

Spatially resolved study of charge-density-wave strain in NbSe₃: Evidence for a finite threshold for creep

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We have used spatially resolved measurements of the low-field charge-density-wave (CDW) response in NbSe₃ to obtain the CDW strain profile with single-wavelength resolution. Below ~ 35 K, the single-particle (linear) resistance R shows significant hysteresis with electric field E for E below a threshold field E_T . The size of the local hysteresis loop $\Delta R(x)$ and thus of the CDW strain $\epsilon(x)$ varies linearly with position to within $20 \mu\text{m}$ of the current contacts. Our results confirm that slip boundary conditions at the current contacts are responsible for the single-particle resistance hysteresis, but not for the “switching” observed at a larger field E_T^* . Although CDW motion is slow and creeplike between E_T and E_T^* , extremely slow hysteresis relaxation below E_T implies that the creep rate changes by at least several orders of magnitude at E_T . This threshold behavior is inconsistent with existing predictions for thermal creep, and highlights the highly unusual character of CDW dynamics at low temperatures.

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The mechanism of low-temperature transport in charge-density-wave (CDW) conductors remains one of the most important problems in the collective dynamics of disordered systems. Below ~ 35 K in NbSe₃, CDW conduction is characterized by two threshold fields E_T and E_T^* . Between E_T and E_T^* , the CDW velocity is extremely small, temperature-activated, and increases exponentially with applied field, suggesting that motion occurs via thermal creep.¹ However, coherent voltage oscillations accompanying this “creep” imply that CDW motion exhibits temporal order. At E_T^* , the CDW velocity in high quality samples with homogeneous current flow jumps by several orders of magnitude; Above E_T^* , conduction is nearly ohmic and weakly temperature dependent. Significant hysteresis is observed in the single-particle resistance below E_T and in the CDW velocity near E_T^* .² Many of these features have been observed in other CDW conductors.^{3,4}

The single-particle resistance and its low-field hysteresis have been extensively studied,^{3,5–8} in part because the resistance couples to CDW strain and thus probes CDW structure. Although originally believed to be intrinsic to the CDW's bulk response in the presence of quenched disorder, a variety of experiments including four-probe,⁶ five-probe⁷ and sliding-contact three-probe transport measurements⁸ indicate that hysteresis is primarily a finite-size effect associated with boundary conditions imposed by current contacts.⁹ Spatial variations in CDW strain in the high-temperature sliding state have been probed using x-ray diffraction¹⁰ and optical transmission,¹¹ but these techniques have not been extended to characterize the hysteretic low-temperature, low-field strain. Most experiments to date have been corrupted by inhomogeneous CDW depinning and current flow associated with crystal defects and contact imperfections.

Here we report spatially resolved measurements of the single-particle resistance in high-quality NbSe₃ crystals at low temperatures. Our results indicate that the CDW strain profile varies linearly with position to within $20 \mu\text{m}$ of the current contacts, that the low-field hysteresis is due to

boundary conditions at current contacts, and that the “switching” at E_T^* is a bulk effect not associated with these boundary conditions. Hysteresis is confined to the region $-E_T < E < E_T$ and shows extremely slow relaxation. This implies an extremely sharp onset for creeplike motion at E_T , providing additional evidence for the remarkable character of CDW transport at low temperatures.

High-quality NbSe₃ crystals with bulk residual resistance ratios $r_R > 300$, homogeneous current flow,¹² and cross-sectional dimensions of roughly $1 \times 5 \mu\text{m}$ were mounted on arrays of $2 \mu\text{m}$ wide gold-topped chromium wires, as discussed in Ref. 13. The differential resistance $R_d = dV/dI_{tot}$ versus applied current I_{tot} was measured as a function of position with a spatial resolution of $20\text{--}70 \mu\text{m}$. To trace out the limiting hysteresis loop, I_{tot} was swept from a large negative value ($< -I_T^*$) to a large positive value ($> I_T^*$) and then back to its initial value. The typical acquisition time for a current sweep was 30 min, sufficient to allow a close approach to steady state.

