

ac-dc interference, complete mode locking, and origin of coherent oscillations in sliding charge-density-wave systems

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We present the results of a detailed experimental study of ac-dc interference phenomena in charge-density-wave (CDW) transport. The dc I - V characteristic of high-quality NbSe₃ crystals, measured in the presence of an applied ac voltage, exhibits a remarkable array of Shapiro steps, including at least 150 subharmonic steps between the dc threshold and the 1/1 harmonic step. The harmonic and low-order subharmonic steps mode lock completely for ac frequencies between 100 kHz and 300 MHz and for ac amplitudes from a fraction of the dc threshold voltage V_T to $75V_T$. The form and magnitude of the Bessel-like oscillations of the harmonic step widths with ac amplitude are independent of applied frequency at frequencies above 20 MHz, demonstrating conclusively that periodic CDW pinning by impurities does not vanish at high fields and frequencies. Surprisingly, even though CDW's are randomly pinned many-degree-of-freedom systems, single-coordinate models reproduce nearly all qualitative features of the Shapiro steps. A comparison between our interference measurements and our earlier coherent oscillation ("narrow-band noise") measurements yields strong evidence that both effects are of bulk origin. We also have observed mode locking in the frequency domain by measuring the coherent oscillation spectrum in the presence of an applied ac voltage. When mode locking occurs, the spectral width of the fundamental peak collapses dramatically, indicating that the CDW's motion becomes almost perfectly periodic with no fluctuations. However, the amplitude of the fundamental does not change, suggesting that phase correlations are not dramatically enhanced. A detailed understanding of these results should yield important insights into the dynamics of charge-density waves and of other many-degree-of-freedom systems.

I. INTRODUCTION

Collective motion of charge-density waves (CDW's) in quasi-one-dimensional conductors such as NbSe₃ results in some of the most remarkable transport effects ever discovered.¹ To minimize their energy of interaction with randomly distributed impurities, incommensurate CDW's deform, losing their translational symmetry and becoming pinned to the lattice.² Above a threshold electric field E_T the CDW becomes depinned and transports charge, resulting in nonlinear dc conduction at fields of millivolts per centimeter.^{3,4} At megahertz frequencies, strongly frequency-dependent ac conduction is observed.^{3,5}

Since the possible pinned configurations do not change when the CDW's space-averaged phase is displaced by $2n\pi$, the CDW's impurity pinning energy is expected to be periodic in such displacements. When a dc voltage (current) is applied, the current (voltage) response contains a coherent oscillating component,^{4,6} which, for historical reasons, is referred to as narrow-band noise (NBN). The Fourier spectrum of these coherent oscillations consists of a fundamental peak, whose frequency ω_n is proportional to the dc CDW current (i.e., the time-averaged CDW velocity) and rich array of harmonic peaks. Coherent oscillation phenomena are directly analogous to the ac Josephson effect, with the roles of current and voltage interchanged.

The most striking manifestations of phase-dependent CDW interaction with impurities are the interference effects observed when a voltage $V(t) = V_{dc} + V_{ac}\cos(\omega t)$ is applied. For example, the dc I - V characteristic exhibits steps where the dc CDW current remains constant or nearly so while the dc voltage varies.⁷ Analogous to steps of constant voltage observed in Josephson junctions, they are commonly referred to as Shapiro steps. These steps are due to mode locking of the internal NBN oscillation frequency ω_n associated with time-averaged CDW motion with the applied ac frequency ω . Both harmonic steps, for which $\omega_n/\omega = p$, and subharmonic steps, for which $\omega_n/\omega = p/q$ ($\neq r$), are observed. A step in the I - V characteristic is said to be *completely* locked if the dc CDW current is indeed constant for some finite range of dc bias.⁸ On most steps the current varies somewhat with dc bias, so that the mode locking is incomplete.

We have earlier published two papers, hereafter referred to as I (Ref. 9) and II (Ref. 10), which are of direct relevance to the present work. Paper I described detailed measurements of coherent oscillation phenomena. These measurements indicated that the effective periodic impurity pinning potential is highly nonsinusoidal, and that its shape varies little with applied field. Paper II described detailed measurements of ac-dc interference phenomena. The widths of the mode-locked steps in the dc I - V characteristic were found to exhibit Bessel-type oscillations with ac amplitude and frequency.¹¹ A single-

coordinate model of CDW motion in a nonsinusoidal potential¹² provided a good qualitative and semiquantitative account of these results.

Here we present additional results for ac-dc interference in NbSe₃ samples of exceptional quality, which yield important new insights into the dynamics of charge-density waves and related many-degree-of-freedom systems. A remarkable array of harmonic and subharmonic steps are observed on the dc I - V characteristic, whose quality rivals that observed in Josephson junctions. These steps mode lock completely and have the form predicted by single-coordinate models over almost the entire experimentally accessible range of ac amplitudes and frequencies. The form and magnitude of the harmonic step-width variation with ac amplitude is independent of applied frequency above 20 MHz, indicating that periodic CDW interaction with impurities does not vanish at high fields and frequencies. Measurements of mode-locking in the frequency domain indicate that CDW velocity coherence is dramatically increased by mode locking, but that there is no similar dramatic increase in phase coherence. A detailed understanding of these results for randomly pinned CDW's is presently lacking.

This paper draws heavily on the results and analysis contained in papers I and II. In the interest of brevity, we have attempted to minimize duplication of discussion; the earlier papers should be consulted for additional details and clarification. Some preliminary results of the present work have appeared elsewhere.¹³

II. EXPERIMENTAL METHODS

The material chosen for this study was NbSe₃, because it shows the effects of ac-dc interference most clearly. NbSe₃ undergoes two independent CDW transitions at 145 and 59 K, respectively, each removing approximately half of the Fermi surface. Crystals of modest purity [a residual resistance ratio of ~ 130 , $E_T(T=120\text{ K}) \sim 60\text{--}80\text{ mV/cm}$] were mounted in a two-probe configuration on a 50- Ω microstrip and placed in an open-cycle refrigeration system with a helium exchange gas. ac and dc voltages were applied using a Marconi 2022 signal generator and a Systron-Donner M107 precision dc voltage source, respectively, and the differential resistance dV/dI was measured using a Princeton Applied Research 5301 lock-in amplifier. Narrow-band noise spectra were measured using a Hewlett-Packard 8558B spectrum analyzer. All measurements were automated through the use of an IBM AT computer.

The differential resistance dV/dI was measured instead of the I - V characteristic because it provides much more sensitive detection of subharmonic steps. Since the CDW resistance R_{CDW} in NbSe₃ is shunted by an Ohmic resistance R_N arising from ungapped portions of the Fermi surface and from quasiparticle excitations across the Peierls gap, the steps of constant CDW current appear as peaks in dV/dI which rise to the value of the shunt resistance R_N .

As was extensively discussed in papers I and II, macroscopic crystal defects and contacts break CDW velocity coherence, resulting in a nonuniform CDW current dis-

tribution, and can play a crucial role in the observed response. In order to elucidate the fundamental transport properties of an impurity-pinned CDW, we have developed sample preparation, selection, and mounting techniques which minimize these effects of defects and contacts. NbSe₃ crystals are identified which have uniform cross sections and which are free of visible defects. One such crystal is carefully removed from the growth, cut to a length of 5–10 mm, and transferred to the microstrip sample mount. Silver paste contacts are then applied to a defect-free portion, avoiding portions damaged by the cuts.

