Experiment versus the Classical Model of Deformable Charge-Density Waves: Interference Phenomena and Mode Locking

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The differential resistance of $NbSe_3$ measured in the presence of combined ac and dc electric fields exhibits interference features. Our experimental results for the dependence of these features on ac amplitude and frequency are in complete and dramatic *disagreement* with predictions of the classical model of deformable charge-density waves, and prove that pinning by impurities does not vanish at large fields and frequencies.

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Incommensurate charge-density waves (CDW's) are formed below a Peierls transition in many quasi one-dimensional metals, such as $NbSe_3$. Normally pinned to the lattice by the phase-dependent energy from impurity fluctuations, the CDW's gradually become depinned and begin to move and transport current when the applied electric field E exceeds a threshold value E_T.[1,2] Two competing theories have been developed over the past several years to account for the various phenomena associated with CDW depinning and transport. One is based on the treatment of CDW metals as macroscopic quantum systems, with depinning occurring by quantum tunneling over large distances in space (~ 1 μm) through a small pinning gap.[3] The other treats the CDW as a charged classical object ("rubber sheet") with internal degrees of freedom and subject to viscous friction.[4,5]

Among the most striking phenomena associated with CDW transport are the steps of constant CDW current, analogous to steps of constant voltage in the Josephson effect, observed on the dc I-V characteristic in the presence of an ac field.[6] These steps (and the corresponding peaks in the differential resistance dV/dI) are due to interference or mode locking of the applied frequency ω with the internal CDW current oscillation frequency ω_n; both harmonic ($\omega_n/\omega = p$) and subharmonic ($\omega_n/\omega = p/q$) steps are observed.[7] Mode locking is said to be complete when the dc CDW current is constant for some finite range of dc voltage, so that the peak in dV/dI attains the value of the normal electron resistance which shunts the CDW.[8] Initial studies[6-8] found that most peaks in dV/dI do not lock completely, and that complete locking is seldom observed for applied frequencies above 10 MHz or in crystals longer than 1 mm.

Earlier, it was shown[9] that ac-dc interference experiments can be accounted for in terms of a simple model in which the only variables are the space-average phase, $\theta(t) = \langle \phi(x,t) \rangle_x$, and a phase-dependent pinning potential, $V(\theta)$, motivated by the tunneling theory. In contrast, a recent Letter by Coppersmith and Littlewood[10] claims to show, with calculations based on the classical deformable CDW model, that mode locking "arises from the presence of many metastable pinned states." Complete mode locking occurs only when "significant relaxation takes place while the field is below the threshold field." They also claim to show explicitly that a single-coordinate description cannot account for experiment. Here new experiments are reported which are in complete and dramatic *disagreement* with predictions of the classical deformable CDW model. The experiments are, however, in excellent agreement with the tunneling theory given in Ref. 3.

We have recently completed a detailed experimental study of ac-dc interference phenomena in $NbSe_3$.[11] Crystal defects other than isolated impurities and contact effects are observed to result in nonuniform current and velocity distribution within CDW crystals. These velocity distributions, which are not included in most theories of CDW transport, play a determining role in the decay of transient "ringing" in the voltage response to current pulses[12] and in broad-band noise generation.[13] They also affect the mode-locking phenomena. In particular, complete locking can only be observed if the intrinsic voltage width of a step is wider than the voltage increment corresponding to the CDW velocity distribution. By selecting crystals which have no visible defects, and by mounting and contacting them carefully, we have obtained samples which display exceptional velocity coherence. Using these crystals, we have shown that the widths of both the harmonic and subharmonic steps exhibit Bessel-function–type oscillations with ac amplitude and frequency. We have made the first observation of similar oscillations in the dc threshold voltage ($p = 0$

step), and have observed complete harmonic and subharmonic locking in samples much longer than have previously been reported. In this Letter, we report a major extension of these measurements to much higher ac amplitudes and frequencies in order to test the classical deformable-CDW model.

Coppersmith and Littlewood[10] (CL) make the following predictions for ac-dc interference and mode locking within the classical model: (1) Damped relaxation of the CDW below threshold is crucial to mode locking. Complete mode locking occurs only when the ac frequency and amplitude are small enough so that significant relaxation can take place while the total field is *below* the dc threshold field E_T. (2) For large ac amplitudes and frequencies such that the time interval $\delta t \approx 2\pi E_T/\omega E_{\mathrm{ac}}$ spent below threshold is small, dV/dI should exhibit peaks but not complete locking. Further, the "wings" (sharp negative dips in dV/dI immediately adjacent to the peak) predicted by single-coordinate models should not be observed; dV/dI should rise smoothly as the peak is approached from either direction.

