Charge-Density-Wave Phase Slip and Current Conversion in $NbSe_3$

M. P. Maher, T. L. Adelman, S. Ramakrishna, J. P. 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

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We report measurements of the phase-slip process by which charge-density-wave (CDW) current is converted to single-particle current at electrical contacts. An excess voltage V_{ps} produces a large static deformation of the CDW phase, which drives formation of dislocation loops. Measurements of the phase-slip rate as a function of V_{ps} for both CDWs in $NbSe_3$ reveal a diodelike relation, analogous to that for phase slip in superfluids. Our results are generally consistent with the predictions of Ramakrishna *et al.*, and may have significant implications for study of CDW dynamics at temperatures well below the Peierls transition.

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The dynamics of charge density waves (CDWs) remains one of the most interesting and rich subjects in condensed-matter physics [1]. The electronic density in the CDW state has the form $\rho(x)=\rho_0+\rho_1\cos[Qx+\phi(x,t)]$, where $Q=2k_F$ is the CDW wave vector. In the presence of impurities, the CDW deforms and becomes pinned to the lattice, leading to spatial variations of its phase ϕ. When an electric field greater than a threshold field E_T is applied, the CDW overcomes the impurity pinning force and slides through the crystal, resulting in collective charge transport and a current $I_c \propto \partial\phi/\partial t$.

Spatial discontinuities in the CDW current, such as occur at electrical contacts, require a means of converting single-particle current into a moving CDW. It has long been accepted that this current conversion occurs via phase slip [2-8], in which dislocations in the CDW superlattice are formed at the boundary between moving and stationary parts of the CDW. Previous experimental work [5-8] has shown that an additional sample-length-independent voltage, known as the phase-slip voltage V_{ps}, is necessary to obtain CDW current, and has provided evidence that CDW phase slip is thermally activated. The data from these experiments have been interpreted using a phenomenological extension of the theory of phase slip in 1D superconducting wires [9].

Here we describe detailed measurements of CDW phase slip in $NbSe_3$. We find that a finite, strongly temperature-dependent threshold voltage V_{ps0} is required to initiate CDW phase slip. Reasonable extensions to the 1D phase-slip model cannot account for this threshold. We show that our results are in general agreement with a theory for 3D phase slip proposed by Ramakrishna *et al.* [10]. Some preliminary results of this investigation have been described elsewhere [11].

As illustrated in Fig. 1, if a voltage V is applied across a portion of a CDW crystal of length L, two regimes of behavior can be crudely distinguished: (I) When the voltage is less than the bulk threshold voltage $V_T=E_TL$ due to impurity pinning, the CDW polarizes from its $V=0$ state but remains pinned. (II) Increasing the voltage beyond the bulk threshold V_T frees the CDW to slide past the impurities. However, the CDW beyond the contacts (where the electric field $E=0$) remains pinned, so that motion between the contacts produces a strain profile given approximately by [4,10]

$$\Sigma=K_z\frac{\partial\phi}{\partial z}=\frac{1}{2}\frac{e\rho_s}{Q}V_{ps}\left[\frac{2z}{L}-1\right], \qquad (1)$$

where z is the distance from one of the contacts, $e\rho_s$ is the CDW charge density, K_z is the CDW elastic constant, and $V_{ps}\sim V-E_TL$ is called the phase-slip voltage. The maximum magnitude of the strain occurs at the contacts (i.e., at $x=0$ and $x=L$) and depends only upon V_{ps} and not upon the contact separation. This strain drives CDW phase slip near the contacts [2-4], allowing time-averaged motion of the CDW between the contacts. The phase-slip rate and thus the magnitude of I_c will increase with increasing V_{ps} and Σ.

A method for determining the I_c-V_{ps} relation from four-probe I-V measurements was introduced by Gill [5] and by Monceau *et al.* [6]. In the usual four-probe configuration (hereafter referred to as the normal configuration), current is injected through the outer pair of contacts, and the induced voltage is measured across the inner pair. In the transposed configuration, current is injected through the inner pair of contacts, so that the voltage measured across the outer pair includes the entire

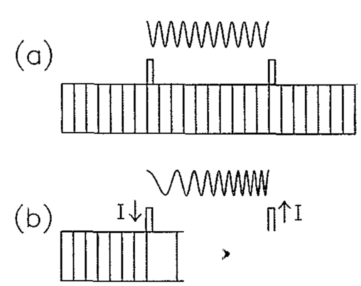

FIG. 1. A CDW sample with two electrical contacts. (a) The applied voltage between the contacts $V=0$, the CDW current $I=0$, and the CDW is unstrained. (b) $V>V_T$ and $I>0$. The CDW is compressed near one contact and stretched at the other. These strains drive formation of vortex loops near the contacts, which allow time-averaged CDW motion.

