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Conduction delays in switching $NbSe_3$: Sensitive dependence on the initial configuration

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In switching charge-density-wave (CDW) conductors, a voltage pulse greater than threshold causes the CDW to slide only after a delay τ. For identical experimental conditions, we have found a class of delays as long as 5 s with fluctuations as large as 5 orders of magnitude. For large τ, the distribution of delays $N(\tau)\propto\tau^{-\gamma}$, with $0.8<\gamma<1.3$. We argue that τ measures the time at which the internal strain somewhere in the CDW exceeds the threshold for phase slippage. The electronic history of the sample, and hence the distribution of initial configurations of the CDW, determines $N(\tau)$.

Sliding charge-density-wave (CDW) conductors have now been established as models for the study of dissipative nonlinear dynamical systems with many equally important degrees of freedom.[1] The CDW in conventional samples depins smoothly at a threshold E_T. Classical models with many degrees of freedom[2] have been successful in explaining the critical behavior of the depinning in conventional samples,[3] hysteresis, and nonexponential relaxations of the CDW polarization,[4] and other observed phenomena. A central feature of these models is the existence of an exponentially large number of metastable states for a pinned CDW.[2]

Switching samples depin abruptly and hysteretically. Zettl and Grüner observed that, on applying current pulses larger than threshold, the CDW began to slide only after a time delay between 1 and 100 μsec, with fluctuations of smaller than 100% from pulse to pulse.[5] We report detailed measurements of the delays near threshold. We find a new class of long switching delays clearly separated from the shorter ones observed by Zettl and Grüner. These long delays may be of order seconds, and fluctuate up to 5 orders of magnitude from one pulse to the next for identical external experimental conditions.

Delayed transitions occur in many driven dynamical systems, from lasers[6] to convecting fluids.[7] However, we know of no physical system exhibiting delays with variability comparable to the long delays in switching CDW's. Our results rule out several theories of switching-CDW conduction. We propose the following mechanism: the switching delay is the time during which the CDW evolves from one of a large number of initial configurations to a configuration in which the internal strain is sufficiently large to tear the CDW.

Samples of freshly grown, nominally pure $NbSe_3$ were mounted in a standard two-probe configuration. The samples were cooled in a helium exchange gas to between 25 and 30 K in a temperature-controlled closed-cycle refrigerator. The rms temperature fluctuations were ±10 mK over an indefinite period of time. Details of the experimental procedure will be published elsewhere.[8]

In initial experiments we applied a train of square pulses to a sample and measured the switching delay for each pulse. For voltages V near the threshold V_c the first delay was between 1 ms and 100 ms, but every subsequent delay was of order 1 μs. The CDW began in an unpolarized state. The first pulse polarized the sample, and for every subsequent pulse the initial state of the sample was highly polarized. This behavior is reminiscent of the pulse sign memory effect.[9] The initial state of the sample in large part determines the switching delay time. For all data presented here, the remanent polarization was erased before each square voltage pulse with a 3-s erasing pulse (discussed below) of the form $V(t)=V_0(\frac{1}{2})(1-\cos\Omega t)\cos(2\pi f t)$, with $V_0=185$ mV $>2V_c$, $2\pi/\Omega=3$ s, and $f=1$ kHz.

Figure 1 shows the amplified CDW current response to four identical voltage pulses applied to a single sample. In order to use the full dynamic range of our digitizer, the ohmic current has been subtracted using a standard bridge circuit. Because the switching delays ranged from 1 μs to 1 s, the current was measured in logarithmic time intervals. The switching time was determined in software after each pulse. After the beginning of each pulse, a displacement current flows as the CDW polarizes, decreasing roughly logarithmically until the abrupt switch. The current traces are nearly identical before the abrupt switches. Thus the macroscopic CDW polarization $P=\int I_{\mathrm{CDW}}(t)dt$ just prior to a switch depends on the switching delay τ. *Switches do not always occur at the same macroscopic polarization of the CDW.*

In presenting distributions which vary over many orders of magnitude, logarithmic binning in a histogram $N'(w=\log\tau)$ is preferable to conventional linear binning in a histogram $N(\tau)$. Figure 2 shows the distribution of delays for a single sample under different experimental conditions. Figure 2(a) shows the shift of $N'(w)$ from long to short delays as V is increased above the threshold V_c. (V_c was defined as the voltage at which 50% of the delays were less than 1 s, 87.8 mV for these data. Changing the percentage criterion from 30% to 70% of the

