Charge-density-wave phase slip in NbSe₃

M.P. MAHER, S. RAMAKRISHNA, D.A. DI CARLO, T.L. ADELMAN, V. AMBEGAOKAR, J.D. BROCK* and R.E. THORNE

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, NY 14853, U.S.A.

** School of Applied and Engineering Physics, Cornell University, Ithaca, NY 14853, U.S.A.*

Abstract : We have studied the phase-slip process by which charge-density-wave (CDW) current is converted to single-particle current at electrical contacts. Transport and X-ray scattering measurements indicate that an excess voltage V_{ps} dropped between current contacts induces a large static deformation of the CDW phase. The measured V_{ps} - and temperature-dependent phase-slip rates are consistent with a model in which CDW dislocation loops are thermally nucleated in the presence of these deformations. The effects of impurities and contact perturbations on the phase slip process are also discussed.

1. Introduction

Discontinuities in charge-density-wave (CDW) current density, such as occur at current contacts, require a means of converting between collective CDW current and single-particle current. This conversion occurs via phase slip, in which CDW phase fronts are locally added or removed, allowing spatial variations of the CDW velocity. Here we give an overview of our recent work on phase slip in the CDW conductor NbSe₃. Experimental and theoretical details are given in Refs. [1-3].

2. Previous Experiments

As first shown by Gill [4] and subsequently by Monceau et al. [5] and Borodin et al. [6], phase slip can be investigated experimentally using four-probe I-V measurements. In the usual four-probe configuration (hereafter referred to as the Normal configuration), current is injected through the outer pair of contacts and the voltage V_{norm} is measured across the inner pair. In the Transposed configuration, current is injected through the inner pair and the voltage V_{trans} is measured across the outer pair. As long as the CDW remains pinned, the voltages V_{norm} and V_{trans} measured for equal currents are the same, as guaranteed by reciprocity. However, once the CDW depins and a finite CDW current I_C flows, $V_{trans}(I_C) > V_{norm}(I_C)$, where the difference $\Delta V(I_C) = V_{trans}(I_C) - V_{norm}(I_C)$ is essentially independent of the inner contact pair separation. Because phase slip occurs between the voltage contacts in the Transposed configuration and outside them in the Normal configuration, Gill suggested that the excess voltage ΔV is required to maintain phase slip at the current contacts.

Experiments by Gill and by Monceau et al. on NbSe₃ showed that I_C , proportional to the phase-slip rate, increased rapidly with ΔV , and that $\Delta V(I_C)$ increased rapidly with decreasing temperature, suggesting that phase slip was thermally activated. Gill showed that an I_C - ΔV relation adapted from the theory of 1-D phase slip in thin superconducting wires provided a good qualitative fit to his data, but that parameter val-

ues extracted from such fits were unphysical. We repeated Gill's experiment using higher purity crystals and higher quality contacts, and found that Gill's fit failed qualitatively at small CDW currents: our data exhibited a strongly temperature-dependent threshold phase slip voltage below which the phase-slip rate I_c was negligible.

3. Phase Slip Theory

Motivated by these experiments, Ramakrishna et al.[1] have developed a theory of CDW phase slip, based upon earlier ideas of Feinberg and Friedel[7] and of Lee and Rice[8] and upon an analogy with phase slip in superfluids. This theory addresses two basic questions: (1) What drives phase slip? and (2) How are CDW phase fronts added and removed? As in superconductors and superfluids, CDW phase slip is driven by gradients $\nabla\phi$ in the phase of the CDW order parameter; i.e., by strain in the CDW superlattice. Unlike these other systems, however, the phase gradients are a consequence of boundary conditions. As shown in Figure 1, for applied electric fields E less than the depinning threshold E_T , the CDW deforms locally but does not depin. For $E > E_T$ the CDW depins from impurities, but beyond the contacts where $E \approx 0$ the CDW remains pinned. Consequently, CDW motion between the contacts results in compression of the CDW near one contact and expansion near the other. The magnitude of the compressions and expansions is determined by the applied voltage in excess of that required to overcome pinning (and, when the CDW slides continuously, damping) forces, defined as the phase-slip voltage V_{ps} . Assuming the electric field between current contacts is uniform and longitudinal, the CDW wavevector varies linearly with position z between the contacts according to:

