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Charge-density-wave transport in quasi-one-dimensional conductors. I. Current oscillations

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We present a detailed experimental study of current oscillation phenomena in charge-density-wave (CDW) transport. The amplitude and harmonic content of the quasiperiodic component of the current oscillations remain large and approximately constant even at very high electric fields, suggesting that the form and magnitude of the impurity pinning potential are approximately independent of applied field. A simple single-coordinate model of CDW motion in a nonsinusoidal pinning potential, motivated by the quantum tunneling theory of CDW depinning, accounts for all the qualitative features of our results. Nearly perfect velocity coherence seems to be characteristic of weak-impurity-pinned CDW's in high-quality $NbSe_3$. However, other types of crystal defects together with contact effects act to broaden the observed distribution of CDW velocities. This velocity distribution plays a crucial role in the "ringing" observed in the CDW response to a current pulse, and also in generating the broadband noise.

I. INTRODUCTION

Collective charge transport by moving charge-density waves (CDW's) has been observed in a number of quasi-one-dimensional conductors, including $NbSe_3$, TaS_3, and $K_{0.3}MoO_3$.[1] For dc electric fields greater than a threshold field, the CDW "slides" through the crystal,[2,3] resulting in a highly nonlinear dc conductance at fields of millivolts per centimeter. At high fields, the dc CDW conductance saturates near the value expected in the absence of a Peierls transition. The small-signal ac conductance is also highly nonlinear at megahertz frequencies, increasing rapidly at low frequencies but saturating to the same value as the high-field dc conductance at high frequencies.[2,4] Quasiperiodic current oscillations accompany dc current flow for applied fields above threshold.[3,5] When dc and ac electric fields are applied together, striking interference phenomena similar to those seen in Josephson junctions are observed.[6] The CDW response can depend on its electrical and thermal history, resulting in a host of hysteresis and "memory" effects.[7]

All of these transport phenomena depend crucially on the phase-dependent CDW interaction with impurities, which destroys the translational invariance of incommensurate CDW's and pins their phase to the lattice. In order to account for CDW depinning and transport, two fundamentally different theoretical approaches have been proposed. In the classical approach[8–11] the CDW is treated as a charged elastic medium ("rubber sheet") which deforms around the impurities and which is subject to viscous friction. Calculations based on this model, hereafter referred to as the classical elastic medium model, have yielded a form for the dc I-V relation[9] which provides a good fit to experiment over a limited range at moderate fields,[12] qualitatively correct features of the linear ac conductance and anomalies in the low-frequency dielectric response,[13] as well as descriptions of hysteresis and other phenomena associated with the metastability of a pinned CDW. However, these calculations are difficult and have often involved simplifying assumptions whose physical relevance is uncertain. Furthermore, no detailed and quantitative comparisons between theory and experiment have been reported.

A second theoretical approach is based on treating CDW conductors as macroscopic quantum systems. In the tunneling model,[14] interaction with impurities creates a small energy gap (different from the Peierls gap) in the CDW's collective excitation spectrum. CDW depinning occurs by coherent tunneling through this pinning gap over large distances in space ($\sim 1\ \mu m$). The tunneling model has been remarkably successful in accounting for experiment. The dc I-V characteristic of $NbSe_3$ has been found to accurately follow a Zener form at all temperatures on the upper CDW transition for fields between 2 and 200 times the dc threshold.[2,12,15] A predicted scaling of ac and dc conductances is also obeyed on the upper transition in $NbSe_3$ over at least two orders of magnitude in both field and frequency.[12,14] Excellent qualitative and semiquantitative agreement has been obtained for a variety of ac mixing experiments[16] which characterize the nonlinear response of both $NbSe_3$ and TaS_3. Furthermore, values of microscopic parameters which are deduced from experiment using tunneling theory expressions have been shown to be consistent with values obtained from independent measurements.[12] However, until now a detailed understanding of current oscillation and ac-dc interference phenomena has been lacking.

With the objective of understanding more about the CDW's interaction with impurities, this paper and its companion[17] (hereafter referred to as I and II, respectively) present a detailed experimental study of the two phenomena which provide the most striking manifestations of this interaction. Paper I describes measurements of the current oscillation phenomena. We find that the amplitude and harmonic content of the quasiperiodic component of these oscillations remains large even at very high electric fields. We show that a simple single-coordinate model of CDW motion, using a nonsinusoidal effective pinning potential previously proposed within the context of the tunneling theory,[18] accounts for all qualitative features of our results. An additional important finding is that nearly perfect velocity coherence seems to be characteristic of weak-impurity-pinned CDW's in high-quality $NbSe_3$. However, other types of crystal defects together with contact effects (which are not usually included in theories of CDW transport) act to broaden the observed distribution of CDW velocities. This velocity distribution plays a crucial role in the "ringing" observed in the CDW response to current pulses, and also in generating the broadband noise which accompanies CDW conduction. Paper II describes detailed measurements of ac-dc interference phenomena. There we show that the same single-coordinate description also accounts for all qualitative and most quantitative features of these interference phenomena. Some early results of this work have been previously published.[19,20]

II. REVIEW OF CURRENT OSCILLATION PHENOMENA

The current oscillations which accompany CDW current flow at applied dc electric fields greater than threshold may be resolved into two components: coherent quasi-periodic current oscillations commonly referred to as narrow-band noise (NBN), and random current oscillations referred to as broadband noise (BBN). The Fourier spectrum of the coherent current oscillations consists of a fundamental peak, whose frequency is proportional to the dc CDW current, and a rich array of harmonics. The Fourier spectrum of the broadband noise decreases smoothly with increasing frequency f as $f^{-\alpha}$, where α is approximately 0.8.[21,22]

The origin of the coherent current oscillations has been the subject of some controversy. The CDW's energy of interaction with impurities is unchanged by a translation through one CDW wavelength, so that the impurity pinning potential is expected to be periodic. With a dc field applied, CDW motion in this potential should result in periodic current oscillations. Alternatively, current oscillations might also result from the periodic formation of vortices required for CDW-to-normal carrier conversion at the contacts.[23] Measurements have been performed which appear to support both mechanisms.[24–26] However, more recent studies involving the application of thermal gradients[27,28] and also current injection from two sets of contacts[29] seem to rule out the contacts as an important NBN source. Current oscillations are thus primarily a bulk phenomenon, associated with the CDW's interaction with impurities and other crystal defects.

Measurements of the current oscillations can yield information about the form of the impurity pinning potential. This may be appreciated by considering the rigid overdamped oscillator (ODO) model[22,30] equation of motion:

$$I_{\mathrm{CDW}}(t) \propto d\theta/dt = \omega_{\mathrm{co}}[V/V_T - \mathscr{V}'(\theta)] \ . \quad (1)$$

Here $\theta(t) = 2\pi x/\lambda_{\mathrm{CDW}}$ is the CDW phase (λ_{CDW} and x are the CDW wavelength and position with respect to the lattice), ω_{co} is the "crossover" frequency, V_T is the threshold voltage, and $\mathscr{V}'(\theta)$ represents the normalized derivative of the impurity pinning potential. This simple equation of motion has been shown to reproduce some qualitative features of CDW transport. For an arbitrary $\mathscr{V}'(\theta)$ at high electric fields $V \gg V_T$, the fundamental drift frequency $\omega_d = d\theta/dt \approx \omega_{\mathrm{co}} V/V_T$ will be nearly constant and the current oscillations are then proportional to $\mathscr{V}'[\omega_{\mathrm{co}}(V/V_T)t]$. Under these conditions, the time-variation of the current oscillations is the same as the space-variation of the pinning force. A similar relation should hold when internal degrees of freedom are included: the CDW phase and the pinning force vary in space and time, so that θ and $\mathscr{V}'(\theta)$ then represent the space average of these quantities. Spectral analysis of the current oscillations can therefore directly yield the magnitudes of the Fourier components of the effective pinning force.