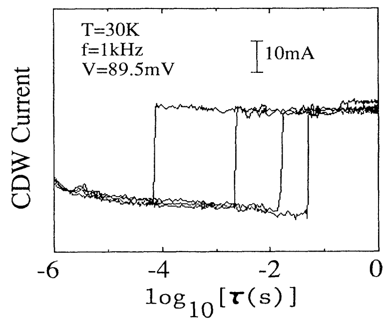

FIG. 1. CDW current response to four identical voltage pulses.

time shifted V_C less than $\pm 0.5\%$.) For the smallest voltage $V=88.6$ mV, the delays are between 100 μs and 1 s. At an intermediate value $V=89.4$ mV, the distribution is bimodal with a peak at a few microseconds, a gap between 10 μs and 100 μs, and a broader peak between 100 μs and 100 ms. For the highest value of $V=90.2$ mV, most of the weight is in the peak near a few microseconds.

Near V_c, the distribution $N(\tau)$ of long delays obeys a power law with a cutoff at short times. Figure 2(b) plots $P(w)=\log_b[N'(w)/10^w]$ for 4000 long delays at $V=89.3$ mV. It can easily be shown that, if $N(\tau)\propto\tau^{-\gamma}$, then $N'(w)\propto 10^{(1-\gamma)w}$ and $P(w)=-\gamma w+\text{cons}$. $P(w)$ in Fig. 2(b) is clearly well fit by a straight line *over at least 4 orders of magnitude*. A least-squares fit of a line to $P(w)$, with points weighted by $\sqrt{N'(w)}$ and including only points with $-0.02>w>-4.1$ (100 μs $<\tau<1$ s), gave us γ. The inset shows the variation of γ from 0.8 to 1.2 as V was varied from 88 to 90 mV (sufficiently close to V_C that few short delays appeared).

The form of $N'(w)$ depends critically on the erasing frequency f. For 50 Hz $<f<5$ kHz, delays were uncorrelated,[10] indicating that the erasing procedure was effective. With all other experimental parameters (including any thermal or other noise) fixed, the width of $N'(w)$ for the long delays drops from 4 to 2 orders of magnitude as f is increased from 50 Hz to 5 kHz.[8] For $f>5$ kHz, correlations develop between successive delays. The sensitive dependence of $N'(w)$ on f shows that external noise is not the dominant cause of fluctuations in τ, in conflict with the explanation of Joos and Murray.[11]

Figure 3 shows the dependence of the average $\langle\tau\rangle$ and the standard deviation σ on the pulse height V with $f=1$ kHz. Between $V=88$ mV and 92 mV, the average delay decreases by 3 *orders of magnitude* and σ is larger than $\langle\tau\rangle$. Near 92 mV, the gap evident in Fig. 2(b) appears. Above 92 mV, only short delays are observed, with $\sigma<\langle\tau\rangle$. Note that the voltage at which the gap occurs is different in Figs. 3 and 2(b). We attribute this to an observed extremely long-term (weeks) drift in the threshold voltage.

Thin, short samples of uniform cross section from freshly grown batches of $NbSe_3$ are most likely to have a single switch. All measurements reported here were performed on a single sample 0.4 mm long with resistance 630 Ω at 25 K and 3.81 kΩ at 295 K. A temperature of 30 K was convenient because switching does not occur much above 30 K in virgin samples,[12] and at much lower temperatures heating becomes a problem. Lowering the temperature to 25 K did not qualitatively change the observed behavior. Measurements were also performed on other samples from two growths. All samples we measured showed long and short delays with a gap in $N(\tau)$ in the range 10 μs–1 ms, a power-law tail in $N(\tau)$ for long delay times, and $\langle\tau\rangle$ decreasing faster than ϵ^{-2} for small