phase-slip voltage. At a fixed CDW current I_c, the voltages $V_{\text{norm}}(I_c)$ and $V_{\text{trans}}(I_c)$ measured in the normal and transposed configurations, respectively, are given by

$$V_{\text{norm}}(I_c) = E_b(I_c)L_{\text{in}} + V_{\text{ps}}(I_c)(L_{\text{in}}/L_{\text{out}}), \tag{2a}$$

$$V_{\text{trans}}(I_c) = E_b(I_c)L_{\text{in}} + V_{\text{ps}}(I_c), \tag{2b}$$

where $E_b(I_c)$ is the bulk electric field required to produce a CDW current I_c, and L_{in} and L_{out} are the separations of the inner and outer pair of contacts, respectively. The phase-slip voltage can thus be calculated using

$$V_{\text{ps}}(I_c) = [V_{\text{trans}}(I_c) - V_{\text{norm}}(I_c)]\left[\frac{L_{\text{out}}}{L_{\text{out}} - L_{\text{in}}}\right]. \tag{3}$$

We have measured the I_c-V_{ps} relation as a function of temperature for both the $T_{P_1} = 145$ K and $T_{P_2} = 59$ K CDWs in $NbSe_3$. Several indium contacts were thermally evaporated through a shadow mask onto high-purity [residual resistivity ratio (RRR) ≈ 280] single crystals. Contact separations varied from 5 to 2500 μm, contact widths were typically 5–10 μm, and contact resistances were 5–10 Ω [12]. Voltage and differential resistance were measured versus current for each individual contact pair in both the normal and transposed configurations, and $V_{\text{ps}}(I_c)$ was calculated using Eq. (3). The separation of the outer contact pair was always at least 4 times the separation of the inner pair, and the inner contacts were always adjacent.

Figure 2 shows I_c-V_{ps} curves for the T_{P_1} CDW at various temperatures, obtained using a sample 8.3 μm wide and 1.2 μm thick. The uniformly high quality of our electrical contacts yielded significantly higher-quality data than previously obtained. At all temperatures, the I_c-V_{ps} relation has a diodelike form: I_c is essentially zero up to a threshold phase slip voltage $V_{\text{ps}0}$, and then increases rapidly for $V_{\text{ps}} > V_{\text{ps}0}$ [13]. The threshold $V_{\text{ps}0}$ increases rapidly with decreasing temperature. Similar

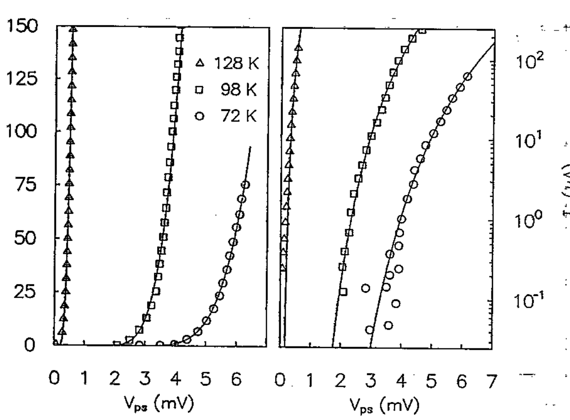

FIG. 2. CDW current I_c and $\log(I_c)$ vs phase-slip voltage V_{ps} for the T_{P_1} CDW in $NbSe_3$ at three temperatures. The solid lines are fits to the data using Eq. (5).

results are obtained for the T_{P_2} CDW, although V_{ps} values for a given I_c are roughly an order of magnitude smaller.

In previous studies [5,7], I_c-V_{ps} data were fitted using

$$I_c = I_0 \exp(-F/T) \sinh[\alpha V_{\text{ps}}/T], \tag{4}$$

which describes 1D phase slip in thin superconducting wires [9]. This equation fails to describe CDW phase slip in two ways. First, I_c in Eq. (4) increases smoothly with V_{ps} and does not exhibit a threshold $V_{\text{ps}0}$. Reasonable fits can be obtained if V_{ps} is replaced with $V_{\text{ps}} - V_{\text{ps}0}$ [5,11]. However, attempts to develop a physical justification for this replacement have been unsuccessful. Second, if the phase slip were one dimensional, in each phase-slip event the CDW amplitude would collapse in the entire cross section A of the sample, and the total charge removed would be $\alpha = e\rho_s\lambda A$. As noted previously [5,7], experimental values of α deduced from fits by Eq. (4) are several orders of magnitude too small; the experimental temperature dependence of α is also inconsistent with theory.

This failure of the 1D phase-slip theory is not surprising. Since the CDWs amplitude coherence length ξ (~10–50 Å in $NbSe_3$) is orders of magnitude smaller than typical sample dimensions, CDW phase slip should be three dimensional. As discussed by Lee and Rice [2] and by Feinberg and Friedel [4], phase slip should thus occur via formation of phase dislocation loops.

