

# Transient structure of a driven charge-density wave in NbSe<sub>3</sub>

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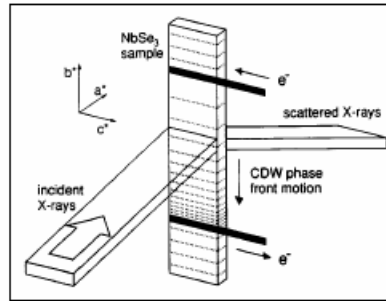
The fundamental statistical physics describing systems which are so far out of thermal equilibrium that the notion of a partition function is not valid is currently not well understood. The goal of this experiment is to study the structural response of a very simple system as it is driven between two distinct steady state configurations which are described by different partition functions, paying close attention to the transition regions. We chose to study the charge-density waves (CDWs) found in quasi-one-dimensional metals because the equilibrium properties of these systems are well understood. Experimentally, the structure of the stationary and the sliding CDW states have both been measured at high resolution. Theory and experiment are in excellent agreement<sup>1</sup>. On the other hand, the dynamics of CDW systems are relatively poorly understood. Although a large number of electronic transport experiments have been performed, the results of these experiments are difficult to interpret in

terms of microscopic models and time-resolved structural data are very limited.

In an effort to address these issues, we used a time-resolved, high-resolution x-ray scattering technique to measure the evolution of the structure of the sliding Q<sub>1</sub> CDW in NbSe<sub>3</sub>, as the direction of the driving electric field is reversed. In a full report of this work<sup>2</sup> we interpret our data using an equation of motion for the phase of the CDW order parameter which pertains at low temperatures and at large applied fields.

Found primarily in one-dimensional metal crystals, the CDW state is characterized by a periodic density wave appearing in the conduction electron density and a concomitant periodic longitudinal lattice-distortion wave. The properties of CDW systems have been studied extensively during the past 15 years. A large fraction of this research has been devoted to studies of the nonlinear electronic transport exhibited by some CDW systems.<sup>3</sup> This nonlinearity can now be explained simply. The incommensurate periodicities of the lattice and the CDW imply that there is no preferred location of the CDW in the crystal. Since there is no energetically preferred location, there can be no elastic restoring forces acting on the CDW. Consequently, an arbitrarily small applied field is able to accelerate the CDW, causing it to slide rigidly through the crystal lattice, producing a collective current.<sup>4</sup>

In any experimental realization, electrical contacts are required to



(Figure 1) Schematic illustration of the experimental configuration. Normal current is injected and extracted from the current electrodes. CDW or collective mode conduction takes place only in the region between the current electrodes.

produce the electric field which drives the CDW into the sliding state. The boundary conditions imposed on the CDW state at the electrical contacts play a crucial role in determining its sliding structure. The system is illustrated schematically in Fig. 1. The electric field is created inside the quasi-one-dimensional metal crystal by applying a voltage between two widely spaced electrodes. In the region between the electrodes, the applied voltage creates the electric field,  $E_{app} = V_{app} / L$ . In the regions of the sample which are outside of the two contacts, the electric field is zero. To achieve a steady-state collective current due to the CDW sliding, current must be injected at one electrode and extracted at the other. Equivalently, CDW phase fronts must be added at one electrode and removed at the other. In the figure above, the dashed lines indicate lines of constant phase of the CDW. In the region of the sample between the contacts, the CDW phase fronts are moving when the CDW is sliding. In the regions of the sample which are outside the electrical contacts, there is no collective



Joel D. Brock (left) and graduate student Emma Sweetland examine the tip of a refrigerator cylinder that is used to chill tiny metal samples for x-ray experiments. (Photo: Charles Harrington/Cornell University Photography)

current and the CDW phase fronts are stationary.