Using this technique, we have obtained NbSe₃ samples with extremely homogeneous current distributions and unprecedented coherence, whose response likely approximates that of a CDW pinned only by impurities and other microscopic defects. The uniformity of the current distribution is best characterized by the spectral width of the NBN fundamental: the fundamental frequency is proportional to the current density, so that the spread in frequencies reflects the spatial distribution and temporal fluctuations of the current density. Figure 1 shows the fundamental spectral width $\delta\omega$ versus fundamental frequency ω_n for a 2.3-mm-long NbSe₃ sample at 117 K. For fundamental frequencies above 10 MHz, the spectral width varies little with frequency and is approximately 7 kHz. Many samples have fundamentals comprised of a few very sharp peaks, or of a single peak but only at low frequencies and dc biases. The remarkable feature of this sample is that the full spectral weight of the fundamental at $\omega_n/2\pi = 200\text{ MHz}$ and $V_{\text{dc}} \sim 50V_T$ is contained within

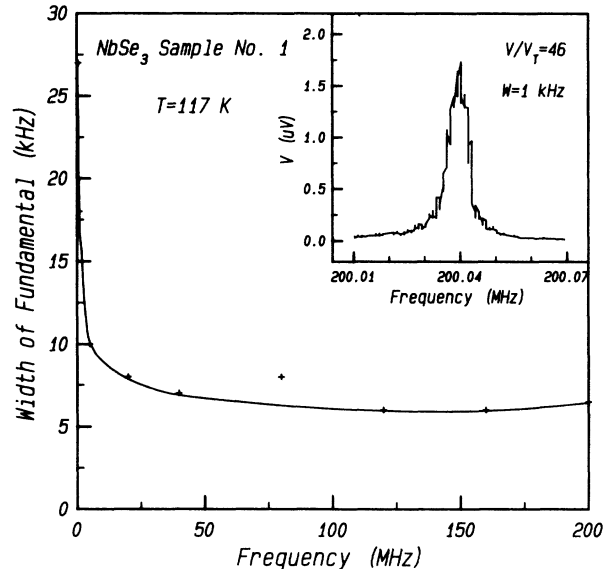


FIG. 1. Spectral width of the NBN fundamental vs fundamental frequency ω_n , for an exceptionally coherent NbSe₃ sample at $T = 117\text{ K}$. The solid line is a guide to the eye. The inset shows the spectrum of the fundamental at $\omega_n/2\pi \sim 200\text{ MHz}$, measured using a 1-kHz spectrum analyzer bandwidth. The spectral width is approximately 7 kHz, corresponding to a $Q \equiv \omega/\delta\omega \sim 30000$.

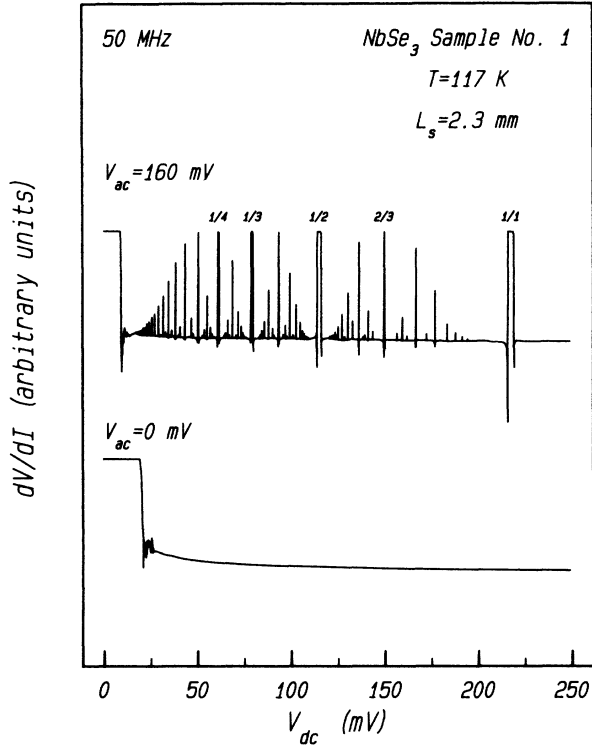


FIG. 2. Differential resistance dV/dI vs dc voltage measured in the presence of a 50-MHz, 160-mV peak ac voltage. The 1/5, 1/4, 1/3, 2/5, 1/2, 2/3, and 1/1 peaks are completely mode locked. Between threshold and the 1/2 step, 96 subharmonic steps, down to $p/q = 1/27$, were observed.

a single peak whose $Q \equiv \omega_n/\delta\omega \sim 30\,000$. The exceptional coherence of this sample is manifested in an extraordinary array of interference peaks in the differential resistance. As shown in Fig. 2, the 1/5, 1/4, 1/3, 2/5, 1/2, 2/3, and 1/1 peaks are all completely mode locked (i.e., dV/dI rises precisely to the value of R_N , the shunt Ohmic resistance measured below threshold). Between threshold and the 1/2 peak, at least 96 subharmonic peaks were visible, down to the $p/q = 1/27$ peak. As the ac amplitude was varied, all steps between threshold and the 1/1 step with $q \leq 5$ locked completely.

III. RESULTS AND ANALYSIS

A. Conditions for complete mode locking

The conditions under which complete mode locking can be observed in the response of an “ideal,” impurity-pinned CDW is an issue of considerable experimental and theoretical interest. In real CDW crystals, the interference features are smeared out by the spatial and temporal CDW current-density distributions associated with defects and contacts and also with thermal gradients induced by Ohmic heating when large fields are applied. For a given dc bias, this distribution, which is reflected in the spectral width of the NBN fundamental, corresponds to a width δV in dc voltage. Assuming that the intrinsic response of a velocity-coherent CDW is to lock complete-

ly, complete locking will only be observed if the intrinsic step width is larger than δV corresponding to the current distribution. (Although this is an oversimplification,¹⁴ the experiments are generally consistent with this relation.) For samples of ordinary quality with broad NBN fundamental widths, most peaks in dV/dI are rounded, exhibiting neither the complete locking nor the negative “wings” adjacent to the mode-locked peaks evident in the data of Fig. 2. (These “wings” result because the dc CDW current jumps abruptly to the mode-locked value on either side of the step.) Even for the high-quality samples used in paper II, complete locking of the 1/1 and 1/2 steps was only observed for applied frequencies between 2 and 30 MHz, and for a limited range of ac amplitudes.

Using exceptionally coherent NbSe₃ samples we have performed a more meaningful evaluation of the ac amplitude and frequency regime for complete mode locking. Figure 3 shows data measured in the low-frequency limit. The applied ac frequency is 200 kHz, roughly an order of magnitude smaller than the dielectric relaxation frequency and 2 orders of magnitude smaller than the crossover frequency ω_{co} (defined as the frequencies of the peaks of the real part of the dielectric constant and in the imaginary part of the ac conductance, respectively). The 1/1, 2/1, and 3/1 steps lock completely. At very low frequencies, the low-order (small p) harmonic steps occur at fields very near threshold and are closely spaced, so that their maximum width is much smaller than at higher frequencies. Further, the effects of crystal imperfections and contacts are relatively more important at low fields, and the width of the current distribution (NBN fundamental)

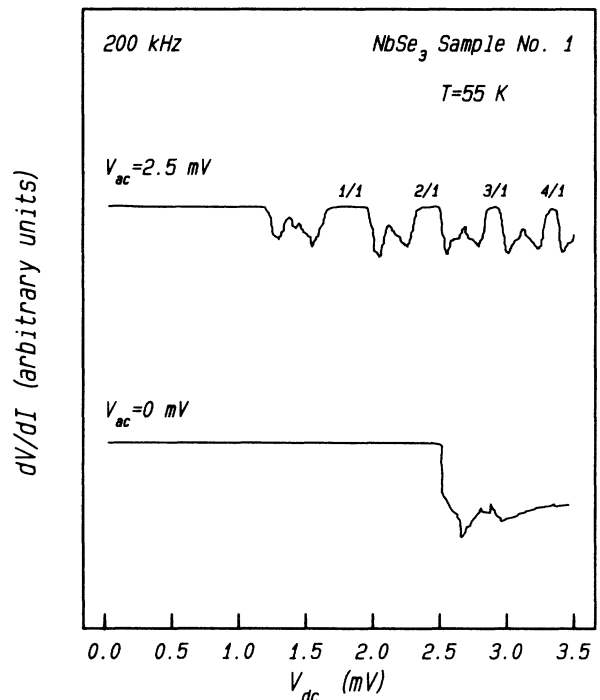


FIG. 3. Differential resistance vs dc voltage measured in the presence of a 200-kHz, 5-mV peak ac voltage. The 1/1, 2/1, and 3/1 peaks mode lock completely.

is large and eventually becomes larger than the intrinsic step width. Complete locking thus was not observed below 100 kHz. At very low frequencies, the peaks in dV/dI wash out into a continuum and dV/dI decreases smoothly above threshold.