The results of our experimental tests of these predictions are summarized in Figs. 1 and 2. The measurements were performed on a crystal of $\mathrm{NbSe_3}$ mounted in a two-probe configuration, with a separation between contacts of 2.5 mm. The dc threshold voltage, $V_T=21$ mV at 117 K, indicates that this crystal was of ordinary purity. The Q of the velocity distribution deduced from current-oscillation spectra for $\omega_n=50$ MHz was 5000. Our major findings are as follows:

(1) In Fig. 1, the 1/1 peak in dV/dI (as well as the 1/2 and 1/3 peaks) exhibits complete locking and sharp negative wings adjacent to the peak. The dc voltage at the 1/1 step is 232 mV, and the peak amplitude of the applied 50-MHz ac voltage is 80 mV. The minimum time-varying voltage when biased at the 1/1 step is thus 152 mV, *7 times greater than the (zero-ac) dc threshold voltage.* Damped relaxation of the CDW below threshold is thus *completely irrelevant* to mode locking.

(2) Figure 2 shows experimental data taken in the limit of large ac amplitude and high frequency. Here the applied frequency is 200 MHz, 20 times the highest frequency at which complete locking has previously been reported. The value of $\delta t \approx 2\pi E_T/\omega E_{\mathrm{ac}}$ for the experimental data shown in Fig. 1 of Ref. 11 was 1.6×10^{-9} s, and is within the stated regime of validity of CL's calculation. The value of δt for the experimental data of Fig. 2 is 2.1×10^{-10} s, a factor of 8 further into CL's assumed regime of validity. This time corresponds to a frequency 3 orders of magnitude greater than the measured dielectric relaxation frequency and 2 orders of magnitude greater than the classical "crossover" frequency in our

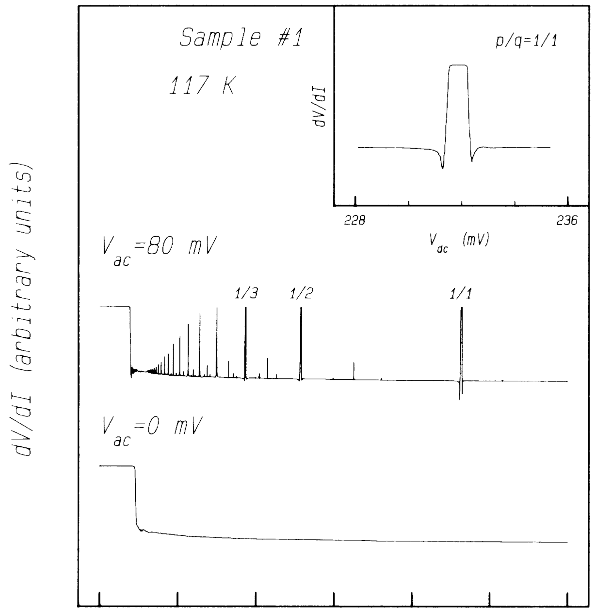

FIG. 1. Differential resistance dV/dI vs dc voltage of a 2.5-mm-long $\mathrm{NbSe_3}$ sample measured in the presence of an 80-mV peak, 50-MHz ac voltage. The 1/1 interference peak (shown on an expanded scale in the inset) locks completely and has sharp negative wings.

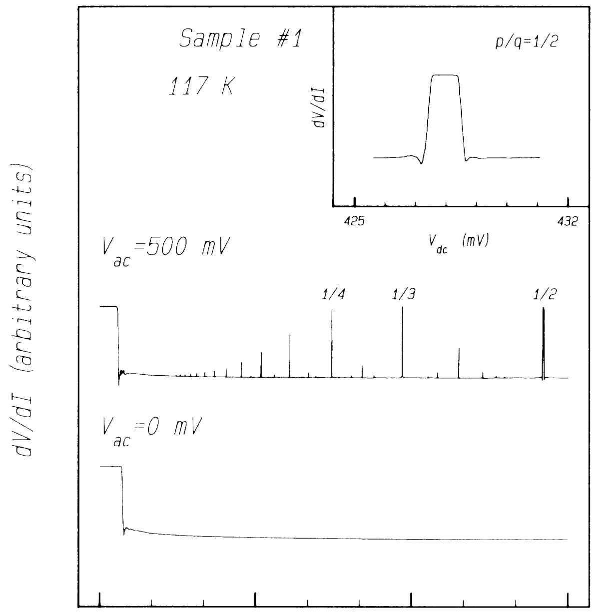

FIG. 2. Differential resistance dV/dI vs dc voltage measured in the presence of a 500-mV peak, 200-MHz ac voltage. The 1/2 interference peak (shown on an expanded scale in the inset) locks completely and has sharp negative wings.

$NbSe_3$ crystals. *The 1/3 and 1/2 steps lock completely and the 1/2 step has sharp negative wings, in complete disagreement with CL's prediction.* We have also observed complete locking and negative wings at 300 MHz on the 1/3 step, corresponding to a value of $\delta t \approx 1.1 \times 10^{-10}$ s.

We observe complete harmonic and subharmonic locking over an extremely broad range of ac frequencies and amplitudes on both CDW transitions in $NbSe_3$, limited at high fields and frequencies by Ohmic heating effects. Whenever complete locking is not observed, it is always consistent with the measured width of the velocity distribution becoming larger than the expected step width.