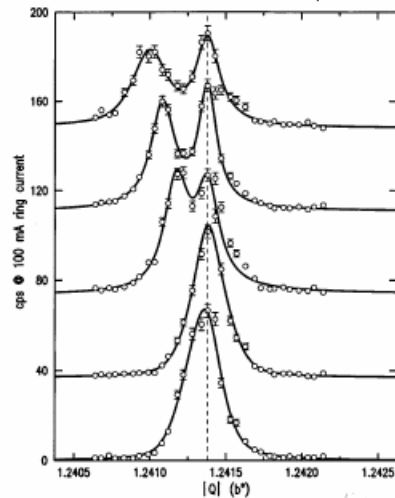
The mechanism which converts normal current to collective current at the electrodes is straightforward. Before the electric field is applied, the CDW is uniform. As the electric field is applied, phase fronts in between the electrodes try to move down, dilating the CDW near the electrode on the top. This strain can be relieved by nucleating a dislocation loop, which quickly grows to be the size of the sample, inserting a new phase front as it does so.<sup>5</sup>

A one-dimensional theory of the steady state current conversion process has been developed by Ramakrishna *et al.*<sup>6,7</sup> One of the specific predictions of this theory is that, as in the discussion above, the CDW should be strained in the region between the electrodes. Using synchrotron-based high resolution x-ray scattering techniques and the facilities at CHESS, DiCarlo *et al.*<sup>8</sup> have observed a position, temperature and electric field dependent strain of the sliding  $Q_1$  CDW in NbSe<sub>2</sub> consistent with this prediction. The goal of the present experiments is to measure the time dependence of this strain field as the sign of the electric field is

reversed.

The x-ray scattering measurements were performed at the F2 experimental station. The storage ring was running at an energy of 5 GeV and the stored positron current typically decayed from 80 to 40 mA during a 50 minute fill cycle. For the purposes of comparing different data sets, we have normalized the data to counts per second at 100 mA ring current. A Si(111) double-bounce monochromator selected a wavelength of 1.5 Å from the white x-ray beam produced by the 24-pole wiggler. A flat Au-coated mirror in the hutch suppressed harmonics of the fundamental wavelength passed by the monochromator. The sagittally-bent second monochromator crystal focused the x-ray beam in the out-of-scattering-plane direction at the sample position. Tantalum slits restricted the x-ray spot size at the sample to approximately 0.8 mm × 3 mm. The resulting x-ray beam contained  $4 \times 10^{10}$  x-rays/second/100mA of stored positron current. The scattered x-rays were analyzed by a triple-bounce channel cut Si(111) crystal and detected by a standard NaI(Tl) scintillator and photomultiplier tube.

To measure the transient structural response of the CDW, we utilized a stroboscopic detection system<sup>9</sup>. The CDW is subjected to a continuous square wave voltage wave form. The half period of the square wave is divided into time intervals of equal size. X-rays detected during a particular time interval are summed over a large number of square wave periods. Typical count rates at the



(Figure 2) A succession of (0 0) reciprocal space scans taken from different time slices of a data set taken at 17 mA. The centers of the time bins, measured from the rising edge of the driving wave form, are 0.014, 0.030, 0.050, 0.062, and 0.078 seconds. Successive scans are displaced by 35 cps @ 100mA.

peak of the (0 1+Q, 0) CDW satellite were on the order of 50 counts/second @ 100 mA of ring current. Therefore, in order to obtain reasonable counting statistics we sum over roughly 10,000 voltage wave form cycles.

All of the data we discuss here were taken at 70 K. The depinning current of this sample was determined by measuring the differential resistance as a function of current with a lock-in amplifier. The depinning current was  $20 \pm 3$  mA.

Close inspection of the data reveal that the CDW satellite peak does not have the same shape or width for the different current directions. This is most clearly seen at lower driving fields. For example, Figure 2 shows a series of longitudinal scans through the (0 1+Q, 0) CDW satellite from different time intervals as the current direction is switched. These data were taken at 70 K and with a driving current of 17 mA. Each scan is offset by 35 cps @ 100 mA for clarity. The top and bottom scans represent the steady-state for the two current directions. The remaining scans show that the CDW satellite peak evolves from a single peak to a bimodal line shape. Data sets taken with higher driving current magnitudes exhibited the same systematic structure. In general, as the driving current magnitude was increased, the area under the stationary peak decreased. At 29 mA, the stationary peak is no longer measurable. The width of the moving peak is slightly broader in the transient regions.