III. EXPERIMENTAL METHODS AND RESULTS

Because it exhibits the most coherent response of any CDW material, $NbSe_3$ is the material of choice for a study of current oscillation phenomena. Measurements were made on several $NbSe_3$ crystals of modest purity (with a residual resistivity ratio of 130), which 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. Several different contact materials were used, including gold, silver, and platinum paste, but no differences in the measured current oscillation spectra were observed. $NbSe_3$ undergoes two CDW transitions at temperatures of 145 and 59 K, respectively. Measurements were made primarily near 115 K on the upper transition but also at 51 K on the lower transition, temperatures which were found to provide the most coherent sample response. The current oscillations could not be measured directly. Instead, the samples were biased with a dc current and the resulting voltage oscillations were measured using a Hewlett-Packard 8558B spectrum analyzer with a Trontek WF or Tektronix 1121 preamplifier. The CDW in $NbSe_3$ is shunted by a normal metallic conductance whose magnitude is approximately three times the limiting high-field CDW conductance at 115 K. The CDW is therefore approximately voltage-biased, so that the measured voltage oscillation amplitude is just the current oscillation amplitude divided by the normal shunt conductance. Because of ohmic heating at high fields, measurements were limited to dc voltages below $40V_T$ on the upper transition and below $100V_T$ on the lower transition.

Crucial to any study of CDW transport is an understanding of the role played by defects and contacts in the observed response. Real CDW materials contain many types of defects in addition to isolated isoelectronic and charged impurities. Virtually all $NbSe_3$ crystals have growth defects which result in a nonuniform cross-sectional area and current density. In material of low purity or that has been intentionally doped, impurity clustering may occur. Because of their quasi-one-dimensional nature, the mechanical coupling between crystal chains is weak; the crystals thus tend to be very fragile and subject to "fraying" along the chain direction which creates multiple independent paths for current flow. Sample preparation, mounting, and contacting procedures as well as thermal cycling can all result in structural damage to the crystal. Furthermore, because of the electrical anisotropy of these crystals, the current flow near the contacts may be very inhomogeneous. In contrast with this rather complicated experimental situation, all theories of CDW transport describe only the CDW's interaction with randomly distributed impurities, and presume CDW motion with a unique time-average velocity. Therefore, in order to obtain convincing experimental tests of theory, the effects of contacts and defects must be minimized and/or well characterized so that the "intrinsic" CDW response may be ascertained. We have expended considerable effort on developing techniques to meet this objective. Using these techniques, we have obtained with some consistency coherent current oscillation spectra exhibiting at least 15 harmonics on the upper CDW transition in $NbSe_3$ in samples up to 5 mm long. The number of harmonics which may be discerned above the background noise level provides a good measure of the sample's coherence. Figure 1 shows the NBN spectrum of a 2 mm long $NbSe_3$ sample at 45 K. The spectrum consists of a single fundamental and 23 harmonics, nearly twice as many as have previously been reported.

In measuring these current oscillation spectra, three experimental facts were found to be particularly important. First, the spectral shape of each harmonic in the current oscillation spectrum is found to be identical to that of the fundamental, and the spectral width of the nth harmonic is simply n times that of the fundamental. (In our notation, the frequency of the nth harmonic is n times the fundamental frequency.) Second, a spectrum analyzer measures the average of the absolute magnitude for the oscillating voltage within a bandwidth W about the measurement frequency. We therefore performed two different types of spectral measurements. At each of several dc biases, spectra were taken first with a bandwidth W larger than that of the highest harmonic of interest, and second, with a bandwidth much smaller than the spectral width of the fundamental. In the latter case, the relevant measure of peak height was approximated by multiplying the measured peak height of the nth harmonic by $\sqrt{n}$. In general, the results obtained using these two methods agreed fairly well; the experimental data presented here represent a composite of these two methods.

A third experimental fact of relevance to the measurements is that the amplitude of the current oscillations fluctuates in time. For relatively poor samples with broad spectral widths, the measured fluctuations in the fundamental amplitude are small. For highly coherent samples, however, the fluctuations can be very large. In one particularly coherent $NbSe_3$ sample the variance of the fundamental amplitude obtained from successive one-minute averages of the spectra was comparable to the mean amplitude. To minimize the effects of these fluctuations on the measurements, the spectra were averaged for long time periods, typically five to ten minutes.

Figure 2 shows the magnitudes of the first three harmonics of the narrow-band noise, normalized by the height of the fundamental, as a function of dc bias for a $NbSe_3$ sample at 114 K. To facilitate comparisons with theory, Table I(a) gives some of the measured parameters of this sample, and Table I(b) gives the measured funda-

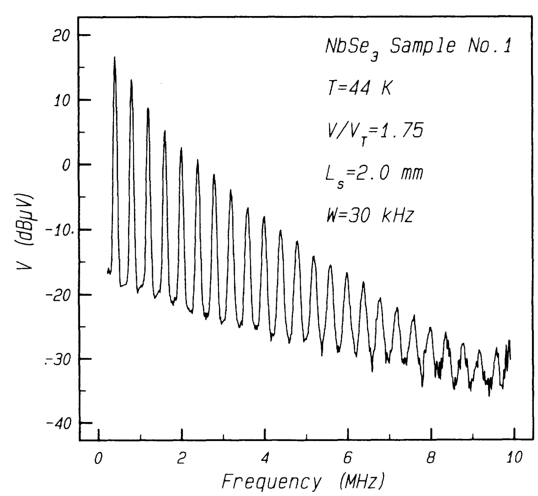

FIG. 1. Spectrum of the voltage oscillations measured in response to an applied dc current of a 2.0-mm-long $NbSe_3$ sample at 44 K. A single fundamental and at least 23 harmonics are visible.

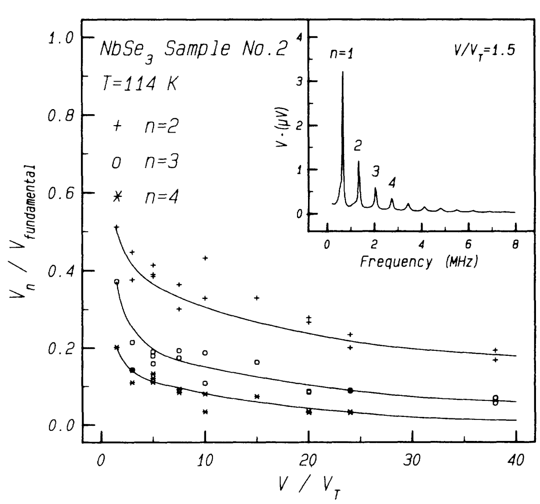

FIG. 2. Amplitudes of the first three harmonics of the current oscillation spectrum in $NbSe_3$ at 114 K normalized by that of the fundamental. Significant harmonic content remains even at very high fields. The solid lines are guides to the eye.