$$\Delta Q(z) = \frac{\partial\phi}{\partial z} = \frac{e\rho_c}{QK_z} V_{ps} \frac{z}{L}, \quad (1)$$

where K is the CDW elastic constant, $e\rho_c$ is CDW charge density, and L is the current contact separation.

Fig. 1. — A CDW crystal with two electrical contacts for (a) $E < E_T$ and (b) $E > E_T$.

Because the transverse dimensions of typical CDW crystals are orders of magnitude larger than the CDW's amplitude coherence length, CDW phase slip must be 3D instead of 1D, so phase front addition and removal occurs by nucleation of dislocation loops in the CDW superlattice. Nucleation of a loop of radius R reduces the strain energy in the CDW by an amount proportional to R^2 ($\propto$ loop area), but requires an energy roughly proportional to R to drive the CDW amplitude to zero at the circumference of the loop. Consequently, the loop energy has a maximum as a function of R , so that loops below a critical radius will shrink and vanish, while those above this radius will grow and remove an entire phase front. Assuming loop nucleation is thermally activated and a CDW strain profile as given in Eq. (1), the CDW current I_c varies with the phase-slip voltage V_{ps} as

$$I_c = I_0 (V_{ps} / V_a) \exp(-V_a / V_{ps}), \quad (2)$$

where the activation voltage V_a for the $T_p=145$ K CDW in NbSe_3 is given by

$$V_a \approx \frac{(1300 \text{ mV} \cdot \text{K})}{T} \left[\frac{\Delta}{\Delta_0} \right]^3, \quad (3)$$

and $I_0 \propto L$. Because the energy of a critical dislocation loop varies inversely with CDW strain, the strain and thus the V_{ps} required to obtain a given nucleation/phase-slip rate decreases with increasing temperature. The phase-slip rate is largest near the contacts, where the magnitude of the strain is largest, and rapidly falls to zero away from the contacts. The L -dependence of I_0 results because for fixed V_{ps} , the fraction of the sample length where the strain exceeds that needed for appreciable phase slip is independent of L , so the corresponding volume and thus the overall nucleation rate increases with L .

4. Comparison between Theory and Experiment

Figure 2 shows our data for the CDW current I_C as a function of the phase slip voltage V_{ps} for the $T_p=145$ K CDW in NbSe_3 . Since V_{ps} is assumed to be dropped uniformly between current contacts and thus makes a contribution to V_{norm} , V_{ps} is calculated from the Transposed-Normal voltage difference ΔV using $V_{ps} = \Delta V [L_{\text{out}} / (L_{\text{out}} - L_{\text{in}})]$, where L_{out} and L_{in} are the separations of the outer and inner contact pairs. The prediction of Eq. (2) (solid lines) provides an excellent qualitative fit to the data. Figure 3 shows the activation voltage V_a obtained from such fits as a function of temperature for seven NbSe_3 crystals. The solid line is given by $V_a = [(4000 \text{ mV K}) / T] (\Delta / \Delta_0)^3$, assuming the usual BCS form for the gap Δ . The overall qualitative and quantitative agreement with the prediction of Eq. (3) is very good, although the measured temperature variation of V_a is somewhat more rapid than predicted. I_0 values obtained from these fits show no indication of the predicted length dependence, although the scatter in these values is enormous: a 20% uncertainty in V_a translates into an order of magnitude uncertainty in I_0 .

Fig. 2. — CDW current I_C vs. phase-slip voltage V_{ps} at three temperatures.