A simple interpretation of this data is that near threshold the CDW shears into two or more independent domains. Some portion of the system remains pinned and does not slide. The CDW in these pinned portions of the sample does not exhibit the time-dependent strain of the sliding portions. Therefore, the CDW satellite peak due to these pinned regions remains fixed. Thus, the depinning current measured above using the lock-in amplifier cannot be interpreted simply as the current at which the CDW is depinned over the

entire crystal.

Figure 3 shows the difference in position of the sliding and pinned peaks as a function of time for one complete period of the driving current wave form. The particular data set shown was taken at a current of 29 mA but is representative of the data taken at all currents. To characterize the time scales of these transients, we fit these relative peak positions to the solution of the driven diffusion equation.<sup>2</sup> The solid line in Figure 3 is the best fit to that solution. For the data set shown, the time constant  $\tau_0$  obtained for the switch to larger values of  $Q$  was  $2.58 \pm 0.37$  msec and for the switch to smaller values of  $Q$  was  $4.71 \pm 0.74$  msec.

In summary, we have used a time-resolved high-resolution x-ray scattering technique to measure the transient structural response of a simple quasi-one-dimensional CDW system as it is driven between two distinct steady state configurations. We find that a simple Langevin equation of motion for the phase of the CDW, which is based on the notion of a background strain which is driving the nucleation of dislocation loops, accurately describes the re-

sponse of the CDW to a reversal in the direction of an applied electric field, well above the threshold to sliding. As the driving field approaches threshold from above, the line shape becomes bimodal and the fraction of the CDW which is stationary increases monotonically. This suggests that, in this sample, the CDW did not depin throughout the entire sample at one well defined voltage.

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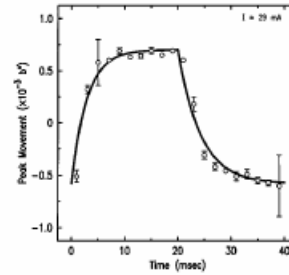
1 D. DiCarlo, R.E. Thome, E. Sweetland, M. Sutton, and J.D. Brock, (submitted to Physical Review B).

2 E. Sweetland, A.C. Finnefrock, W.J. Podulka, M. Sutton, J.D. Brock, D. DiCarlo, R.E. Thome, Submitted to Physical Review B.

3 For comprehensive reviews of the electronic properties of CDW systems see P. Monosau, in *Electronic Properties of Quasi-One-Dimensional Materials* (Reidel, Dordrecht, 1985), Pt. II, p. 139; G. Grüner, *Rev. Mod. Phys.* 60, 1129 (1988).

4 H. Fröhlich, *Proc. R. Soc. London, Ser. A* 223, 296 (1954).

5 For a comprehensive review of previous work on CDW phase slip, see F. Ya. Nad', in *Charge Density Waves in Solids*, edited by L.P. Gor'kov



(Figure 3) Positions of the moving peak are shown for a data set taken at a current of 29 mA ( $I_c \approx 20$  mA). The solid lines are the best fit to a driven diffusion equation.

and G. Grüner (Elsevier, Amsterdam, 1989), p. 189.

6 S. Ramakrishna, M. P. Maher, V. Ambegaokar and U. Eckern, *Phys. Rev. Lett.* 68, 2066 (1992). Note that the definition of  $z$  used in this paper differs from that used in the present one.

7 Satish Ramakrishna, *Phys. Rev. B* 48, 5025 (1993). Note that the definition of  $z$  used in this paper differs from that used in the present one.

8 D. A. DiCarlo, E. Sweetland, M. Sutton, J. D. Brock and R. E. Thome, *Phys. Rev. Lett.* 70, 845 (1993). Note that the definition of  $z$  used in this paper differs from that used in the present one.

Similarly, contrary to the present paper, this paper combines  $K_c$  and  $\Delta^2$  into one variable.

9 Similar techniques have been used to study  $K_0\text{MoO}_7$ . See, for example, T. Tamegai, K. Tsutsumi and S. Kagoshima, *Synth. Metals* 19, 923 (1987) and references therein.