Figure 4 shows data measured in the small ac amplitude regime, where the applied ac voltage is much smaller than the dc voltage at which a step occurs. Even though the peak ac voltage is only one-third of the dc bias at the 1/1 step and the total time-varying voltage is always at least 7 times greater than the zero-ac dc threshold, the 1/1 step locks completely. In this regime, the step width is roughly proportional to the applied ac voltage. For smaller ac voltages, the step width became narrower than the voltage increment corresponding to the measured current distribution, and complete locking was not observed.

Figure 5 shows data measured in the high-frequency limit. The applied ac frequency is 200 MHz, roughly 2 orders of magnitude greater than the dielectric relaxation frequency, an order of magnitude greater than the crossover frequency ω_{co} , and within a factor of 5 of the estimated pinning frequency. The 1/3 and 1/2 steps both lock completely. Complete locking was also observed on the 1/3 step at 300 MHz, for a peak ac amplitude of 610 mV. The peak magnitude of the total applied voltage (ac + dc) in the latter case was $\sim 50V_T$; measurements at higher frequencies were not attempted because of poten-

tial damage due to Ohmic heating. At high frequencies the step width becomes small compared to the dc bias at which the step occurs, so that in order to observe complete locking the width of the NBN fundamental (current distribution) must be very small compared to the fundamental frequency (mean current). Since in most samples the width of the NBN fundamental increases with increasing bias, observing complete locking at high frequencies is very difficult.

Figure 6 shows data measured in the large ac-amplitude limit. The ac frequency is 50 MHz and the peak ac amplitude, 200 mV, is 75 times greater than the zero-ac dc threshold V_T and 6 times greater than the dc bias near the 1/1 step. Again, the 1/1 peak locks completely.

Similar results were obtained as a function of frequency and ac amplitude for both CDW's in NbSe_3 , with two exceptions. First, the negative wings adjacent to the mode-locked peaks tended to be much sharper for the CDW which forms below 59 K. This could result because thermal fluctuations, which may perturb the CDW and result in intermittent loss of mode lock near the edge of a step and thus in rounding of dV/dI , are less important at low temperatures. Second, the low-temperature CDW did not lock completely at very high frequencies (above 80 MHz). The NBN fundamental width for this CDW always broadened substantially at large dc biases, so that the current distribution became larger than the step width at high frequencies. The reason for this broadening is not clear, but its onset appeared to coincide with

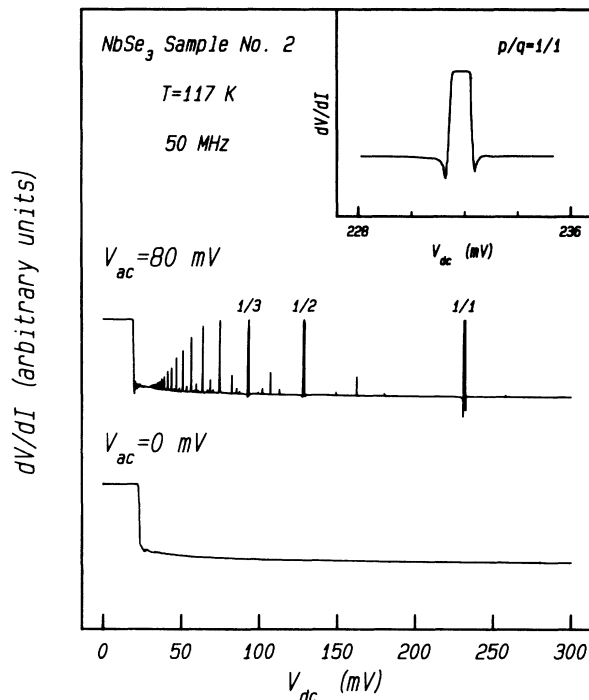


FIG. 4. Differential resistance vs dc voltage measured in the presence of an 80-mV peak, 50-MHz ac voltage. The 1/1 interference peak (shown on an expanded scale in the inset) as well as the 1/2 and 1/3 peaks lock completely. The total (ac + dc) voltage at the 1/1 peak is always at least 7 times greater than the zero-ac dc threshold voltage.

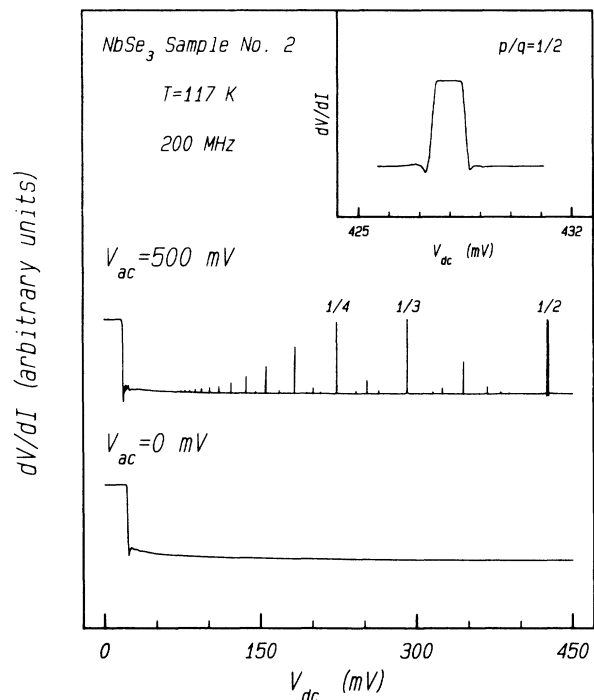


FIG. 5. Differential resistance vs dc voltage measured in the presence of a 500-mV peak, 200-MHz ac voltage. The 1/2 interference peak (shown on an expanded scale in the inset) locks completely.

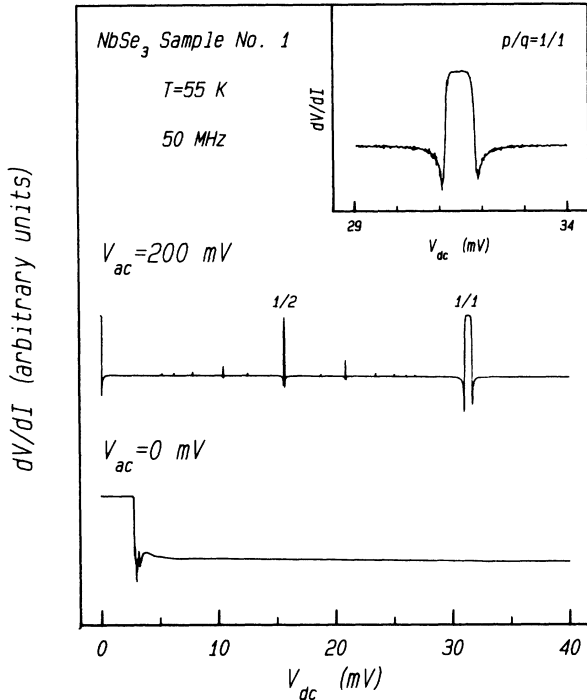


FIG. 6. Differential resistance vs dc voltage measured in the presence of a 200-mV peak, 50-MHz ac voltage. The 1/1 interference peak (shown on an expanded scale in the inset) locks completely and has sharp negative wings. The peak ac amplitude is 75 times the zero-ac dc threshold voltage and 6 times the dc bias at which the 1/1 step occurs.

the onset of NBN due to depinning of the CDW which forms below 145 K. The broadening may thus be associated with interaction between the two CDW's. It should also be noted that all of the measurements discussed here were made at temperatures which yielded the narrowest NBN spectral width and therefore the most coherent response. At other temperatures, the NBN fundamental broadened,⁹ smearing out the interference features and making observation of complete locking more difficult or impossible.