TABLE I. (a) Experimental parameters for $NbSe_3$ sample No. 2. The dc I-V characteristic is fit using the Zener form $I_{CDW}(V)=G_aV+G_bV\exp(-V_0/V)$. The crossover frequency ω_{co} is defined as the frequency of the peak in the imaginary part of the ac conductance. The room-temperature resistance was 87 Ω. (b) Fundamental frequency of the current oscillations at selected dc biases for $NbSe_3$ sample No. 2 at 114 K.

(a)

L_s (mm)	0.64
G_a ($m\Omega^{-1}$)	16.1
G_b ($m\Omega^{-1}$)	4.0
V_T (mV)	5.5
V_0 (mV)	17.9
$\omega_{co}/2\pi$ (MHz)	20
I_{CDW}/ν_n (μA/MHz)	5.6

(b)

V/V_T	ν_n (MHz)
1.5	0.85
3.0	5.4
5.0	11.9
10.0	29.8
20.0	66.6
38.0	130

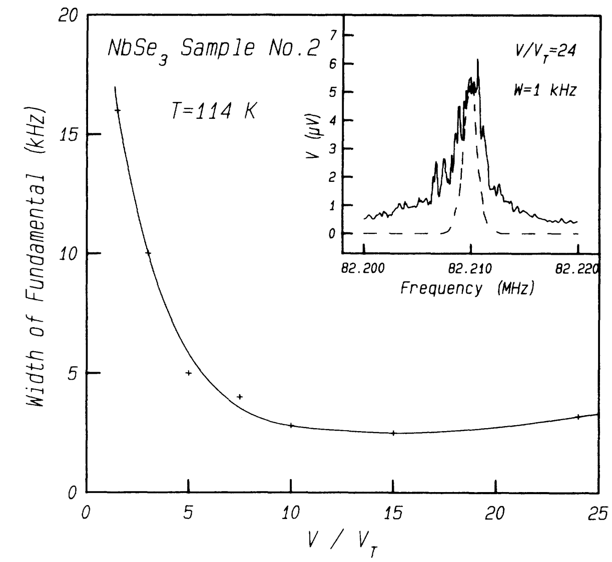

FIG. 3. Spectral width of the current oscillation fundamental vs dc bias. The inset shows the spectrum of the fundamental at 82 MHz (solid line) and of a sine wave from a signal generator (dotted line) measured using a 1-kHz spectrum analyzer bandwidth.

mental oscillation frequency at selected dc biases. Near threshold, the harmonic content is very rich, but as the dc bias increases the harmonic content at first rapidly decreases. Even at $V/V_T=38$, however, where the fundamental oscillation frequency is over six times the measured "crossover" frequency (experimentally defined as the peak in the imaginary part of the ac conductance), the magnitude of the second harmonic remains 20% of that of the fundamental. For the lower CDW at $T=51$ K in the same sample and for $V/V_T=100$ ($\nu_n=290$ MHz), the second harmonic amplitude was still 17% of the fundamental. Measurements in TaS_3 give similar results: at 170 K and for $V/V_T=25$, the second harmonic amplitude is 32% of the fundamental. These measured harmonic amplitudes all represent lower bounds: broadband noise, background and instrumental noise, and bandwidth limitations all tend to decrease the measured amplitudes of the harmonics relative to the fundamental. Extrapolating these results, we expect that the harmonic content will remain large even at frequencies of the order of the pinning frequency, which is thought to be approximately 1 GHz for our $NbSe_3$ samples. The fact that the second harmonic does not vanish at high electric fields was previously discussed by Weger *et al.*[31] in terms of a relaxation oscillator model.

The fundamental for the sample of Fig. 2 at 114 K consisted of two extremely sharp, closely spaced peaks, one approximately seven times larger than the other. Figure 3 shows the measured spectral width of the larger of these two peaks versus dc bias. The spectral width is seen to decrease rapidly near threshold but saturates to a constant value at moderate fields. Because of nonuniform heat sinking along the length of the crystal, thermal gradients arising from ohmic heating broaden the fundamental at very high fields. The response of this sample was exceptionally coherent: at a drift frequency of 84 MHz, the spectral width of the fundamental was less than 3 kHz, corresponding to a $Q\equiv\omega/\delta\omega\approx 30\,000$. As we discuss below, the observed spectral width depends upon both contact effects and crystal defects. In samples of ordinary quality, the spectral width of the fundamental appears much broader and tends to broaden further with increasing field. However, the individual peaks which comprise the fundamental are observed to narrow. We therefore believe that the behavior shown in Fig. 3 represents the ideal "intrinsic" response of the CDW in $NbSe_3$.

The measured spectral width of the current oscillations also depends strongly upon temperature. As shown in Fig. 4 for the upper transition in $NbSe_3$, the spectral width of the fundamental is rather large near the transition, decreases to a minimum around 120 K, and then increases rapidly at lower temperatures. This sample was not of exceptional quality; however, we observe similar although less pronounced broadening even in our best samples. The general behavior shown in Fig. 4 is also observed on the lower transition in $NbSe_3$ and in TaS_3 (Ref. 32) as well. This behavior likely results from a competition between two factors. First, as the temperature decreases the CDW amplitude grows, so that establishing regions with different average CDW velocities becomes energetically less favorable. On the other hand, the process of depinning at crystal defects and some other strong pinning centers is probably thermally activated. Recent measurements by Gill[33] indicate that this is true of the phase-slip which occurs at the contacts. At high temperatures, therefore, phase-slip may occur easily, so that the velocity broadening near the transition is likely associated with the vanishing CDW amplitude. At low temperatures, phase

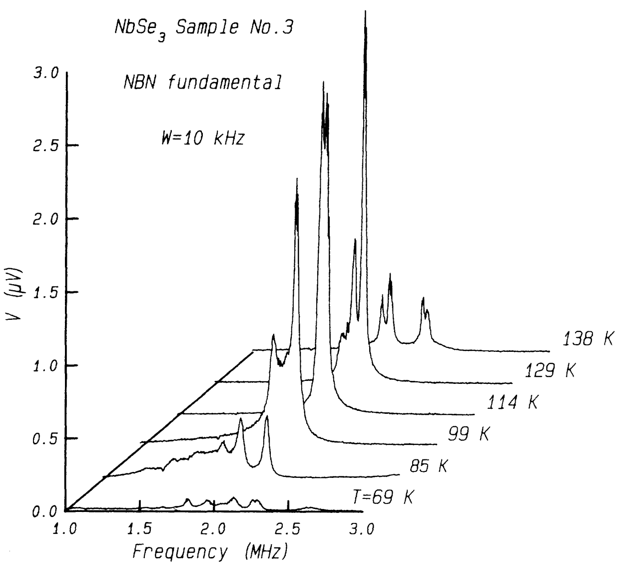

FIG. 4. Temperature dependence of the shape of the current oscillation fundamental. The fundamental broadens significantly at low temperatures and at temperatures near the CDW transition.

slip may become more difficult, resulting in substantial nonuniformities in the current distribution.