Ramakrishna *et al.* [10] have used an analogy with superfluids to develop a quantitative theory of 3D CDW phase slip. In the superfluid case, the strain $\nabla\phi$ which drives vortex loop formation is proportional to the superfluid velocity v_f; for a spatially uniform v_f the rate of vortex formation will also be uniform. In the CDW case, the strain $\nabla\phi$ is a consequence of boundary conditions and is largest near the current contacts, so dislocation loop formation occurs primarily in their vicinity. Assuming a CDW strain profile as given in Eq. (1), Ramakrishna *et al.* [10] found that a phase-slip voltage V_{ps} generates CDW current I_c at a rate given approximately by

$$I_c = I_0 (V_{\text{ps}}/V_a) \exp(-V_a/V_{\text{ps}}), \tag{5}$$

where

$$I_0 \approx \omega \frac{e\rho_s}{Q} \frac{A^2}{4\xi^3} L, \tag{6a}$$

$$V_a \approx \frac{\pi^2 Q K_x^2}{e\rho_s k_B T} \approx \frac{(3200\text{ mV K})}{T}\left[\frac{\Delta}{\Delta_0}\right]^3, \tag{6b}$$

ω is the attempt frequency, ξ^3 is the amplitude coherence volume, Δ is the CDW gap, and K_x is the transverse elasticity. Equation (5) describes thermally activated dislocation loop nucleation, where the barrier height is inversely proportional to the strain and thus to V_{ps}. This strain dependence of the barrier yields a sharp onset of phase slip at a finite V_{ps}. At smaller V_{ps}, phase slip still occurs, but at a very small rate.

The I_c-V_{ps} data were fitted by Eq. (5) using a nonlinear least-squares fitting routine with two adjustable parameters: I_0 and the activation voltage V_a. The quality of these fits (as indicated by the solid lines in Fig. 2) is generally excellent, suggesting that Eq. (5) has the current functional form.

More detailed tests of the theory of Ramakrishna *et al.* are complicated because of an unusual scatter in the experimental data. As long as the CDW remains pinned, the normal and transposed *I-V* curves agree exactly and are completely reproducible. When the CDW is depinned, repeated measurements in the normal configuration using a given set of contacts yield essentially identical *I-V* characteristics. However, using the same contacts, repeated measurements in the transposed configuration yield slightly different *I-V* characteristics. These differences translate into apparently random variations in V_{ps} for a given I_c of $\sim$10%–20%. The magnitude of these variations is roughly independent of the inner contact pair separation, and dominates the scatter in $V_{ps}(I_c)$ values obtained for different contact pairs [14].

In order to account for the scatter in V_{ps}, Eq. (5) was fitted to a large number of $I_c(V_{ps})$ curves obtained from repeated measurements on several different-length sections of the same crystal. $V_a(T)$ was then calculated as the average value of V_a obtained from these fits. I_0 was determined by fitting the $I_c(V_{ps})$ data a second time with V_a fixed at this average value.

Figure 3 shows the activation voltage V_a versus temperature for the T_{P_1} CDW in six undoped $NbSe_3$ crystals of comparable thickness. The solid line is a fit of the form $V_a(T) = [(4000 \text{ mV K})/T](\Delta/\Delta_0)^3$, assuming the usual BCS form for the gap Δ. The overall qualitative and quantitative agreement with the theoretical prediction of Eq. (6b) is very good, although the measured temperature variation of V_a is somewhat more rapid than predicted. A similar fit for the T_{P_2} CDW yields $V_a(T) = [(100 \text{ mV K})/T](\Delta/\Delta_0)^3$. The quantitative discrepancy may reflect differences in the elasticity of the two CDWs not accounted for in Ref. [10].

Figure 4 shows I_0 versus contact separation L for the crystal of Fig. 2. The large scatter in I_0 is due to the scatter in V_{ps}: For typical values of V_a, V_{ps}, and I_c, a 20% variation in V_{ps} will give an order of magnitude variation in I_0. In spite of this scatter, I_0 appears to be approximately independent of length L, contrary to the prediction of Eq. (6a).

A length dependence of I_0 is predicted because, for the linear strain profile of Eq. (1) assumed by Ramakrishna *et al.*, the volume in which the strain Σ exceeds the value at which appreciable dislocation nucleation occurs, and thus the phase-slip rate, scales with L. In our experiment, the electrical contacts were applied to one side of the crystal. Calculations of the electric field and strain profiles for this configuration indicate that the strain is enhanced in the immediate vicinity of the contacts by roughly a factor of 2, and that both the size of the region of enhanced strain and the magnitude of the enhancement are approximately independent of current contact separation. Since most of the phase slip occurs in this region, a much weaker length dependence of I_0 is thus expected. Calculations of the phase-slip rate for this strain profile can be described using Eq. (5), with a V_a value roughly a factor of 2 smaller.