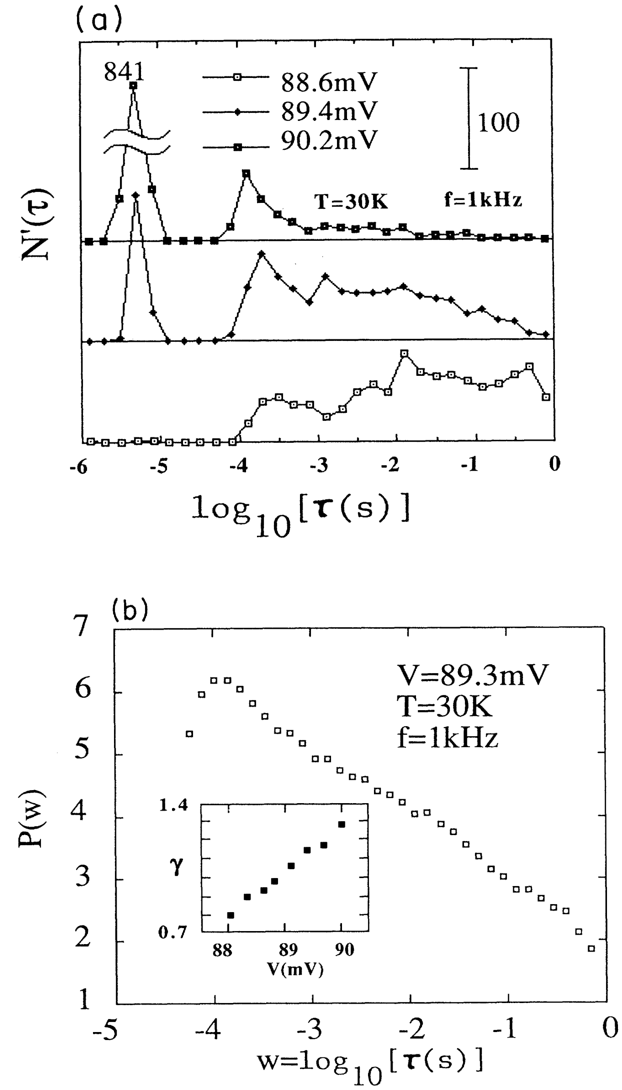

FIG. 2. Distributions of delays. (a) Dependence on V: Three distributions $N'(w)$ (where $w=\log\tau$) of 1024 delays each, binned in logarithmic increments. The distribution shifts to shorter times as V is increased. A gap in $N'(w)$ appears between 10 and 100 μs. (b) Power law: For $\tau>10^{-4}$ s, $N(\tau)\propto\tau^{-\gamma}$. This is evident here because $P(w)=\log_{10}[N'(w)/10^w]$ lies on a straight line over 4 orders of magnitude (see text). Inset graph shows the dependence of the exponent γ on the pulse height V. Standard error on γ was of order 5%.

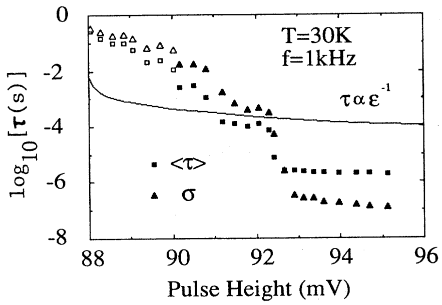

FIG. 3. Average $\langle \tau \rangle$ (squares) and standard deviation σ (triangles). Only distributions with fewer than 10% of delays longer than 1 second are included. The σ and $\langle \tau \rangle$ represented by hollow symbols are systematically low: some of these delays were longer than 1 s but are included in the average as 1-s delays. Each $\langle \tau \rangle$ is the average of 1024 delays. The solid line represents $\langle \tau \rangle(\epsilon) \propto \epsilon^{-1}$ (for $V_c = 87.8$ mV) predicted for the model of Strogatz *et al.* (Ref. 16).

ϵ, where $\epsilon = (V - V_c)/V_c$. The exact position of the gap and the exact form of $\langle \tau \rangle(\epsilon)$ vary from sample to sample. Initial observations by Zettl and Grüner[5] are consistent with the *short* delays we have observed.

Several theories have been proposed to explain CDW conduction in switching samples. Hall *et al.* [12] have proposed that switching samples contain a few "ultrastrong pinning centers"[13] which prevent the intact CDW from sliding. The CDW can slide only when the internal strains become sufficiently large to cause tears, or phase slips, in the fabric of the condensate. Inui *et al.* [14] proposed a many-body Hamiltonian embodying these ideas, and they numerically investigated a 1-degree-of-freedom version. Strogatz *et al.* [15] have proposed a different, exactly soluble many-body Hamiltonian that is isomorphic to the mean-field x-y model. Each of these models shows delayed switching,[16] with $\tau \propto \epsilon^{-\beta}$, where $\epsilon = (V - V_c)/V_c$. For the model of Strogatz *et al.*, $\beta = 1$ (solid line in Fig. 3) and for any 1-degree-of-freedom model, $\beta = \frac{1}{2}$. Using our operational definition of V_c to define ϵ, the average τ decreases faster than ϵ^{-2} for $0.005 < \epsilon < 0.05$, ruling out the model of Strogatz *et al.* and all 1-degree-of-freedom models. There are no fluctuations in τ reported for the model of Strogatz *et al.*

We analyze our results by modifying successful classical models of conventional sliding CDW conduction to include ultrastrong pinning centers. A discretized phase-dynamical model that has been studied numerically by a number of authors is[2]

$$\frac{d\phi_i}{dt} = \frac{\phi_{i+1} - \phi_i}{x_{i+1} - x_i} - \frac{\phi_i - \phi_{i-1}}{x_i - x_{i-1}} + \frac{1}{2} e\,(x_{i+1} - x_{i-1}) + V \sin(\theta_i + \phi_i) \;, \tag{1}$$

where ϕ_i is the phase of the CDW at the ith impurity site, x_i is the random dimensionless position of the ith impurity, e is the dimensionless electric field, V is the strength of the impurity pinning potential, and θ_i is a random phase.