One of the most striking results of our measurements is that the magnitude of the current oscillations remains large even at very high fields. As shown in Fig. 5 for the $NbSe_3$ sample of Fig. 2, the total oscillating voltage within the fundamental, measured using a spectrum analyzer bandwidth W much larger than the spectral width of the fundamental (upper curve), increases rapidly near threshold but saturates toward a constant value at high fields. This measured fundamental amplitude depends upon dephasing effects (to be discussed later), but the constant spectral width shown in Fig. 3 suggests that these effects become bias independent at high electric fields. In fact, the spectral width of the fundamental at $V/V_T=38$ was significantly broadened due to heating effects, yet the oscillating amplitude appeared undiminished. From the discussion of Sec. II, the experimental results illustrated in Figs. 2 through 5 therefore indicate that the CDW moves in a highly nonsinusoidal potential whose magnitude and shape are approximately independent of the applied electric field.

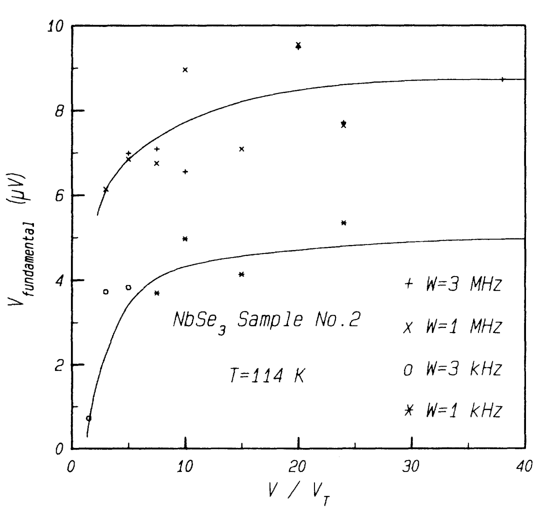

FIG. 5. Magnitude of the voltage oscillation fundamental measured using four different spectrum analyzer bandwidths W. The upper curve represents the fundamental voltage measured with W much larger than its spectral width. The results using $W=3$ MHz and $W=1$ kHz only differ by a factor of 2, indicating the exceptional coherence of this sample.

IV. ANALYSIS AND DISCUSSION

A. Coherent current oscillations

Several models have been proposed to account for current oscillation phenomena. The simplest classical model is the rigid overdamped oscillator model of Eq. (1) with a sinusoidal pinning potential.[22,30] This model has numerous deficiencies. First, it provides a poor fit to the observed dc I-V characteristic, rising much too rapidly near threshold. Second, it predicts that the harmonic content of the current oscillations should be negligible at fields greater than approximately three times threshold. Third, the model is unphysical because the time-average pinning energy is always zero. Fourth, measured values of the model parameters do not even approximately satisfy the predicted relations. One way to see this last inconsistently is to use the measured values of ω_{co}, the limiting high-field conductance G_b, and the ratio of the CDW current to the measured fundamental oscillation frequency I_{CDW}/ω_n in the ODO model relation

$$V_T=(\omega_{co}/G_b)/(I_{CDW}/\omega_n) . \quad (2)$$

As shown in Table II(a), the calculated threshold voltage is then found to be 5 and 8 times larger in $NbSe_3$ and TaS_3, respectively, than is measured. A similar disagreement results in Table II(b) when values of the pinning frequency ω_p and the CDW effective mass m^* inferred from millimeter-wave conductivity measurements[34] are used in the ODO model relation

$$E_T=m^*\omega_p^2\lambda_{CDW}/2\pi e . \quad (3)$$

The ODO model thus does not provide an adequate parameterization of experiment.

Weger *et al.* proposed a relaxation oscillator model for the current oscillations.[31,35] This model does give large harmonic content at high fields, but is otherwise very similar to the ODO model and has most of its other deficiencies. A predicted sawtoothlike waveform for the current oscillations was shown to be consistent with measured spectra near threshold, where the harmonic amplitudes fall off with increasing order n as $1/n$ or $1/n^2$. However, direct measurements of the current waveform near threshold show that it more closely resembles periodic, widely spaced spikes. Knowledge of only the magnitudes of the Fourier components provides insufficient information to uniquely reconstruct the current waveform

TABLE II. Experimental parameters are obtained from (a) dc and rf measurements and (b) millimeter wave measurements (from Ref. 36). The threshold fields calculated using these parameters in the ODO model expressions Eqs. (2) and (3) are much larger than are measured in $NbSe_3$, TaS_3, and $(TaSe_4)_2I$.

(a)

	G_b ($m\Omega^{-1}$)	$\omega_{co}/2\pi$ (MHz)	I_{CDW}/ω_n (μA/MHz)	V_T (expt.) (mV)	V_T [Eq. (2)] (mV)
$NbSe_3$ 114 K	4.0	20	4.9	5.4	24.5
TaS_3 185 K	2.3	70	16.1	63	480

(b)

	m^*/m_e	$\omega_p/2\pi$ (MHz)	E_T (mV/cm)	E_T (mV/cm)
$NbSe_3$ 45 K	117	2600	30	400
TaS_3 160 K	940	4300	150	8700
$(TaSe_4)_2I$ 150 K	10^4	34 000	$\approx$1500	5.8×10^6

(and thus the pinning force).

More sophisticated classical treatments are based on the classical elastic medium model. Because the calculations are difficult, few predictions for the form of the current oscillations have thus far been made. Fisher[11] has shown that near threshold the CDW velocity within a correlation volume should exhibit harmonics up to some cutoff frequency Ω, above which the amplitude of the harmonics will rapidly decrease. The ratio of Ω to the fundamental oscillation frequency is predicted to vary with dc bias as $(V/V_T-1)^{\mu-\zeta}$, where $\mu<\zeta$; i.e., the harmonic content should decrease with increasing dc bias. Experimentally, no sharp cutoff in the harmonic content is observed: the harmonic amplitudes decrease smoothly with increasing order n at all fields. Further, the values of μ and ζ have only been calculated using a mean field theory; the results do not appear to provide a good quantitative account of experiment, nor have we found any obvious way to deduce these parameters from our measurements. A quantitative comparison thus does not appear possible at this time. Using a different approach which includes finite size effects, Klemm and Schrieffer[10] have predicted that in very short samples the pinning potential should be sinusoidal. However, as the sample length L_s increases the potential is predicted to become more structured, with the ratio of the second harmonic of the potential to the fundamental increasing as $L_s^{1/2}$. Figure 6 shows the results of measurements of this ratio made on a single sample of $NbSe_3$ at 121 K. Measurements on very short samples (<0.15 mm) are difficult because inhomogeneous current injection and damage caused by the contracts result in spectra that are very broad and also in uncertainties in the effective sample length. Although there is some scatter in the data, it is clear that the harmonic amplitude in $NbSe_3$ has no length dependence for sample lengths between 0.25 and 4 mm. Contacts could conceivably act as strong pinning centers in short samples and thus introduce additional harmonic content. However, the fact that the harmonic content does not vary with sample length argues against this. These results may indicate either that the theory is not appropriate, or that finite size effects are not important in $NbSe_3$ samples of the lengths and purities which are usually studied.