Fig. 3. — Phase slip activation voltage V_a versus temperature for six undoped NbSe_3 crystals (open symbols) and one Ta-doped crystal (*).

5. Additional Experiments

To investigate the effects of impurities on phase slip, we performed measurements on both undoped ($\text{RRR}=280$) and Ta-doped ($\text{RRR}=35$) crystals. One might expect impurities to pin the dislocation loops and hinder their growth. However, as indicated in Figure 3, the V_a values are the same for both undoped (open symbols) and doped (*) crystals, even though their threshold fields differ by a factor of 6. This result

is consistent with estimates of Ramakrishna which suggest that loop pinning forces are very weak compared with the strain-related Peach-Koehler force which drives growth of the loops.

We have also investigated the effects of contact geometry and perturbations on phase slip. The current and voltage contacts used in the experiments described above were 5-10 micron wide indium strips, evaporated onto the side of the crystal. Consequently, current injection is inhomogeneous and the electric field and CDW strain must be enhanced in the immediate vicinity of the current contact. Furthermore, the voltage contacts shunt part of the current out of the crystal so that the electric field and CDW strain are modified in their vicinity. To improve our measurements, we have used completely nonperturbing voltage contacts (metallized atomic force microscope tips) and we have ion implanted the crystal near the current contacts to pin the CDW strongly, so that CDW sliding and phase slip occur only where the electric field is uniform and purely longitudinal. The measured I_C - V_{ps} data have the same form as in Figure 2, and the $V_a(T)$ curves obtained by fitting this data have the same form as in Figure 3. Estimates of strain enhancements near side current contacts suggested that V_a values should be roughly 15% larger in the case of uniform fields. The experimental V_a values are slightly higher than those in Figure 2 and consistent with this estimate, although the scatter in the data is significant. Experiments using implanted contacts to test for the predicted length dependence of I_0 are currently in progress.

Finally, an important assumption in our analysis of transport measurements is that the excess voltage V_{ps} produces a strain in the CDW. We have directly measured this strain by high-resolution X-ray scattering.[3] We find that the CDW wavevector shifts longitudinally when electric fields above threshold are applied. The magnitude of the shift is largest near the current contacts, has opposite signs in the two halves of the crystal, and varies roughly linearly with position between the current contacts. Furthermore, the shift's variation with current and temperature is essentially the same as the measured variation of V_{ps} . These results confirm the identification of V_{ps} with the CDW strain.

6. Conclusion

The theory of Ramakrishna et al. provides a surprisingly good account of transport and X-ray scattering measurements of phase slip in NbSe₃. Future work will explore the dynamics of phase slip and its possible role in broadband noise generation, and the effects of phase-slip-related metastable CDW deformations on memory effects.

References

- [1] S. Ramakrishna, M. P. Maher, V. Ambegaokar, and U. Eckern, Phys. Rev. Lett. **68** (1992) 2066.
- [2] M. P. Maher, T. L. Adelman, S. Ramakrishna, J. P. McCarten, D. A. DiCarlo, and R. E. Thorne, Phys. Rev. Lett. **68** (1992) 3084.
- [3] D. DiCarlo, E. Sweetland, M. Sutton, J. D. Brock, and R. E. Thorne, Phys. Rev. Lett. **70** (1993) 845.
- [4] J. C. Gill, J. Phys. C **19** (1986) 6589.
- [5] P. Monceau, M. Renard, J. Richard, and M. C. Saint-Lager, Physica **143B** (1986) 49.
- [6] D. V. Borodin, S. V. Zaitsev-Zotov, and F. Ya. Nad', Sov. Phys. JETP **66** (1987) 793.
- [7] D. Feinberg and J. Friedel, in *Low-Dimensional Electronic Properties of Molybdenum Bronzes and Oxides*, edited by C. Schlenker (Kluwer, Academic, Dordrecht, 1989) 407.
- [8] P. A. Lee and T. M. Rice, Phys. Rev. B **19** (1979) 3970.