Phase-dynamical models are only valid when the local strain, or phase gradient $\phi' = d\phi/dx$ of the CDW is smaller than a critical value ϕ'_c. A phase gradient larger than ϕ'_c will cause the CDW to tear by nucleating a phase vortex.[12,14] The sparsely distributed, extremely strong impurities that pin CDW in switching samples prevent the CDW from sliding even above the critical voltage for depinning in conventional samples.[12] In our picture, as the voltage across the sample is increased, the local strain somewhere in the sample will eventually exceed ϕ'_c. At this point, a large portion of the CDW begins to slide.

We have assumed that, on application of a voltage pulse, phase-slippage does not occur until $t = \tau$. Thus the details of the dynamics of phase slippage are unimportant in modeling τ. We can qualitatively explain our data with two simple modifications to Eq.(1). (1) The presence of a single extremely strong pinning center is modeled by changing the boundary condition to fix the phase at one end of the chain. (2) Each "spring" is assigned a breaking threshold $(\phi_{i+1} - \phi_i)t = \Delta\phi_i^t = \phi'_c l_i$ where $l_i = x_{i+1} - x_i$ is the distance between impurities. The state of a static CDW in configuration space can be defined by the vector $\mathbf{v} = (\Delta\phi_1, \Delta\phi_2, \ldots, \Delta\phi_N)$, where $\Delta\phi_i = \phi_{i+1} - \phi_i$. The intersection of the planes defined by $\Delta\phi_i = \Delta\phi_i^t$ defines the surface of a "hyperrectangle"[17] in the configuration space of the CDW. We call this surface the phase-slip boundary (PSB). The volume enclosed by the PSB contains all phase-slip-free configurations of the CDW. At any point exterior to the PSB, phase slippage must occur and the CDW must slide. The switching delay τ is then the time it takes for the CDW to evolve from one of an exponentially large number of metastable configurations to the PSB.

This simple picture qualitatively explains many of our observations.

(1) Memory of previous switch: A CDW begins from a relaxed state. A pulse applied to this CDW will cause a switch after a relatively long delay τ_0 and place the CDW in a highly polarized configuration. A second pulse will cause a switch in a shorter time τ_1, because the highly polarized configuration is closer to the PSB.

(2) Displacement current: The current that flows before the switch in Fig. 1 is the displacement current of a polarizing CDW.

(3) τ and the microscopic polarization: The switch in this model will not always occur at the same macroscopic CDW polarization because the condition for switching is that the *local* phase gradient $\phi'_i > \phi'_c$.

(4) Distribution of delays and erasing pulse: Each of the exponentially large number of metastable configurations of the CDW should take a different time to evolve to the PSB. Contributions to the maximum width of the distribution of delays come from distributions in both initial configurations and in $\Delta\phi_i^t$. As the erasing frequency f is increased, smaller and smaller subsets of allowed initial configurations are sampled, as shown by the decreasing width of $N'(w)$.

The erasing frequency at which erasing pulses become ineffective, 5 kHz, is close to the reciprocal of the position of the long-time edge of the gap in Fig. 2(a) $\tau \approx 10^{-4}$ s. Such high-frequency pulses could induce no long switches, although with amplitude $2V_C$ they repeatedly depinned the CDW after short switches. Apparently, many long switches are required to access a wide distribution of initial configurations. However, to understand the central result of this paper, that the switching delay is extremely sensitive to the initial configuration, requires dynamical simulations beyond the scope of this paper. A clarification of dynamic issues will shed light on the distribution and evolution of internal strains in CDW conductors, quantities that have been inaccessible to previous experiments.

Our experimental results may also be relevant to other continuum systems in which large, inhomogeneous internal strains can build up. The dynamics of earthquake faults have been modeled by a chain of identical masses with nonlinear damping coupled by Hooke's law springs.[18] In an earthquake fault, the local response can be elastic only up to a critical strain. For larger strains, slippage occurs. The time it takes for such an event to occur is the time it takes for the earthquake fault to evolve from its initial configuration to the phase-slip boundary.

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