In the classical elastic medium model, deformations of the moving CDW around pinning centers should result in a nonsinusoidal space-averaged pinning force. However, these models also predict that such deformations, and therefore the pinning potential, should become smaller with increasing dc field and vanish in the high field limit. Experimentally, the amplitude of the current oscillations is approximately constant and the magnitude of the second harmonic varies by less than a factor of two for fields ranging over one-and-one-half orders of magnitude. Further, the harmonic content of the current oscillations is still large at fields of 40 to 100 times threshold, where the dc CDW conductance is within a few percent of its

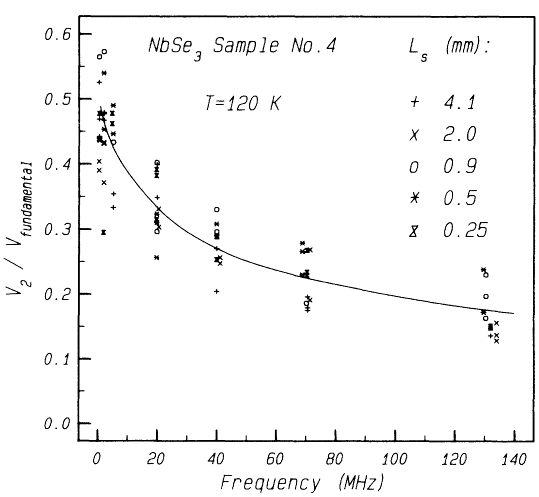

FIG. 6. Amplitude of the second harmonic of the voltage oscillations normalized by that of the fundamental vs fundamental frequency, for several different sample lengths L_s. The measurements were performed on a single crystal of $NbSe_3$ using three different spectrum analyzer bandwidths. No dependence on sample length is observed.

limiting high-field value. In the absence of explicit calculations, it is not clear how these results can be compatible with vanishing deformations and pinning.

Current oscillations could also result from vortex formation required for CDW-to-normal carrier conversion at the contacts or at "cracks" in the crystal.[23] As previously mentioned, recent experiments by several groups[27–29] appear to rule this out as the primary NBN generation mechanism. Further evidence against the vortex hypothesis is provided by the almost perfectly coherent spectra which are observed, as in Fig. 3. Within this model the creation of a vortex modulates the sample current and voltage, and the rate of vortex formation will be proportional to the current discontinuity between any two velocity-coherent regions. Our contacts are very large compared to the sample width and thickness, and are often made only to one side of the crystal. Because of electrical anisotropy, this must result in a highly inhomogenous current distribution near the contacts. That vortex formation could occur sufficiently regularly under such conditions to yield voltage oscillations with Q's of $\sim 30\,000$ seems improbable.

An alternative description of current oscillation phenomena has been proposed within the context of the tunneling theory.[18] The CDW charge density may be written as

$$\rho(x,t)=\rho_0+\rho_1\cos[2k_Fx+\phi(x,t)]\ , \tag{4}$$

where $2k_Fx$ is the phase of a uniform static CDW and $\phi(x,t)$ describes the variations in the CDW electron density (CDW compressions and expansions) which give rise to pinning. Using an idealized weak-pinning model in which the pinning is periodic, two equivalent ground states are hypothesized, $\phi_A(x)$ and $\phi_B(x)$, that differ by π in average phase, and represent opposite signs for the charge variation. This is illustrated in Fig. 7(a) where the length L_d corresponds to the Lee-Rice phase-coherence length.

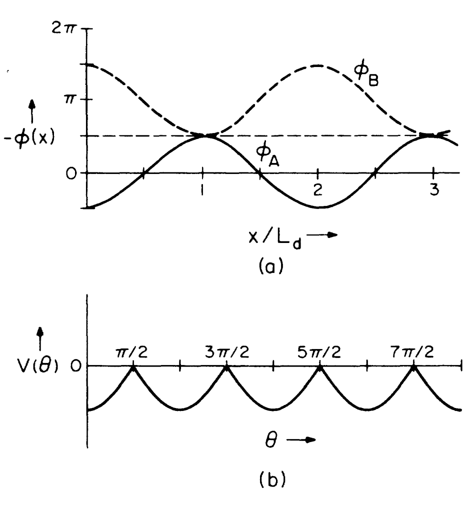

FIG. 7. (a) Phase variations $\phi_A(x)$ and $\phi_B(x)$ that minimize the impurity pinning energy. (b) Impurity pinning potential $\mathscr{V}(\theta)$ given in Eq. (5).

The phase is assumed to have the optimum values required to minimize the pinning energy at $x=0$ and for integral multiples of L_d; both ground states have the same phase (mod 2π) at these points. When a current flows, the space average of the phase decreases in time as $-\omega_d t$. For $-\pi/2<\omega_d t<\pi/2$ (mod 2π), ϕ_A has the lowest energy, while for $\pi/2<\omega_d t<3\pi/2$ (mod 2π), ϕ_B has the lowest energy. To maintain a net negative pinning energy, the system alternates between the ϕ_A and ϕ_B states. This alternation, which corresponds to the periodic deformations of a depinned CDW in the classical elastic medium model, results in an effective pinning potential of the general form

$$\mathscr{V}(\theta)\propto\begin{cases}-\cos\theta \ \text{ for } -\pi/2<\theta<\pi/2 \ (\text{mod}\, 2\pi)\ , & \text{(5a)}\\ \cos\theta \ \text{ for } \pi/2<\theta<3\pi/2 \ (\text{mod}\, 2\pi)\ . & \text{(5b)}\end{cases}$$

The periodicity of the pinning in this case is π in phase, so that $\omega_n=2\omega_d$. The Fourier components of this model pinning potential are found to be given by

$$a_q=(4/\pi)(-1)^{q+1}/(4q^2-1)\ . \tag{6}$$

The cusps in $\mathscr{V}(\theta)$, illustrated in Fig. 7(b), thus introduce a rich array of harmonics into $\mathscr{V}'(\theta)$ whose amplitudes decrease as $1/q$ for large q. In real CDW systems, the pinning is not precisely periodic in space, so that the actual potential is expected to be more rounded. A nonsinusoidal potential similar to Eq. (5) has been proposed by Zawadowski *et al.*[36] to describe interaction of the CDW with a single impurity; such a potential may also result from polarization at strong pinning centers.[37] Coherent Zener tunneling of CDW electrons drives the motion within the pinning potential according to the tunneling theory. The alternation between two equivalent ground states allows a net pinning energy to be maintained for drift frequencies well above the pinning frequency. The harmonic content and amplitude of the current oscillations are thus expected to persist undiminished at high fields.

A complete equation of motion which allows a detailed calculation of current oscillations within the tunneling theory has not been derived. We therefore use a simple model which we believe contains most of the important physics. Acceleration of CDW electrons is determined in this model not by the applied electric field, but by the difference between this force and the pinning force $\mathscr{V}'(\theta)$. The CDW current is thus approximately written as

$$I_{\text{CDW}}(t)=G_b V_{\text{eff}}\exp(-V_0/V_{\text{eff}})\ . \tag{7}$$

Here $V_{\text{eff}}(t)=V(t)-V_M\mathscr{V}'(\theta)$ and $I_{\text{CDW}}/A=n_c(\lambda_{\text{CDW}}/2\pi)d\theta/dt$, where V_M is the maximum magnitude of the pinning force, $\max|\mathscr{V}'(\theta)|=1$, and n_c is the density of CDW electrons. In solving this first-order differential equation for the space-average phase θ, some value of V_M must be assumed. As will be discussed in paper II, measurements of ac-dc interference phenomena indicate that the appropriate magnitude in $NbSe_3$ is approximately one-third of the measured dc threshold field. With this assumption, Eq. (7) yields fits to the measured dc I-V characteristic in $NbSe_3$ which are generally excellent, and indeed somewhat better than previous Zener fits at fields near threshold.