The resulting measurements of single-particle resistivity $\rho_s(x) \propto R_d(x)$ for $|I| < I_T$ were used to determine the CDW strain profile $\epsilon(x) = (1/Q)(\partial\theta/\partial x)$, where Q is the CDW wave vector and θ is the CDW phase.^{3,13} CDW strain corresponds to a modulation of the condensed carrier density, and induces a modulation of the single-particle density and resistivity, $\delta\rho_s/\rho_s = \eta\epsilon + \mathcal{O}(\epsilon^2)$. In a semimetal with one CDW band and one metallic band, $\eta \approx n_c/n_s$. In NbSe₃ at low temperatures both hole and electron pockets are present and η cannot easily be determined.¹⁴ Using $n_c = 10^{21} \text{ cm}^{-3}$ and $n_s = 5 \times 10^{18} \text{ cm}^{-3}$ in the two-band expression of Ref. 14 yields an upper bound of $\eta \approx 200$.

Figure 1 shows the differential resistance R_d as a function of current and position between current contacts for a NbSe₃ sample at $T = 25$ K. The abrupt drop in R_d at $I_T^* = 220 \mu\text{A}$ indicates a “switch” to the high-velocity CDW state,³ and is independent of position. At lower currents R_d exhibits hysteresis, where dc I - V measurements reflect changes in the absolute resistance $R = V/I_{tot}$. For constant

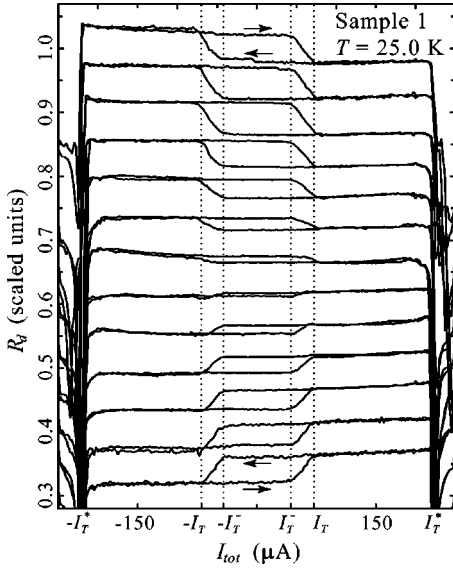


FIG. 1. Differential resistance R_d versus driving current I_{tot} for NbSe₃ sample 1. Each curve is obtained from a different segment of the sample; from top to bottom, the segments are centered at $x = -345, -325, -280, -210, -140, -70, 0, 70, 140, 210, 280, 325,$ and $345 \mu\text{m}$, respectively, with current contacts at $\pm 355 \mu\text{m}$. The curves are vertically offset. The low-field sample resistance per unit length is $\sim 830 \Omega/\text{cm}$.

I_{tot} , the value of R_d in the hysteresis loop is stable over a time scale of at least several hours for $T < 30 \text{ K}$. For reasons to be discussed below, we associate the value of I_{tot} that bounds this hysteretic region with the threshold current I_T .

Figure 1 indicates that R_d varies systematically with position x , exhibiting the approximate symmetry $R_d(x, I) = R_d(-x, -I)$. We define the size of the hysteresis loop as $\Delta R_d = R_d^+(0) - R_d^-(0)$, where R_d^+ and R_d^- correspond to the values of R_d at $I_{tot} = 0$ obtained when I_{tot} is increasing and decreasing, respectively, through $I_{tot} = 0$. As shown in Fig. 2, $\Delta R_d/R_d$ varies linearly with position x between the current contacts and passes through zero near the sample midpoint $x = 0$. Behavior similar to that in Fig. 1 has been observed in more than eight samples.

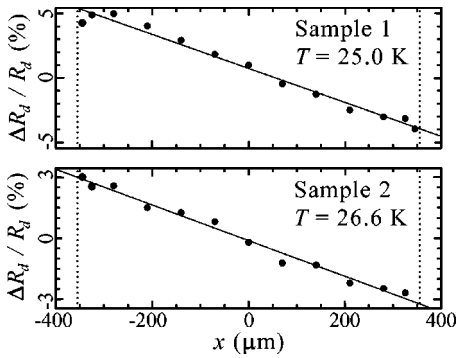


FIG. 2. Hysteresis loop height $\Delta R_d = [R_d^+(0) - R_d^-(0)]/\bar{R}_d$ versus position x for samples 1 and 2. The straight line is the form expected for strain-assisted phase slip near the current injection contacts. The vertical dotted lines represent the position of the current contacts.