In summary, complete mode locking to an applied ac field is observed over an extremely broad range of ac frequencies (100 kHz to 300 MHz) and amplitudes (up to $75V_T$), limited at high fields and frequencies by Ohmic heating effects. The form of the completely mode-locked peaks in dV/dI —flat tops with sharp negative “wings”—is identical to that predicted for single-coordinate locking, as in the rigid overdamped oscillator model¹⁵ and the resistively shunted junction (RSJ) model.¹⁶ Whenever complete locking is not observed, the expected step width is always smaller than the voltage δV corresponding to the measured NBN spectral width. Complete mode locking thus appears to be a fundamental feature of the CDW response. However, even “ideal,” impurity-pinned CDW's likely have some minimum step width for complete locking, perhaps due to thermal fluctuations. Residual defects and contact effects make even our best crystals unsuitable for determining this minimum width.

B. High-frequency variation of the step width and pinning force

The mode-locked step-width variation with applied frequency yields important information about the dynamic interaction between the CDW and pinning centers. As discussed in paper II and illustrated in Fig. 7, the step width exhibits Bessel-type oscillations with ac amplitude. At low frequencies, the maximum step width is limited by the spacing in dc bias between the steps, which vanishes as the applied frequency goes to zero. At high frequencies the spacing between harmonic steps becomes large, and the step width is limited by the maximum time-averaged pinning force which can be developed.

The single-coordinate analysis of Ref. 12 is useful in interpreting experiments in the high-frequency regime. This analysis assumes that the CDW is biased with dc and ac currents, and that the space-averaged CDW phase θ moves in a nonsinusoidal periodic potential $V(\theta)$. With no mode locking, the time-averaged pinning force due to this motion is small. When mode locking occurs, the time-averaged pinning energy is lowered, and the time-averaged pinning force varies so as to precisely cancel changes in the applied dc bias, thereby keeping the dc CDW current constant. The width of the p/q step, calculated as twice the maximum magnitude of the time-averaged pinning force, is given by¹²

$$\delta V/V_T = \max_{\theta} \left[2 \sum (q\alpha) a_q J_p \left[\frac{q\alpha\omega_1}{\omega} \right] \sin(q\alpha\theta) \right], \quad (1)$$

where $(q\alpha)a_q$ is the q th Fourier component of the pinning force, J_p is the p th-order Bessel function, ω_1 is proportional to the amplitude of the applied ac current, and

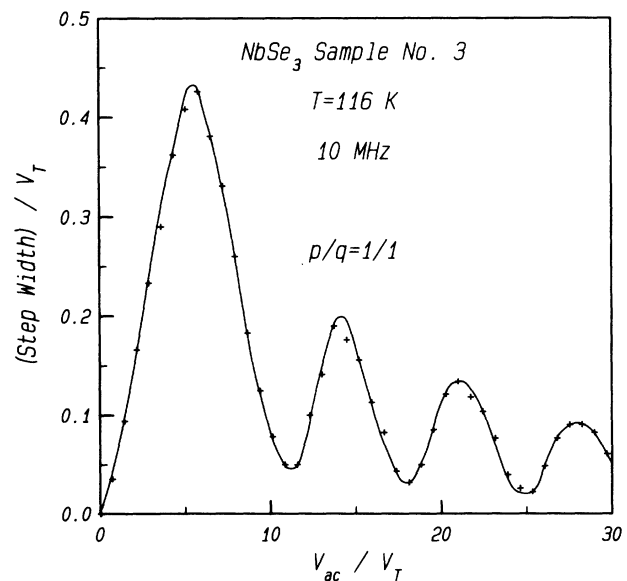


FIG. 7. Voltage width of the 1/1 step vs peak ac amplitude, normalized by the zero-ac dc threshold voltage, for an applied frequency of 10 MHz. The decay of the oscillations with increasing ac amplitude is larger (by roughly a factor of 2) and the minima are much more rounded than is predicted by single-coordinate models. The solid line is a guide to the eye.

α is 1 or 2 for pinning periodicities of 2π or π , respectively. Since the sum is over all p and q such that $\omega_n/\omega = p/q$, the width of the p/q step depends only upon pinning-force Fourier components of order q and higher. Experimentally, the widths of the steps decrease fairly quickly with increasing q , indicating that the Fourier components of the pinning force also decrease quickly. Thus, a reasonable approximation is to retain only the first term in Eq. (1); the width of the p/q step is then proportional to the magnitude of the q th Fourier component of the effective pinning force.

Figure 8 shows the maximum ac-amplitude-dependent width of the 1/1 step (i.e., the magnitude of the first step-width maximum versus ac amplitude) versus ac frequency. The lower curve indicates the completely mode-locked width, while the upper curve is the width obtained by integrating under the peak in dV/dI , which compensates for rounding near the edges of the step due to the finite width of the current distribution. At high frequencies the maximum width saturates, indicating that the magnitude of the fundamental component of the effective pinning force also saturates.

As discussed in paper I and shown much more convincingly in Fig. 9, a similar saturation of the magnitude of the NBN fundamental at high fundamental frequencies (dc fields) is observed. In a single-coordinate model, the fundamental amplitude is proportional to the magnitude of the fundamental component of the pinning force. But real CDW's are many-degree-of-freedom systems, and the NBN is an incoherent sum of oscillations in different regions. Any variations in the number of oscillators or in the phase correlations between oscillators with electric

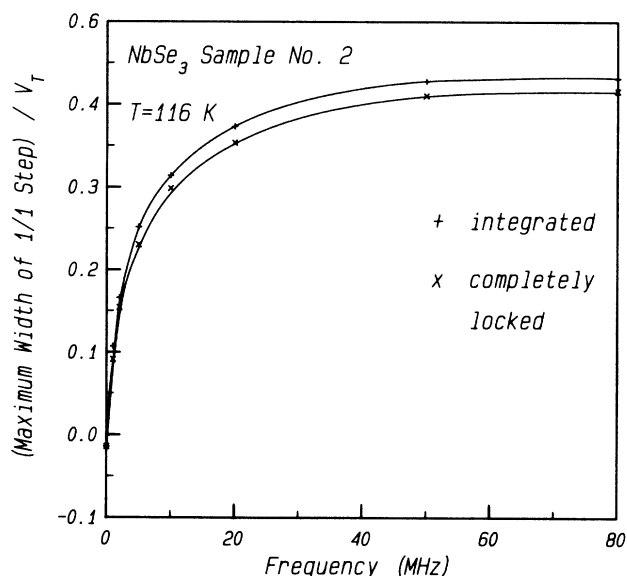


FIG. 8. Maximum ac-amplitude-dependent width of the 1/1 step vs ac frequency. The upper curve is the step width obtained using the integrated measure described in the text, while the lower curve is the completely locked width. At high frequencies, the step width saturates to a constant value. The solid lines are guides to the eye.