Figure 8 shows the harmonic content of the current oscillations calculated using Eq. (7) with $\mathscr{V}'(\theta)$ as given by Eq. (5) and with $V_M = V_T/3$. The parameters V_0, V_T, and G_b were obtained from fits to the dc I-V characteristic in $NbSe_3$. The predictions are seen to be in reasonable qualitative agreement with the experimental results of Fig. 2. However, the predicted harmonic content saturates more quickly to a constant value, and at high fields remains somewhat larger than is measured. This seems to indicate that the effective pinning potential is more rounded than in Eq. (5), and that it may even have some slight dependence on applied electric field. Figure 9 shows the predicted amplitude of the current oscillation fundamental as a function of dc bias. The oscillation amplitude increases rapidly near threshold but saturates at high fields, similar to the behavior observed in Fig. 5.

Additional evidence for nonvanishing pinning at high electric fields may be provided by measurements of the dc CDW conductance. Fleming *et al.*[38] have shown that in both $(TaSe_4)_2I$ and $K_{0.3}MoO_3$ the high-field CDW conductance is temperature-activated and limited to approximately the value of the normal ohmic conductance over a very broad temperature range. Dissipation due to normal electron screening of CDW charge fluctuations is thought to reduce the CDW conductance in this region.[37] This apparent limiting of the dc CDW conductance by the normal screening indicates that CDW charge fluctuations, and therefore the pinning by impurities, remains significant even at very high fields, as predicted by the tunneling theory. In the classical elastic medium model the charge fluctuations and pinning vanish at high fields, so that the limiting high-field CDW conductance is predicted[39] to remain independent of the normal electron conductance, contrary to experiment.

B. Voltage "ringing" in response to current pulses

Oscillations in the CDW response may be observed directly in pulse "ringing" experiments. When a pulse of current greater than the threshold value is applied to a CDW crystal, the voltage exhibits an oscillatory component with a frequency appropriate to the steady-state CDW current.[40] The waveform usually resembles a damped sine wave, although in highly coherent $NbSe_3$ samples near threshold the waveform tends to resemble a damped full-wave-rectified sine wave. The initial amplitude of the ringing is found to be orders of magnitude larger than the final steady-state oscillation amplitude (usually measured using a spectrum analyzer). The simplest and most obvious explanation for the observed decay in the amplitude of these oscillations is that of CDW dephasing. Before the pulse, all regions of the CDW are relaxed in metastable configurations corresponding to local minima of the pinning potential. When the current pulse is first applied, these different regions begin moving together in phase, so that the total oscillating voltage is, roughly speaking, a simple linear sum of the oscillating voltages in each region. Eventually, however, the correlations are lost, and the oscillating voltages in the various regions dephase. In the steady state, the oscillation amplitude should then be roughly $1/\sqrt{N}$ of its value at the start of the pulse, where N is the number of phase coherent regions within the sample. Experimentally, the steady-state oscillation amplitude in $NbSe_3$ is found to be approximately three orders of magnitude smaller than the initial ringing amplitude, consistent with estimates of order 10^6 for the number of phase-coherent domains within a sample.

The time rate of decay of the oscillations depends upon the distribution of velocities within the crystal, which should also be reflected in the spectral width of the NBN fundamental. We indeed find that the number of cycles required for the ringing to decay is approximately equal to the Q of the fundamental. Figure 10 shows the

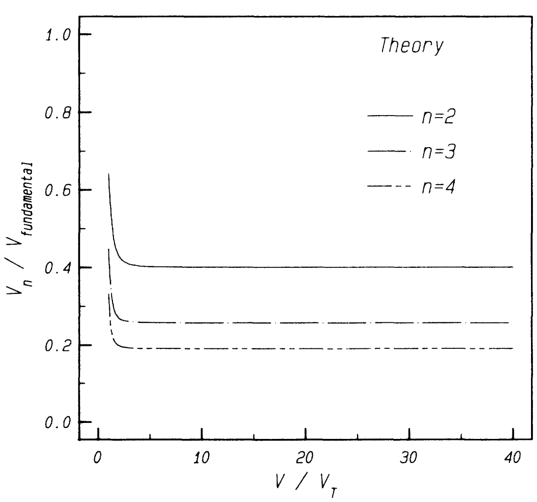

FIG. 8. Amplitudes of the first three harmonics of the current oscillation spectrum normalized by that of the fundamental, calculated using Eq. (7) with $\mathscr{V}(\theta)$ as given in Eq. (5).

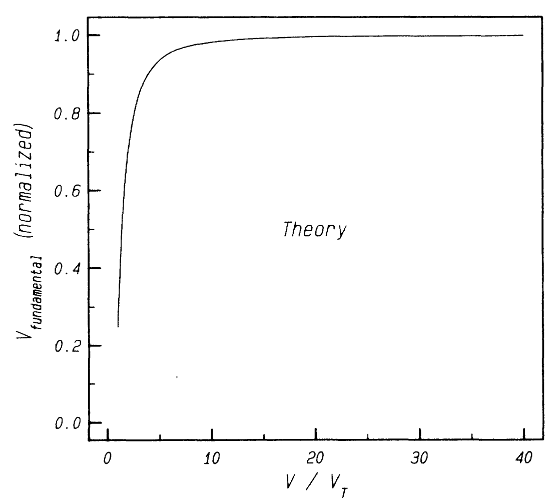

FIG. 9. Amplitude of the fundamental vs dc bias, calculated using Eq. (7) with $\mathscr{V}(\theta)$ as given in Eq. (5).

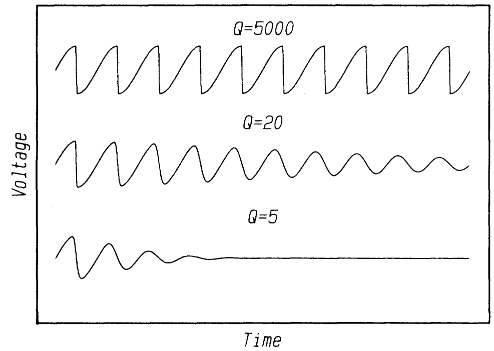

FIG. 10. Effect of dephasing on the CDW voltage response to a current pulse. A Gaussian distribution of velocities is assumed, with a width corresponding to three different values of $Q=\omega/\delta\omega$. The underlying form of the voltage oscillations is taken to be the derivative of the pinning potential of Eq. (5).