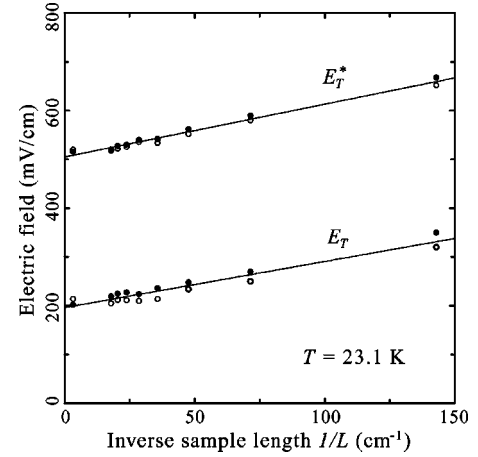


FIG. 3. E_T and E_T^* versus distance L between the current contacts for sample 3. E_T and E_T^* were determined from differential resistance measurements as shown in Fig. 1. The solid lines represent fits to $E = E_p^0 + V_{ps}/L$, with $V_{ps} = 0.94$ and 1.08 mV for E_T and E_T^* , respectively. Open and solid symbols represent negative and positive drive current polarities, respectively.

The force applied to the CDW between the current contacts also produces CDW strain beyond them, and this strain can be measured using arrays of contacts.¹³ Below I_T^* at $T \approx 25 \text{ K}$, the CDW current $I_c < 10^{-3} I_{tot}$ and so changes in total resistance are almost entirely due to changes in single-particle resistance. Measurements using a six-probe configuration indicate that for $x > 20 \mu\text{m}$ beyond the contacts, $R_d(x)$ and thus the strain remain independent of I_{tot} as I_{tot} is swept through the switching current I_T^* . Consequently, the CDW strain profile beyond and between the contacts does not change appreciably at I_T^* .¹⁵

$R_d(I_{tot})$ was measured for several values of current contact separation L and currents I_T, I_T^* and the corresponding fields E_T, E_T^* extracted as in Fig. 1. Figure 3 shows that both $E_T(L)$ and $E_T^*(L)$ vary linearly with $1/L$ and have a finite intercept. The intercepts determine the bulk (large L) values of E_T and E_T^* , and the slopes determine the ‘‘phase-slip voltage’’ V_{ps} near these fields.⁹ Fits to the two lines in Fig. 3 yield slopes that agree to within 15%. Since for $I_c \approx 0$, V_{ps} is proportional to the CDW strain near the current contacts,⁹ the strain magnitudes near E_T and E_T^* are essentially the same. This provides additional evidence that the transition at E_T^* is not associated with phase slip at the current contacts.

The hysteresis observed in R_d for $|I_{tot}| < I_T$ is readily understood to be a consequence of boundary conditions at the current contacts.⁹ We use the model of Adelman *et al.*,¹³ which provides a semiquantitative description of the spatiotemporal response of the CDW current, strain, and phase-slip rate to changes in applied current at higher temperatures in NbSe₃. As will be discussed elsewhere, this model is equivalent to a model recently proposed by Brazovskii *et al.*¹⁶

At current contacts CDW current must be converted to/from single-particle current, and this requires strain-induced phase slip via formation of CDW dislocations.¹⁷ Conservation of total current implies that the CDW current density is given by