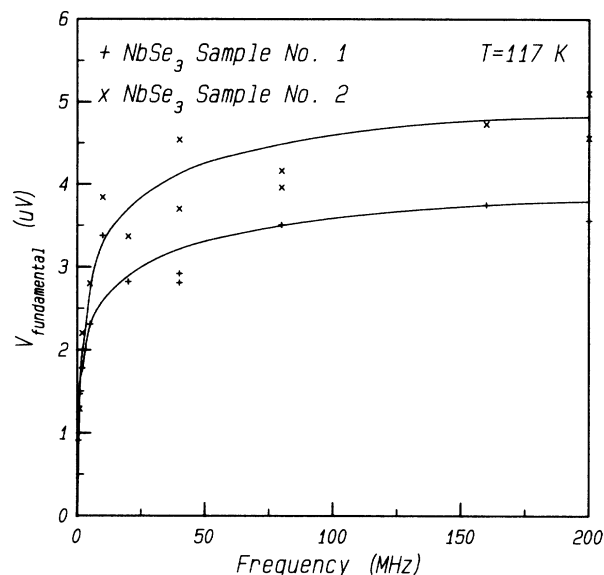


FIG. 9. Magnitude of the NBN fundamental vs fundamental frequency ω_n , measured using a spectrum-analyzer bandwidth much larger than the fundamental spectral width. At high fields (fundamental frequencies), the fundamental amplitude saturates. The solid lines are guides to the eye.

field, including those variations associated with macroscopic crystal defects, would be expected to affect the NBN amplitude. For this reason, no conclusions regarding the pinning force could be drawn in paper I from NBN measurements alone. However, taken together with the present results for mode locking, they provide compelling evidence that the magnitude of the fundamental component of the effective periodic pinning force is independent of field and frequency at high fields and frequencies.

We note that the ratio of the maximum 1/1 step width at high frequencies to the dc threshold V_T varies somewhat from sample to sample and tends to be largest in the most coherent samples. For example, the ratio for an exceptionally coherent NbSe₃ sample at $T = 51$ K was 0.7, a factor of 2 larger than that for the sample used in paper II. The sample-to-sample variation is likely due to the effects of crystal defects other than impurities and of contacts. Defects and contact effects tend to increase the dc threshold, with the most coherent samples from a growth usually having the lowest thresholds. By creating nonuniform current distributions which smear out the steps, they tend to decrease the maximum measured step width, so that the ratio $\delta V/V_T$ is expected to decrease with decreasing sample quality.

C. Origin of the coherent oscillations

Throughout this series of papers we have assumed that the coherent oscillations (narrow-band noise) and ac-dc interference are bulk effects arising from CDW interaction with impurities and other defects. However, it has been proposed¹⁷ that narrow-band noise (NBN) might also arise from the periodic formation of vortices re-

quired for CDW-to-normal carrier conversion at the contacts. The most obvious way to differentiate between these mechanisms is to measure the sample-length dependence of the NBN amplitude: for bulk generation, the amplitude should scale as $L_s^{1/2}$, whereas for contact generation, the amplitude should be length independent. Unfortunately, such measurements are difficult, and different results have been obtained by different groups.¹⁸ The most convincing of such measurements, using non-perturbative probes, indicate bulk generation.¹⁹ More recent studies involving the application of thermal gradients²⁰ and also current injection from two sets of contacts²¹ have also supported the bulk generation hypothesis. These experiments are not conclusive, however, and some controversy has remained.

A comparison between the present ac-dc interference experiments and our earlier NBN experiments provides strong additional evidence for NBN generation in the bulk. Figure 10(a) shows the variation of the amplitude of the second harmonic of the NBN, normalized by the fundamental amplitude, versus fundamental frequency. Figure 10(b) shows the maximum ac-amplitude-dependent width of the 1/2 step at applied frequency $\omega = 2\omega_n$, normalized by that of the 1/1 step at applied

frequency $\omega = \omega_n$, versus the fundamental NBN frequency ω_n on the step. (In other words, the step width comparison is made for the same NBN fundamental frequency, or for approximately the same dc bias.) These quantities have essentially the same absolute magnitudes and both show the same slow decrease with increasing fundamental frequency. From the discussion in Sec. III B, this similarity together with that between Figs. 8 and 9 suggests that the effective periodic potentials which give rise to NBN and ac-dc interference have the same form and thus are of the same origin. Since the measured voltage width of the interference steps is proportional to the sample length, the effective potential must therefore be of bulk origin.

D. Deviations from single-coordinate behavior

Papers I and II showed that many qualitative features of the CDW response—including the ac amplitude and frequency variation of the widths of the interference steps in the dc I - V characteristic, the jumps and inductive dips in the bias-dependent ac conductance,²² and the inductive loops in phase-space plots of the response to ac fields²³—could be accounted for using a single-coordinate model of overdamped CDW motion in a nonsinusoidal periodic potential. Further, the present measurements of the detailed form of the mode-locked steps, of complete mode locking at essentially all ac amplitudes and frequencies, and of the variation of the maximum ac amplitude-dependent step width with ac frequency also are all consistent with a single-coordinate description.

However, a complete description of a pinned CDW must obviously involve many degrees of freedom that reflect random disorder. It is thus interesting to ask, what deviations from single-coordinate behavior are observed experimentally, since such deviations should provide important clues as to the correct many degree-of-freedom description. From the present measurements on samples of exceptional quality, we note the following features which seem fundamental to the response of an impurity-pinned velocity-coherent CDW and which are unlikely to be explained in terms of a single coordinate.

(1) The ratio of the maximum ac-amplitude-dependent step width to the zero-ac dc threshold voltage is smaller than single-coordinate models predict by a factor of approximately 2.

(2) The effective periodic potential deduced from the single-coordinate analysis gradually becomes more rounded as the electric field (time-averaged CDW velocity) is increased.

(3) The oscillations of the interference step widths with ac amplitude exhibit smooth minima, as in Fig. 7. Single-coordinate models predict sharp, downward pointing cusps at the minima.^{12,16,22,24} (Also see Fig. 9 in paper II.)

(4) The magnitude of the oscillations of the interference step widths decreases much more rapidly with increasing ac amplitude than is predicted, as may be seen by comparing Fig. 7 with Fig. 10 in paper II.

In an attempt to gain further insight into the fourth discrepancy, we have measured the ratio of the magni-

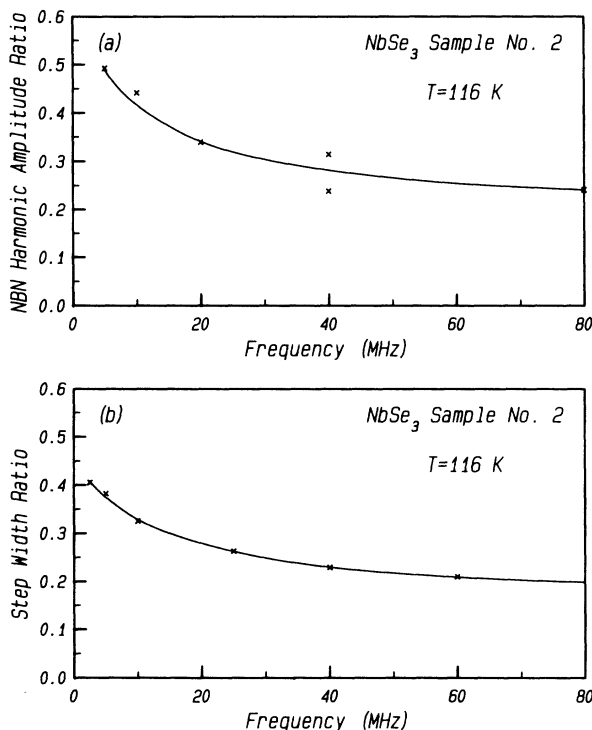


FIG. 10. (a) Ratio of the amplitudes of the second and first harmonics of the NBN, $V_2/V_{\text{fundamental}}$, vs fundamental frequency ω_n . (b) Ratio of the maximum ac-amplitude-dependent width of the 1/2 step at applied frequency $\omega = 2\omega_n$ to that of the 1/1 step at applied frequency $\omega = \omega_n$ vs the fundamental NBN frequency ω_n on the step. The solid lines are guides to the eye. The similarity between (a) and (b) suggests that the potentials which give rise to the NBN and to the interference steps have the same form and are of the same origin.