predicted pulse-response voltage waveform, assuming a Gaussian distribution of CDW velocities and that the generated voltage in each domain has the same form as the pinning force of Eq. (5) (as predicted at large dc bias). Typical $NbSe_3$ samples exhibit Q's of 20, while Q's in TaS_3 and $K_{0.3}MoO_3$ are usually less than 5. Note that even though the underlying voltage waveform is highly nonsinusoidal, the predicted response for the lower Q's resembles a damped sinusoid. Any additional phase shifts between different regions of the crystal associated with how the current-carrying state is established will lead to further rounding of the waveform. The results in Fig. 10 for $Q=5$ may be seen to closely resemble the observed ringing behavior in $K_{0.3}MoO_3$ near threshold.[41] Clearly, it would be desirable to observe the ringing waveform at high fields, since there it should have precisely the form of the pinning force. There are several practical difficulties associated with this, however. First, the oscillation period at sufficiently high fields is at most tens of nanoseconds, which is short compared to both the R-C time constants in our measurements and to the decay time for "ringing" on most pulse generator outputs. Second, the oscillation amplitude at such fields is on the order of 0.1% of the pulse amplitude. Third, even in very coherent samples there is no guarantee that dephasing effects would not completely obscure the underlying waveform. As a result, we have thus far been unable to perform convincing measurements of the ringing at high electric fields.[42]

Dephasing effects have been discussed previously,[43] although their physical origin has not really been described. More recently, it has been suggested[44] that the dephasing picture is consistent with predictions[45] based on the classical elastic medium model. In this model, the CDW moves with a *unique* time-average velocity (no "tearing" of the "rubber sheet" is allowed); the apparent distribution of velocities is due to time-dependent fluctuations associated with the CDW's interaction with impurities. Our experiments, however, clearly indicate that the velocity distributions responsible for CDW dephasing in typical crystals are not described by this model. The distribution of time-average CDW velocities within a crystal[46] is observed to be associated with crystal defects (perhaps including some types of impurities) and contact effects. With appropriate experimental technique, these effects due to defects and contacts are found to be greatly reduced. Figure 11 shows the voltage response to a current pulse for a 2.8-mm-long sample of $NbSe_3$ at 121 K. Each 10 μs segment represents an average of 64 traces, each with an oscillation amplitude comparable to the averaged amplitude. The seven averaged segments were acquired successively over a period of 20 min. The voltage oscillations persist with little decay for over 350 cycles; the measured Q of the narrow-band noise fundamental at the steady-state oscillation frequency was 700. Even though these results indicate significantly greater coherence than has previously been reported, we believe that they are still limited by residual crystal defects and contact effects. Intrinsic ringing decay times associated solely with velocity-coherent CDW motion in the presence of impurities must be at least thousands of cycles of the oscillations. We believe that observed ringing decay times in TaS_3 and $K_{0.3}MoO_3$ are much shorter than in $NbSe_3$, because both are more anisotropic, so that inhomogeneities in the current distributions are greater, and both have shorter transverse coherence lengths. Furthermore, TaS_3 is much more fibrous and therefore more susceptible to growth defects and damage induced by handling, and $K_{0.3}MoO_3$ tends to be of lower crystalline quality as evidenced by hysteresis and other metastable phenomena which are much larger than in other materials. With careful experimental technique, TaS_3 samples can be obtained with relatively few defects; we have observed ring-

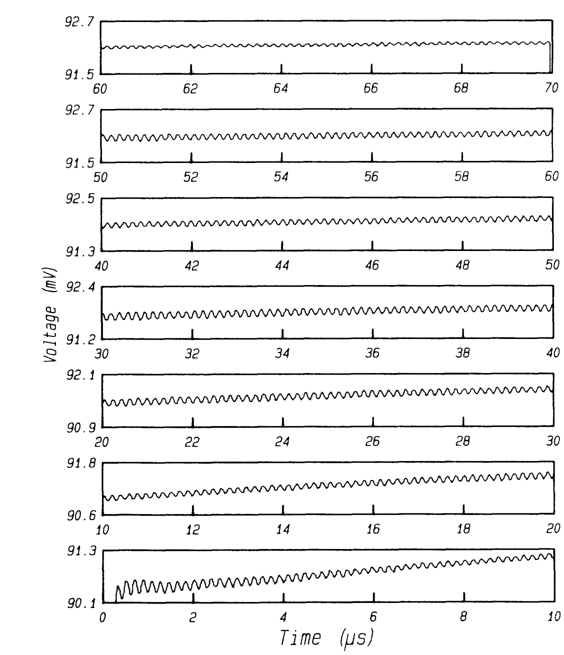

FIG. 11. Voltage response to a current pulse of a 2.8-mm $NbSe_3$ sample at 121 K. The voltage waveform is an average of 64 successive traces. The voltage oscillations persist with little decay for over 350 cycles.

ing waveforms in TaS_3 which persist for tens of cycles, comparable to those more typically observed in $NbSe_3$.

We note one further interesting feature of the ringing: the initial voltage oscillation amplitude is considerably smaller than one would predict from the measured dc threshold field. For example, in the ODO model of Eq. (1), the amplitude of the voltage oscillations is predicted to be $V_{ac}=2G_b V_T/(G_a+G_b)$, where G_a is the shunting normal electron conductance and G_b is the limiting high-field CDW conductance. For the sample of Fig. 11, V_{ac} is calculated to be 6.5 mV, whereas the measured (single-shot) oscillation amplitude is only 0.3 to 0.7 mV. A similar calculation based on Eq. (7) yields an oscillation amplitude near threshold which is roughly a factor of two smaller than the ODO result. One possible explanation for this discrepancy is that dephasing effects (e.g., a distribution of initial phases) may be important even at the start of the pulse. A second possibility is that the amplitude of the periodic component of the pinning force felt by the sliding CDW may be significantly smaller than the dc threshold field. As we shall discuss in paper II, measurements of interference phenomena in $NbSe_3$ suggest that $V_M \approx V_T/3$, i.e., the amplitude of the periodic component is roughly one-third the measured dc threshold field. Use of this pinning force magnitude in the model of Eq. (7) yields voltage oscillation amplitudes within a factor of 2 or 3 of the measured amplitudes in highly coherent samples.

C. Broadband noise

In addition to the coherent quasiperiodic current oscillations, all CDW materials exhibit random current oscillations or broadband noise (BBN) for applied fields above threshold. Previous experimental studies[5,21,47,48] have established that: (i) the power spectrum of the BBN has an $f^{-\alpha}$ frequency dependence with α near 1 and is nearly field independent except near V_T; and (ii) the rms BBN voltage δV scales as $(L_s/A)^{1/2}$ where L_s and A are the sample length and cross-sectional area, respectively, indicating that BBN is generated throughout the volume of the sample. The origin of the BBN has generally been accepted to be phase fluctuations associated with the interaction of a deformable CDW with random impurities. A threshold-field fluctuation model[48] has accounted for some features of the BBN; and a predicted relation between the BBN amplitude and dR/dV, where R is the sample resistance and V is the dc voltage, has been verified experimentally. Using this model, a dynamic coherence length ξ_D has been derived. Measurements of BBN in TaS_3 have been interpreted[49] to show that this length follows a scaling behavior near threshold, i.e., $\xi_D \propto (E-E_T)^{-\nu}$, supporting Fisher's contention[11] that CDW depinning is a dynamical critical phenomenon.