An important assumption of our analysis is that the strains which produce phase slip are large compared with those associated with CDW pinning by impurities. This assumption is supported by experiment. First, from the

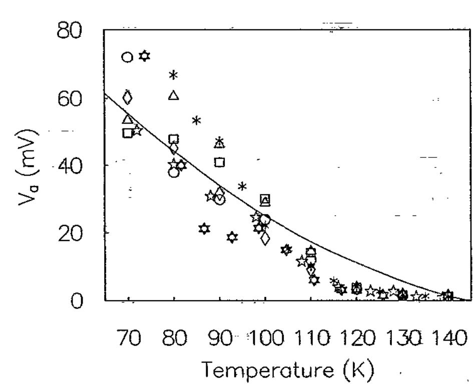

FIG. 3. Temperature dependence of the activation voltage V_a, obtained from fits by Eq. (5) to experimental data for six undoped crystals with RRR $\approx$ 280 (open symbols) and for a Ta-doped crystal with RRR = 35 (asterisks). The solid line is given by $V_a = [(4000 \text{ mV K})/T](\Delta/\Delta_0)^3$.

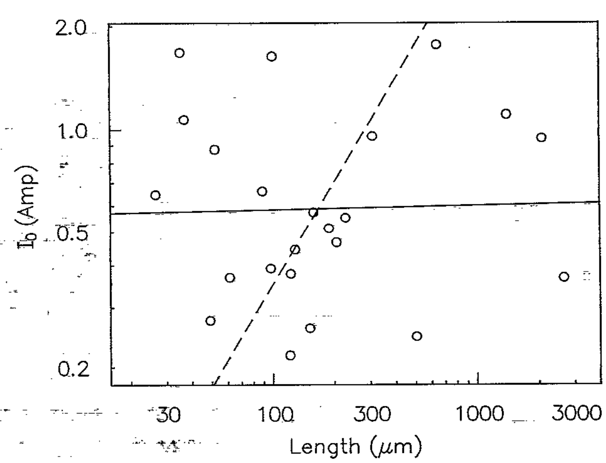

FIG. 4. Dependence of the prefactor I_0 obtained from fits by Eq. (5) on contact separation L. The dashed line is the best linear fit of the form $I_0 = aL$. The solid line is the best power-law fit $I_0 = bL^\gamma$. Although the scatter in I_0 values is large, I_0 appears to be approximately independent of L.

measured V_{ps} values and Eq. (1), we estimate that the phase-slip strains are on the order of 1% below 80 K, more than 2 orders of magnitude larger than typical pinning strains in undoped $NbSe_3$, and that they become comparable to pinning strains only at temperatures within a few degrees of T_P. Second, measurements of the I_c-V_{ps} relation in a Ta-doped crystal with RRR = 35, in which the pinning strains are significantly larger than in undoped crystals, yield $V_a(T)$ essentially identical to that for undoped crystals, as shown in Fig. 3. Third, in high-quality $NbSe_3$ crystals, the CDW velocity is coherent to within one part in 10000. If the phase-slip strains were comparable to pinning strains, then phase slip would occur throughout the crystal and the CDW response would be highly incoherent [15].

Large strains and phase-slip voltages may have significant implications for measurements of CDW dynamics. Substantial phase slip occurs in these measurements, since they are generally performed using a two-probe configuration, or using a four-probe configuration with strongly perturbing voltage contacts. At temperatures below 90 K, the spectral width of the coherent voltage oscillations (narrow-band noise) observed in response to dc currents broadens dramatically, and the Shapiro steps observed in response to combined ac and dc voltages smear out and the mode locking becomes incomplete. Since V_{ps} is large and increasing rapidly in this temperature range, these effects may result from the large phase-slip strains and from fluctuations associated with thermal dislocation nucleation.

Phase slip has been invoked in explanations of a wide variety of CDW phenomena, including CDW-to-normal carrier conversion, narrow-band noise generation [4], broad-band noise generation [15], and "switching" [16]. The present results help to establish the basic character of this process, and provide evidence for the thermal dislocation nucleation model of Ramakrishna *et al.*

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[13] Gill (Ref. [5]) noted the presence of a small but only weakly temperature-dependent threshold phase-slip voltage in his data. A larger scatter and a more limited data set obscured the deficiencies of Eq. (4).

[14] Since in the transposed configuration the depinned length is typically only a few CDW correlation lengths, and since large strains and phase slip occur at the inner contacts, depinning and repinning of the CDW may leave it in different metastable configurations with measurably different threshold voltages. Threshold voltage variations have also been observed in small TaS_3 samples; see S. V. Zaitsev-Zotov, V. Ya. Pokrovskii, and J. C. Gill, J. Phys. (France) I **2**, 111 (1992).

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