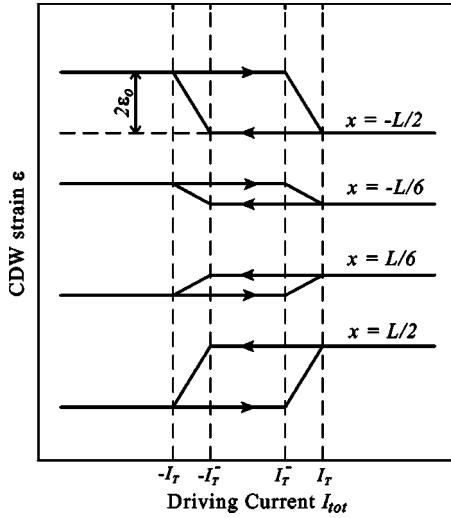


FIG. 4. Strain profile versus driving current and position, as predicted by Eqs. (2) and (3). The current contacts are at $x = \pm L/2$.

$$j_c(x, t) = \frac{1}{\rho_s + \rho_c} \left[\rho_s j_{tot} - E_p(j_c) + \kappa \frac{\partial \epsilon(x, t)}{\partial x} \right], \quad (1)$$

where ρ_s is the single-particle resistivity, ρ_c is the CDW resistivity,¹⁸ $E_p(j_c)$ describes the effective force due to pinning, and $\kappa \equiv (Q^2/en_c)K$ where K is the CDW elastic constant. The pinning force E_p always opposes the effective force $E_{eff} = \rho_s j_{tot} + \kappa(\partial \epsilon / \partial x)$. When the CDW is pinned, $E_p(0)$ acts like a static friction force, self-adjusting up to a maximum value $E_p^0 = |E_p(0)|_{max}$ (corresponding to the large- L limit of E_T) so as to keep the net force $E_{eff} - E_p$ driving CDW motion equal to zero. When the CDW depins and $|E_{eff}| < E_T^*$, $j_c \ll j_{tot}$. $E_p(j_c)$ is then essentially constant and can be written as $E_p(j_c) = \text{sgn}(j_c)E_p^0$.

In our experiments, j_{tot} is swept from a large negative value ($< j_T^*$) to a large positive value ($> j_T^*$). Assuming that the phase-slip rate increases extremely steeply from zero at a critical strain ϵ_c ,¹³ the solution to Eq. (1) for $\epsilon(x)$ between the current contacts is

$$\epsilon(x, j_{tot}) = [2x/L] f^+(j_{tot}) \epsilon_c, \quad (2)$$

where

$$f^+(j_{tot}) = \begin{cases} -1, & j_{tot} < j_T^- \\ (\rho_s L / 2\kappa \epsilon_c)(j_{tot} - j_P), & j_T^- < j_{tot} < j_T \\ 1, & j_T < j_{tot}. \end{cases} \quad (3)$$

Here $j_P = E_p^0 / \rho_s$, $j_T = j_P + 2\kappa \epsilon_c / \rho_s L$ and $j_T^- = j_P - 2\kappa \epsilon_c / \rho_s L$. The solution for a sweep from $j_{tot} > j_T^*$ to $j_{tot} < -j_T^*$ is obtained by replacing $f^+(j_{tot})$ with $f^-(j_{tot}) = -f^+(-j_{tot})$. This result, plotted in Fig. 4, is consistent with the position and current dependence of the observed single-particle resistance ($\propto \epsilon$) in Fig. 1. Typical fit parameters obtained at $T = 25$ K are $\eta \epsilon_c = 0.04$, $E_p^0 = 50$ mV/cm, and $2\kappa \epsilon_c / E_p = 180$ μm . As in previous work, E_T is predicted to vary with distance L between the

current contacts as $E_T = E_p^0 + 2\kappa \epsilon_c / L \equiv E_p^0 + V_{ps} / L$, consistent with Fig. 3. A fit to the data of Fig. 3 yields $V_{ps} = 0.94$ mV. V_{ps} is also given by $L(E_T - E_T^-) / 2$. Using experimental values for E_T and E_T^- for the sample of Fig. 3 yields $V_{ps} = 1.1$ mV, in agreement with the value obtained from E_T versus L .