Figure 3 shows our experimental test of the most well-known prediction of the classical deformable CDW model: The high-field dc CDW conductance is predicted to be of the form $G_{\mathrm{CDW}}(E) = a - bE^{-1/2}$, where a and b are constants.[4] The data, measured in a highly coherent $NbSe_3$ sample on the upper CDW transition, extend from 2 to 150 times the dc threshold field. As was shown in Ref. 4, the predicted form (indicated by the dashed line in Fig. 3) provides a reasonable fit to experiment for fields between 3 and 15 times threshold. The perturbation calculation of Ref. 4 is not valid at such low fields, however, and at higher fields we observe significant deviations from $E^{-1/2}$ behavior. Although measurements at even higher fields would be desirable, there is currently *no* experimental evidence which supports this prediction of the classical deformable CDW model. In contrast, we have shown[14] that the Zener form predicted by the quantum tunneling theory[3] (indicated by the solid curve in Fig. 3) provides an exact fit to experiment for fields between 2 and up to 150 times threshold at all temperatures on the upper transition in $NbSe_3$.

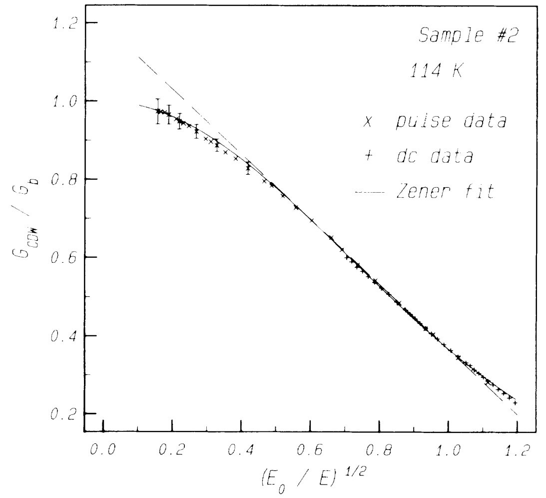

FIG. 3. dc CDW conductance vs $(E_0/E)^{1/2}$ for a highly coherent 2-mm-long $NbSe_3$ crystal. Significant deviation from the asymptotic behavior predicted by the classical deformable CDW model (dashed line) is observed at high fields. The solid curve is a fit to the Zener form $G_{\mathrm{CDW}} = G_b \exp(-E_0/E)$ of the tunneling model using $E_0 = 215$ mV/cm, $E_T = 75$ mV/cm, and $G_b = 1.554$ mS.

We believe that the failure of CL's calculations to account for our mode-locking experiments reflects a fundamental deficiency of the model[4] on which they are based. This model describes the CDW as an extended elastic medium which executes overdamped motion and deforms in the presence of impurities. When the CDW moves, the deformations continuously relax toward the lowest-energy configuration, thereby maintaining a net impurity pinning energy. When the CDW moves very quickly, significant relaxation cannot occur, so that the deformations and the pinning energy vanish.[15] The characteristic frequency for this relaxation, the CDW dielectric relaxation frequency, is measured to be roughly 5 MHz in $NbSe_3$,[16] consistent with simple theoretical estimates. Our observations of complete harmonic and subharmonic locking at very large ac frequencies and amplitudes therefore indicate that *the strength of CDW pinning is undiminished from its value at small fields and frequencies in a regime where significant relaxation cannot occur within the classical model.* Three additional pieces of experimental evidence support this contention. First, the maximum width in voltage of the 1/1 step versus ac amplitude, which should for high frequencies become proportional to the maximum magnitude of the pinning force, is indeed observed to saturate at high frequencies.[11] Second, the harmonic content and magnitude of the current oscillations, which are again determined by the magnitude and shape of the pinning force, also saturate at high dc fields.[12,13] Third, the high-field dc CDW conductances of $(TaSe_4)_2I$ and $K_{0.3}MoO_3$ are observed to be temperature activated and limited to approximately the value of the normal Ohmic conductance over a very broad temperature range.[17] In these materials, dissipation due to normal-carrier screening of the CDW charge fluctuations associated with pinning reduces the conductivity of the collective mode.[18] The temperature-activated high-field CDW conductance thus indicates that the charge fluctuations, and therefore the pinning, persist undiminished at very high electric fields. Together, *these experimental results prove that periodic CDW pinning by impurities does not vanish at large fields and frequencies, disproving the most fundamental prediction of the classical deformable-CDW model.*

In contrast, nonvanishing pinning at high fields is completely consistent with the quantum tunneling theory. In this theory, acceleration by tunneling between pinned states allows the pinning energy to be maintained. We have recently shown[11,13] that a simple single-coordinate model, motivated by the tunneling theory, for a CDW moving in a nonsinusoidal periodic potential provides an excellent and *semiquantitative* account not only of the ac-induced steps in the dc *I-V* characteristic, but also of the "dips" and "jumps" in the bias-dependent ac conductance,[19] the current oscillations,[2] and the inductive loops seen in the response to large ac voltages.[20] The present results show that the two predictions of the classical deformable CDW model for ac-dc interference which differ qualitatively from those of our single-coordinate picture are in clear disagreement with experiment. The failures of the classical model described here prove that it is fundamentally deficient. This provides strong additional evidence for the hypothesis that quantum tunneling underlies CDW dynamics.

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