Our experimental results indicate that broadband noise is associated primarily with the wide distribution of time-average CDW velocities that are present in most crystals. As previously discussed, this distribution is due to crystal defects and to contact effects, which together tend to fragment the crystal into different current-carrying regions. We find that for crystals from a given growth tube with comparable threshold fields and resistances, those exhibiting NBN spectra with the broadest spectral widths also exhibit the largest BBN amplitudes. Conversely, the most coherent samples, such as the $NbSe_3$ samples 1, 2, and 5 discussed here, exhibit almost no BBN except at dc biases very near threshold. The increase in BBN observed with decreasing temperature is consistent with this viewpoint since, as illustrated in Fig. 4, the NBN broadens significantly at low temperatures.[50] The vanishing of the BBN observed[51] when complete CDW mode-locking to an applied ac electric field occurs is also consistent with this viewpoint, since complete mode-locking implies that the CDW moves with a unique time-average velocity throughout the entire crystal volume. (Phase fluctuations arising from the CDW's interaction with impurities are still present, however.)

To illustrate this point further, we have studied the effects of intentionally induced damage on current oscillation spectra in $NbSe_3$. Measurements were performed first on an undamaged crystal. The crystal was then covered with a commercially available heat-sinking compound; at low temperatures the compound freezes and the resulting stress damages the crystal. The low-field two-probe sample resistance at the measurement temperature was not changed by this procedure, and no BBN could be measured below threshold. As shown in Fig. 12, stress-induced damage fragments the crystal into regions with different velocities, resulting in substantial broadening of the NBN spectrum. Figure 13 shows the BBN spectrum measured below 1 MHz when the NBN fundamental was at 40 MHz; the large separation of the NBN fundamental frequency from the BBN measurement frequency ensured that only "true" BBN was measured. The sample before damage was not of high quality, having a spectral width at 40 MHz of approximately 3 MHz; it thus exhibited substantial BBN. In spite of this, the broadband noise power was doubled as a result of damage. It is interesting

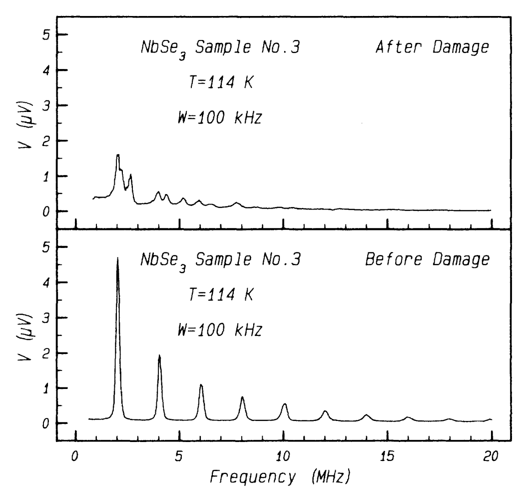

FIG. 12. Current oscillation spectrum of a 1.0-mm $NbSe_3$ sample at 114 K. Stress-induced damage results in significant broadening of the CDW velocity distribution within the crystal, as indicated by the broadening of the spectral peaks.

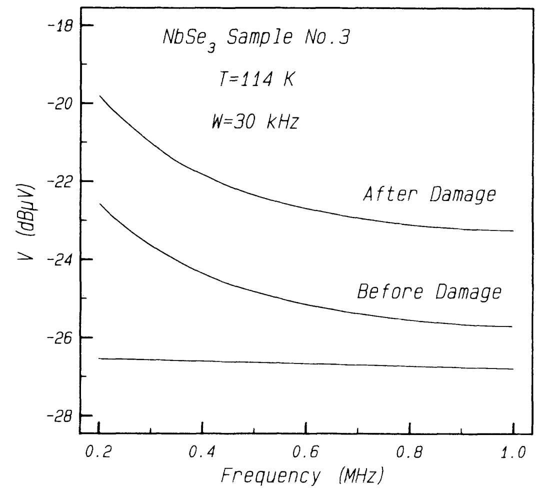

FIG. 13. Broadband noise spectrum of the $NbSe_3$ sample of Fig. 12, where the NBN fundamental frequency is 40 MHz. Stress-induced damage results in a substantial increase in the BBN amplitude. The bottom trace indicates the noise level measured for applied dc fields below threshold.

to note that crystal damage smears out the threshold transition and increases the hysteresis near threshold. Conversely, the most coherent samples from a given growth also tend to have the lowest threshold fields and show virtually no hysteresis or broadband noise. We believe that the defects responsible for the velocity distribution must play an important role in determining the threshold field and in phenomena associated with CDW metastability. We also believe that the present interpretation is consistent with the threshold-fluctuation model of Ref. 48.

In measuring broadband noise, it is crucial to separate those broad spectral components which are actually a sum of coherent oscillations in different regions with different time-average CDW velocities from truly random, incoherent oscillations. We believe that broadening of the NBN due to damage and due to an applied thermal gradient was responsible for the apparent observation[25,52] of nonlinear CDW conduction without NBN: the broad NBN spectrum could not be distinguished from BBN. We have subsequently determined that NBN always accompanies CDW conduction.[28]

Although broadband noise is clearly associated with a distribution of velocities, the underlying mechanism by which the BBN is generated is uncertain. Fluctuations likely occur at the boundary between regions with different time-average velocities, perhaps arising from the phase-slip process. The observed $1/f$ spectrum may thus represent the distribution of beat frequencies between regions with different velocities. CDW metastability associated with pinning by both weak and strong impurities, as well as by the "fragmenting" defects is also likely to play an important role. However, it is now certain that the observed BBN cannot be explained by any previous treatment of the classical elastic medium model.[53] Previous calculations do not include processes in which the phase difference between spatially separated points diverges in time. Our results clearly indicate that such processes are in fact crucial to BBN generation. In our highly coherent $NbSe_3$ samples, where they are nearly absent, the BBN amplitude is very small. In the same samples at temperatures where they are less coherent, in coherent samples which have been structurally damaged so that they become less coherent, and in samples which are initially less coherent, the BBN amplitude is large and roughly proportional to the width of the velocity distribution. We also note that TaS_3 crystals, which were used in the study of Ref. 49, generally contain a broad distribution of velocities, and therefore exhibit broad NBN spectra. As a result, it is essentially impossible to separate coherent oscillations from random noise at biases near threshold. Together, these facts make it unlikely that measurements of BBN in TaS_3 will yield information about any scaling behavior which may exist near the CDW depinning transition.

V. CONCLUSION

Measurements of current oscillations provide a useful probe of the pinning in charge-density wave systems. Although most theories of CDW transport describe only velocity-coherent CDW motion in the presence of individual impurities, crystal defects and contact effects result in nonuniform current and velocity distributions which play an important and sometimes crucial role in the observed phenomena. From our measurements, we infer the following behavior for an "ideal," weak-impurity-pinned CDW: The CDW executes overdamped motion in a periodic pinning potential whose magnitude is approximately independent of field and whose shape is highly nonsinusoidal but may have some (small) field dependence. This motion is characterized by nearly perfect velocity (but not phase) coherence in samples of arbitrary size, and by the generation of little or no broadband noise. Our results are generally consistent with a form for the pinning potential previously proposed within the context of the quantum tunneling theory. On the other hand, the apparent saturation of the shape and magnitude of the pinning potential at high electric fields and drift velocities appears to be inconsistent with the classical elastic medium description of CDW dynamics. A complete quantitative account of our findings is generally lacking, however, and further theoretical efforts are needed.

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