The following picture emerges from this analysis. For $|j_{tot}| < j_T^-$, the CDW is pinned by impurities, $\epsilon(x)$ is independent of j_{tot} and (in the present experiment) drives the CDW in the direction opposite to that in which it was last depinned, and E_p adjusts to cancel changes in j_{tot} and keep $j_c = 0$. For $j_T^- < |j_{tot}| < j_T$, E_p remains constant at its maximum value E_p^0 and the strain profile adjusts to cancel changes in j_{tot} , reaching a slope equal to that for $j_{tot} < j_T^-$ but with opposite sign at $j_{tot} = j_T$. Although there is no steady-state CDW motion between j_T^- and j_T , $\epsilon(x)$ rearranges and a polarization transient flows each time j_{tot} is changed. For $|j_{tot}| > j_T$, phase slip occurs near the current contacts producing steady-state CDW motion, and (with our approximation for the slip-strain relation) the strain profile remains independent of driving current.

The magnitude of the differential resistance hysteresis ΔR_d near a current contact is $\Delta R_d / R_d \approx 4\%$ at 25 K. Using $\eta = 200$, this corresponds to a strain $\epsilon \approx 10^{-4}$. This strain is an order of magnitude smaller than typical strains observed at 90 K by x-ray diffraction for the T_{P_1} CDW,¹⁰ and is consistent with an upper bound on the strain obtained from our x-ray measurements at $T = 25$ K. The CDW's midsample displacement associated with this strain profile is ~ 25 wavelengths in our 0.7 mm length samples.

The present results have several consequences. First, they indicate that single-particle resistance hysteresis primarily arises from contact effects, and that any intrinsic hysteresis is extremely small, consistent with earlier results. Second, the absence of a significant change in CDW strain both between and beyond the current contacts at the switching threshold E_T^* shows that switching is not associated with phase slip at contacts. The linearity of the strain profile implies that switching is also not associated with phase slip centers between the contacts.²

Third, the saturation of R_d to a nearly constant and history-independent value for $I_T < I_{tot} < I_T^*$ shows that the CDW undergoes slow steady-state motion between I_T and I_T^* . If instead there were no motion or the motion were impeded by boundary conditions at the current contacts, the single-particle resistance and CDW strain would increase with increasing I_{tot} for $|I_{tot}| > I_T$. Evidence for CDW motion above I_T has been provided by observations of broadband noise¹⁹ and, more compellingly, of highly coherent voltage oscillations and of the transient response.¹ As I_{tot} is increased from I_T^- to I_T , the single-particle resistance moves toward its $I_{tot} > I_T$ value. The CDW strain profile is changed by a transient CDW polarization current, but the persistence of hysteresis on long-time scales shows that the steady-state current is approximately zero. The net effect of phase-slip boundary conditions is to shift the threshold field for the onset of steady-state CDW sliding from the bulk threshold E_p^0 to $E_p^0 + 2\kappa \epsilon_c / L$.

Finally, we emphasize what we believe to be our most important result: the abrupt onset of temperature-activated, creeplike CDW motion at I_T . In every other collective transport system of which we are aware, thermal creep eliminates the sharp zero-temperature threshold, and the collective mode's velocity increases smoothly as the applied force is increased from zero. From the data of Ref. 1 at $T = 25.9$ K, the CDW velocity increases roughly exponentially from ~ 10 wavelengths per second (~ 100 Å/s) at $1.1 I_T$ to $\sim 10^3$ wavelengths per second at $2.8 I_T$. In the present measurements, single-particle resistance changes due to CDW displacements of roughly one wavelength can be resolved. Since the hysteretic resistance below I_T shows no relaxation on a time scale of hours, the CDW velocity at, for example, $0.9 I_T$ must be less than 10^{-4} wavelengths per

second. Consequently, the CDW velocity increase between $0.9 I_T$ and $1.1 I_T$ is at least 10^5 times larger than its increase from 1.1 to $1.3 I_T$; the characteristic current scale describing this increase differs by a similar factor. This threshold behavior of the creep velocity is inconsistent with all existing theories of elastic and plastic creep in CDW's.^{4,20} Based on this fact and our earlier measurements of coherent oscillations, CDW transport in the low-temperature regime appears to result from an interplay between collective dynamics and thermal fluctuations that is without precedent within the broader context of collective transport in disordered media.

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