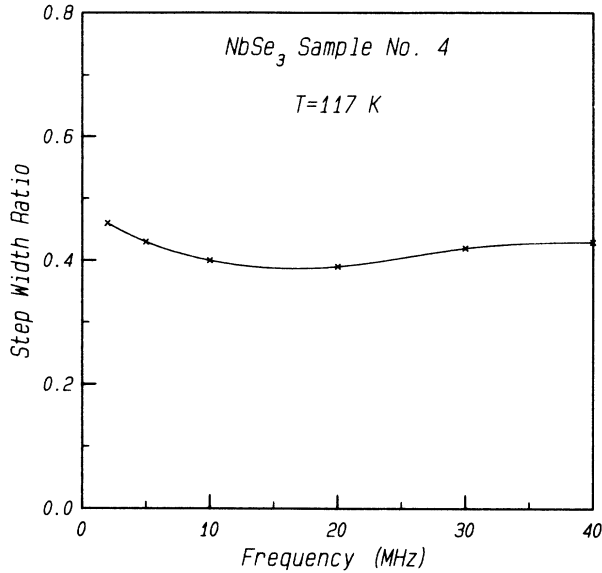


FIG. 11. Ratio of the second to the first maximum in the ac amplitude dependent width of the 1/1 step vs applied ac frequency. The solid lines are guides to the eye.

tudes of the second and first maxima of the 1/1 step-width oscillations as a function of applied ac frequency. As shown in Fig. 11, this ratio is nearly constant, indicating that the form of the oscillations with ac amplitude is independent of frequency.

The single-coordinate analysis of Ref. 12 and paper II is helpful in interpreting the significance of this result. The width of a step is determined in part by the ac-induced back-and-forth displacements of the space-averaged CDW phase, which are proportional to the integral of the time-varying current. Successive maxima of the step width correspond to those displacements (ac amplitudes) which allow the CDW to spend the most time in one part of pinning potential, and therefore to feel the largest time-averaged pinning force. For example, near the first maximum the CDW spends most of its time in one well of the potential, and then jumps to the next well. Near the second maximum, the CDW hops back one well, forward two, back one, and so on. Therefore, the data of Figs. 8 and 11 indicate that *the mode-locked width of the 1/1 step depends only upon the back-and-forth displacements of the space-averaged CDW phase, and not upon the rate at which these displacements are executed.* This is equivalent to our earlier statement that the magnitude of the fundamental component of the effective pinning potential is independent of frequency.

E. Mode locking in the frequency domain

ac-dc interference phenomena involve locking of the internal NBN frequency ω_n to the applied frequency ω . We have observed this locking directly by measuring NBN spectra in the presence of an applied ac voltage.^{25,26} Brown *et al.*²⁷ earlier studied ac voltage effects on NBN using NbSe₃ samples which did not exhibit complete mode locking. Some of the present results have been du-

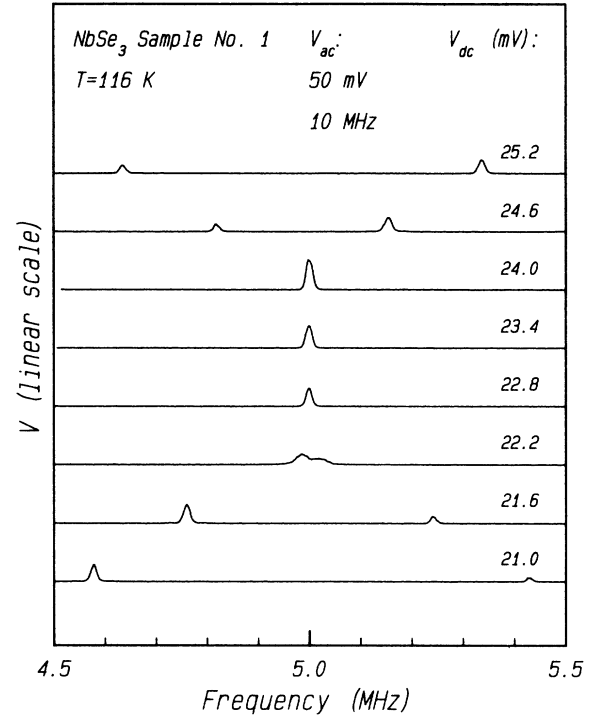


FIG. 12. Averaged NBN spectra measured in the presence of a 10-MHz, 50-mV peak ac voltage, for several dc biases near the 1/2 interference step. The larger of the two peaks at each bias is the NBN fundamental ω_n , while the second peak is the mixed signal at frequency $\omega - \omega_n$. Between $V_{dc} = 22.8$ and 24.0 mV, the NBN peak is locked to one-half the applied frequency ω .

uplicated by Bhattacharya *et al.*²⁸

Figure 12 shows NBN spectra, measured in the presence of a 10-MHz, 50-mV peak ac voltage, for several dc biases near the $p/q = 1/2$ I - V step. (Measurements of mode locking on harmonic steps are difficult because the NBN fundamental is obscured by the much larger spectral peaks due to the applied ac voltage and its harmonics.) For biases where two peaks are present, the larger of the two is the NBN fundamental ω_n , and the second peak is the mixed signal at frequency $\omega - \omega_n$. As the dc bias increases, ω_n increases and the two peaks approach one another. Then for a finite range of dc bias, corresponding to the mode-locked width of the 1/2 peak in dV/dI , a single peak is observed whose frequency is locked to $\omega_n = \omega/2$. At larger biases, the NBN unlocks, and two peaks are again observed. For biases very near those which yield mode locking, the peak positions and amplitudes fluctuate greatly. These fluctuations appear to be between the locked and unlocked states, and may be responsible in part for the large-amplitude low-frequency noise observed in dV/dI adjacent to the mode-locked steps, and for rounding near the edges of the steps.

When mode locking occurs, CDW velocity coherence is dramatically increased. Figure 13 shows the NBN fundamental at 5 MHz with no ac applied, and when mode locked to a 10-MHz, 50-mV peak ac voltage, measured using a spectrum analyzer bandwidth (100 Hz) which is much narrower than the unlocked width. When no ac is applied, the fundamental width, which reflects the width

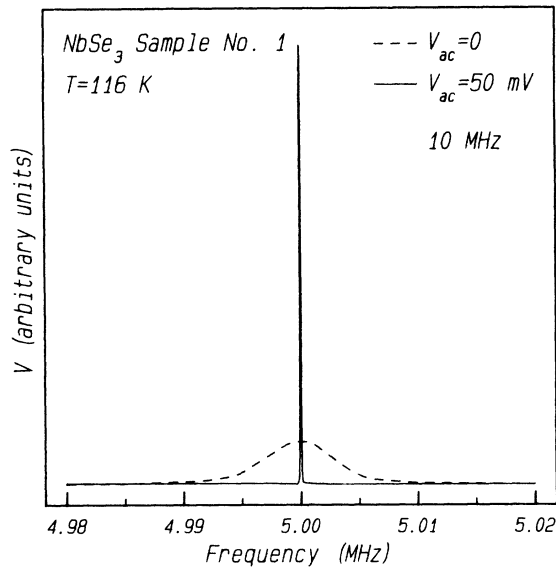


FIG. 13. Averaged spectrum of the NBN fundamental at $\omega_n/2\pi$ MHz with no ac applied and when mode locked to a 10-MHz, 50-mV peak ac voltage, measured using a spectrum-analyzer bandwidth (100 Hz) much smaller than the zero-ac fundamental width. The NBN fundamental sharpens dramatically when mode locking occurs.

of the velocity (current) distribution, is roughly 5 kHz. With ac applied but no mode locking, the spectral width is essentially unchanged. When mode locked, however, the fundamental sharpens dramatically: the spectral width becomes less than the 100-Hz minimum bandwidth of the spectrum analyzer, corresponding to a Q in excess of 50 000. This indicates that the CDW's motion has become essentially perfectly periodic, that fluctuations have been dramatically reduced, and that the CDW moves with a unique time-averaged velocity. This direct observation of the collapse of the CDW velocity distribution is consistent with previous measurements²⁹ which show that the $1/f$ -like wide-bandwidth noise (referred to as broadband noise) vanishes on a mode locked step. Furthermore, this provides strong additional evidence for our contention in paper I that the broadband noise amplitude is correlated with the width of the NBN fundamental (current distribution).

