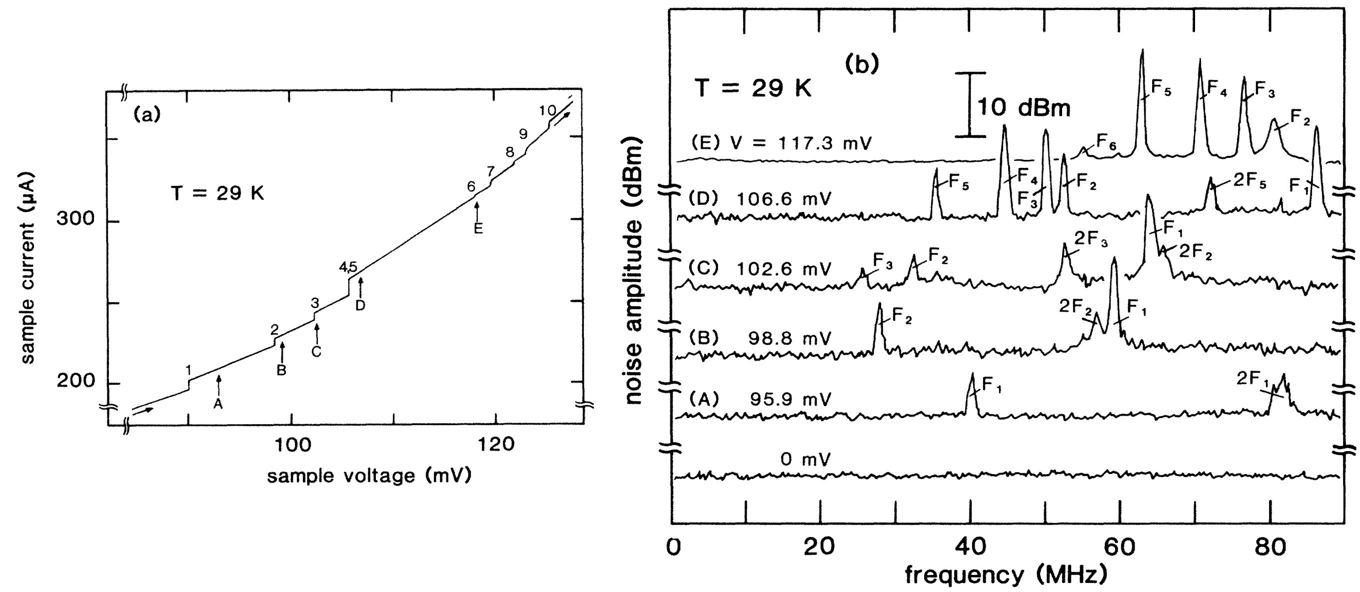

FIG. 3. (a) I-V trace for $NbSe_3$. Ten distinct switches are identified. The lettered arrows refer to bias values at which corresponding noise spectra of (b) were recorded. (b) Noise spectra for the $NbSe_3$ sample of (a). The peaks are identified with the switch at which the noise peak first appears.

Both Fe_xNbSe_3 and $NbSe_3$ display highly temperature-dependent switching behavior.[2,3] In the temperature regime of switching, the threshold fields for the onset of switching are roughly temperature independent, although the associated hysteresis grows with decreasing temperature.[7] For the samples studied here, switching was no longer observed above 50 K in Fe_xNbSe_3 or above 30 K in $NbSe_3$. The experiments described above were repeated at various temperatures within and above the switching regime. Near the upper temperature limit of the switching regime, the threshold fields for the different sample segments converged, and at temperatures above the switching regime, the smooth onset of nonlinear conduction was found to be associated with a *uniform* depinning of the entire CDW throughout the crystal (verified by both the I-V characteristics and narrow-band noise spectra). The formation of phase-slip centers is thus a highly temperature-dependent process, and it appears that, at higher temperatures, any existing phase-slip centers are eliminated, or at least rendered inactive.

Our experimental results disprove all models of switching proposed to date, including the CDW domain analogy to the kinetic Ising model by Joos and Murray,[8] underdamped classical rigid-particle motion,[9] or the "self-blocking" model of uniform CDW depinning by Jánossy, Mihály, and Mihály.[10] These models do not consider phase-slip phenomena, and our experiments clearly demonstrate that switching corresponds to the onset of CDW phase slip at either side of well-defined phase-slip centers in the crystalline bulk. We suggest that *amplitude* dynamics of the CDW condensate play an important role in the switching process. When the threshold field E_T is exceeded in a switching sample, excess CDW phase will be delivered to the phase-slip center by the adjacent depinned region with the greater CDW velocity. The excess phase can be relieved only if the CDW amplitude suffers periodic collapse at the phase-slip interface. The necessary "breaking" of the CDW amplitude at the phase-slip center introduces an additional effective pinning potential. Indeed, previous experiments[7] on $NbSe_3$ have shown that E_T for switching samples far exceeds that expected only on the basis of the (weak) impurity pinning potential in the crystal. This suggests that the additional energy term needed for amplitude suppression at the phase-slip center (which may be a special type of strong-impurity pinning site) may well dominate the conventional depinning energy normally associated with phase mode excitations. A similar effect has been suggested by Gill[11] to account for unusual behavior of E_T in $NbSe_3$, resulting from various contact configurations where the CDW is "broken" by the injected current.

To determine the magnitude of the additional energy term introduced by the phase-slip center, we have measured E_T for a $NbSe_3$ crystal before and after physically cutting away that part of the crystal containing a phase-slip center. The resulting change in E_T at 26 K was from 300 mV/cm (switching present) to 60 mV/cm (switching absent). On an atomic scale, the phase-slip pinning energy contribution is then (240 mV/cm)$a_0 e \approx 7\times 10^{-6}$ meV, roughly eight times greater than the minimum impurity pinning energy at 48 K.

The inevitable phase-slip process at the phase-slip centers suggests that some of the ideas developed for CDW dynamics near a metallic contact by Gor'kov,[12] and independently by Ong, Verma, and Maki,[13] may be applicable to switching. For switching in Fe_xNbSe_3 and $NbSe_3$, "one-dimensional" uniform amplitude collapse across the width of the sample, as suggested by Gor'kov, may be appropriate. However, we strongly emphasize that CDW dynamics at metallic contacts are fundamentally *very different* from CDW dynamics near phase-slip centers, for the simple reason that samples with attached metal contacts, but no phase-slip centers, do not display switching. Clearly, the precise nature of the CDW dynamics at the phase-slip interface is of critical importance in an understanding of switching-related CDW phenomena such as hysteresis,[2] bistability and $1/f$ noise,[14] and period-doubling routes to chaos.[9]

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