The most interesting result in Fig. 12 is that the amplitude of the NBN fundamental does not change significantly when mode locking occurs. This is illustrated more clearly in Fig. 14, where the spectrum-analyzer bandwidth is much larger than the spectral width of the fundamental, so that the peak heights reflect the total oscillating voltage within the fundamental. The averaged fundamental amplitudes when mode locked on the 1/2 step and when no ac is applied are the same to within a factor of 2. However, fluctuations in the mode-locked amplitude are much greater (at least on the time scale of the spectrum analyzer measurement). Although initial measurements seemed to indicate some variation in the mode-locked amplitude with dc bias on the step, subsequent measurements show no clear evidence for such a variation.

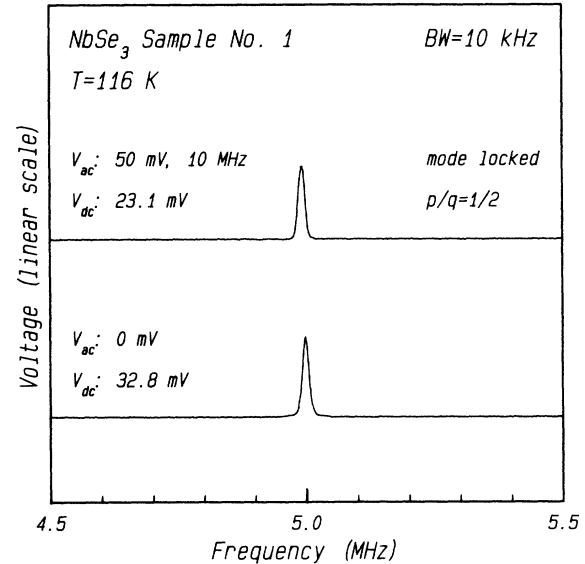


FIG. 14. Averaged spectrum of the NBN fundamental at $\omega_n/2\pi = 5$ MHz with no ac applied and when mode locked to a 10-MHz, 50-MV peak ac voltage, measured using a spectrum-analyzer bandwidth (10 kHz) much larger than the zero-ac fundamental width. The total time-averaged oscillating voltage within the fundamental is essentially unchanged when mode locking occurs, although fluctuations in the mode-locked amplitude are greater.

A possible explanation for these results, which provides insight into the relation between the dynamics of randomly pinned CDW's and the single-coordinate analysis of paper II, may be as follows. When a dc voltage is applied, the phase of the more weakly pinned regions of the crystal will advance before that of the more strongly pinned regions. The oscillating currents from each region will have different phases, so that the total oscillating current will be small (of order \sqrt{N} , where N is the number of regions) compared to the dc current. When ac and dc voltages are applied together and mode locking occurs, the single-coordinate analysis predicts that the phase of the oscillating current will lock to the phase of the applied ac voltage. One might naively expect that each region in the randomly pinned system would behave similarly, so that phase correlations would be enhanced, the CDW motion would become more rigid, and the current oscillation amplitude would become large. Experimentally, the average NBN amplitude is essentially unchanged when mode locking occurs. This suggests that mode locking involves much more subtle changes in the phase correlations, perhaps indicating that the local pinning energy dominates over the electric field energy associated with phase locking to the applied ac. Further, that subtle changes can result in a time-averaged polarization force which is comparable to the dc threshold suggests that the classical depinning field associated with rigid displacements of the CDW from the equilibrium pinned configuration must be much larger—perhaps by orders of magnitude—than the measured threshold. Differences in the correlations associated with depinning

at threshold and with mode locking might then account, in part, for the discrepancy between the predicted and measured step width to threshold ratio.

IV. DISCUSSION

Perhaps the most surprising finding of our study of ac-dc interference is how “clean” and simple the results are, since they reflect the response of a randomly pinned many-degree-of-freedom system. The quality of the mode locking in dV/dI as seen, for example, in Fig. 2, rivals that observed in Josephson junctions. Much previous experimental and theoretical work has focused on the very low-field and low-frequency CDW response, where metastability due to random impurity pinning is important. In contrast, what is most striking about ac-dc interference in high-quality NbSe₃ crystals is that this disorder seems so *unimportant*.

The success of the single-coordinate model of Ref. 12 and paper II in accounting for these results is also surprising. Indeed, nearly all important qualitative features of experiment are reproduced by this model, and deviations from its predictions seem relatively minor. This success suggests that this model's essential features—overdamped motion and a nonsinusoidal periodic effective potential whose magnitude is independent of field and frequency—must be features of any more complete model of CDW transport. It may also indicate that a reduction in the effective number of CDW degrees of freedom occurs during mode locking.

A number of calculations of ac-dc interference effects^{30–34} based on the classical elastic medium (Fukuyama-Lee-Rice) model^{2,30} and related models have been reported. These calculations have yielded anomalies in the dc I - V characteristic and in the bias-dependent ac conductance. Early calculations^{30–32} predicted neither complete mode locking nor negative “wings” in dV/dI ; instead, the peaks in dV/dI were rounded. Complete locking was subsequently predicted,³³ but only for small ac frequencies and amplitudes such that the time during which the total applied field remained below threshold was large, thereby allowing the CDW sufficient time to deform. Complete locking has recently been obtained in numerical simulations.³⁵ However, to date no calculation has yielded complete locking over the full range of ac amplitudes and frequencies for which we observe it experimentally.

Since calculations of mode locking in the classical elastic medium model are difficult, the significance of this failure to account for our results is not clear. However, our observation of a nearly frequency-independent mode-locking response, or equivalently, of nonvanishing periodic pinning, seems to be fundamentally inconsistent with this model.¹³

In this semiclassical model, the CDW is described as a charged 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, with a characteristic relaxation time which increases with the wavelength

of the deformation. When the CDW moves very quickly, highly dissipative short-wavelength deformations are excited, leading to a viscous drag force which increases with increasing velocity. However, the long-wavelength deformations which provide the dominant contribution to CDW polarization cannot relax significantly during each drift period T_d , and must vanish in the high-velocity limit. The characteristic frequency for relaxation of deformations of wavelength comparable to the Lee-Rice phase-phase correlation length L_d is the dielectric relaxation frequency, measured to be a few MHz in NbSe₃, consistent with simple theoretical estimates. Periodic CDW polarization and the periodic pinning force should vanish above this frequency, so that the mode-locking response should exhibit substantial frequency dependence. Our observation of frequency-independent mode locking and nonvanishing periodic pinning at frequencies nearly two orders of magnitude above this frequency therefore appears to be inconsistent with this classical model.

At this time, it is not clear if our results can be reconciled with the classical elastic medium model; given the model's complicated dynamics, the above argument seems a bit too simple to be completely convincing. It has been suggested,³⁶ based on the negative dip observed in dV/dI just above threshold in Figs. 3–6, that finite size effects may be important in our results. Although such dips are predicted by finite-size-effect calculations,³⁷ they could result from many mechanisms, and there is little other evidence which indicates the importance of these effects in samples of ordinary dimensions. In particular, we do not observe⁹ a predicted strong variation of the harmonic content of the NBN with sample length.³⁸ It is important to emphasize that the only distinguishing feature of the NbSe₃ samples studied here is that they are free of macroscopic defects and thus have highly uniform current distributions; their size and purity are typical of samples studied by other groups. If finite-size effects are important in these samples, they must be important in all experimental data measured to date.

It has also been suggested³⁵ that short-wavelength deformations may interact more strongly with individual impurities than long-wavelength deformations, offsetting the decrease in the magnitude of the deformations, and allowing periodic pinning to persist at high frequencies. Periodic pinning might also persist if the relevant CDW relaxation frequency is much higher³⁹ or the pinning is strong⁴⁰ and/or due to phase-slip centers. Further study is needed to determine if these ideas can explain our two crucial observations: (1) the I - V steps mode lock completely throughout the experimentally accessible range of ac amplitudes and frequencies; and (2) the harmonic mode locking response shows no applied frequency dependence and the NBN fundamental amplitude shows no fundamental frequency dependence for frequencies above 20 MHz.

One feature of our frequency-domain measurements of mode locking appears to be inconsistent with some recent calculations based on the classical elastic medium model. Fisher⁴¹ has shown that the ratio of the coherent current oscillation amplitude $\langle j_{ac}^2 \rangle$ to the dc current j_{dc} is given by

$$\frac{\langle j_{ac}^2 \rangle^{1/2}}{j_{dc}} \sim \left[\frac{\xi^d}{V} \right]^{1/2}, \quad (2)$$

where ξ is the dynamic velocity-velocity correlation length, d is the dimension of the system, and V is the volume of the sample. Matsukawa³⁴ has found that on a mode-locked step, ξ diverges, so that the current oscillation amplitude should grow dramatically. We observe no such increase in the time-averaged NBN amplitude when mode locking occurs.

It is interesting to compare our ac-dc interference experiments with those for randomly pinned superconducting vortex lattices. A strong formal analogy exists between the vortex lattice model of Schmid and Hauger⁴² and the classical elastic CDW model, which has been exploited extensively in calculations of CDW dynamics. As with CDW's, the pinning of vortex lattices is phase dependent, so that interference anomalies are observed on their flux-flow I - V characteristic when dc and ac excitations are simultaneously applied.⁴³ However, complete locking of these anomalies has not been observed,⁴³ nor has it been predicted theoretically.⁴² This discrepancy may reflect a fundamental difference between the dynamics of CDW's and vortex lattices. Alternatively, it may be due to deficiencies in both the experiment and theory of the latter. Additional experiments on randomly pinned vortex lattices might help to resolve this issue.

A detailed description of ac-dc interference effects within a quantum tunneling model⁴⁴ that includes disorder has not been given. However, the observation of non-vanishing periodic pinning at high fields and frequencies and the absence of frequency dependence in the mode-locking response would appear to be consistent with this model.⁴² In an electric field, the CDW tunnels between pinned states rather than classically deforming between them, and this allows the pinning energy to be maintained even at high frequencies. The relevant relaxation time for this process is roughly the CDW relaxation time τ which determines the limiting high-field conductance, and is on the order of 10^{-11} s. Tunneling would thus be expected to result in frequency-independent mode locking and to allow the pinning energy to be maintained throughout the experimentally accessible range of fields and frequencies, as is observed.

V. COMMENTS ON CHAOS

In a number of nonlinear systems with competing periodicities, transitions from simple periodic or quasiperiodic behavior to apparently random but deterministic behavior known as chaos are observed.⁴⁶ The question of whether CDW systems exhibit chaotic behavior has been studied experimentally. Brown *et al.*⁴⁷ reported devil's-staircase behavior in NbSe₃, and showed that a fractal dimension $D \sim 0.91$ could be calculated from the measured widths of subharmonic steps, very close to that obtained from simulations of circle maps. The significance of this result is not clear. The samples used were of ordinary quality so that the peaks in dV/dI were rounded and did not approach complete locking. Consequently, the step widths likely differed substantially from the intrinsic

widths which would be measured in a highly coherent sample, so that the uncertainty in the calculated fractal dimension must be very large. More importantly, no chaotic behavior was observed, so that there is no reason to believe that the measurements were made near criticality, where the formula used to derive the fractal dimension is valid. Hall *et al.*⁴⁸ claimed to observe chaotic behavior in NbSe₃ samples which exhibit abrupt switching from one dc current to another. Such samples usually contain macroscopic defects and often have highly inhomogeneous current distributions, resulting in complex behavior. Several ac-induced steps in the dc I - V characteristic were identified as mode-locked harmonic steps. At the edge of each step, complex NBN spectra were observed, and were interpreted as evidence for a transition to chaos via a period-doubling route. This evidence for chaotic response does not seem very convincing. The widths of the steps in the I - V characteristic were all nearly equal in size and varied little with ac amplitude, and no subharmonic steps were observed, in striking contrast to the behavior which we have observed. Further, it is unlikely that these steps were due to harmonic mode locking, because their spacing in dc bias (~ 2 mV for a sample with $V_T \sim 20$ mV) was far too small for the applied ac frequency of 15 MHz. The origin of the complex NBN spectra observed is uncertain, but given the poor quality of the samples any connection with chaos is not obvious.

From the above discussion, it seems that no convincing evidence for chaotic response in CDW systems has been found. Most CDW crystals exhibit complex, apparently random behavior (hysteresis, switching, broad-band noise), due to the effects of macroscopic defects and contacts, due to metastability inherent to a randomly pinned CDW, and due to thermal effects, which may be difficult to distinguish from chaos. Since these effects are small in the exceptionally coherent NbSe₃ samples studied here, these samples would seem well-suited for a search for chaos.

Preliminary measurements on these samples reveal no clear evidence for chaotic response. The onset of chaos is typically characterized by the appearance of wide-bandwidth noise and by hysteresis at the edge of some of the larger steps. No ac amplitude-dependent wide-bandwidth noise has been observed, although the sensitivity of the measurements may not have been sufficient for a meaningful test. However, hysteresis has been observed at temperatures below 59 K. For some ac amplitudes and frequencies the CDW current was occasionally observed to switch from being completely locked on the 1/1 step to another value, which may have been unlocked or locked on a much smaller step. This switching occurred on a time-scale of approximately 1 s, and was observed even when the dc bias was held constant. Abrupt switching of the current from zero (locked on the 0/1 step) to being completely locked on the 1/2 step was also observed, but this effect was not reproducible. Such hysteresis and switching could easily result from metastability associated with random impurity pinning (or with residual macroscopic defects) and from random (perhaps thermally induced) perturbations of the CDW. Consequently, while additional experiments are needed, CDW

materials of currently available quality do not appear well-suited to study of chaotic dynamics.

VI. CONCLUSIONS

ac-dc interference and mode locking effects in high-quality NbSe₃ crystals reveal many interesting features of the pinning and dynamics of charge-density-wave systems. Given that these effects represent the response of a macroscopically homogeneous system and result from CDW interaction with randomly distributed impurities, they seem remarkably simple and beautiful. Nearly all important qualitative features of our experiments, including the form of the mode-locked I - V steps and the variation of their widths with ac amplitude and frequency, are reproduced by a single-coordinate model of overdamped CDW motion in a nonsinusoidal periodic potential. The reason for this apparent simplicity is not obvious. Further, our observations of the apparent saturation of the magnitude of the effective pinning force at high fields and

frequencies, the absence of significant frequency dependence to the mode locking response, and the absence of any significant increase in the NBN amplitude when mode locking occurs, seem puzzling from the point of view of the classical elastic medium model, although they appear to be consistent with the quantum tunneling model. A detailed understanding of these results should yield important insights into the dynamics of charge-density waves and other many-degree-of-freedom systems.

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