Harmonic Generation and Scaling Behavior in Sliding-Charge-Density-Wave Conductors

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The generation of harmonics of a small ac modulation field is used, in conjunction with the standard current-voltage measurement, to study the depinning behavior is well-characterized and high-purity samples of the charge-density-wave conductor $NbSe_3$. The results show that the current carried by the charge density wave obeys a scaling law: $I_{CDW}=[(V-V_T)/V_T]^\zeta$ with $\zeta=1.23\pm0.07$. Implications of this result on the various theoretical models of the dynamics of the sliding state are discussed.

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The onset of sliding of the incommensurate charge-density-wave (CDW) condensate[1] in inferred from the appearance of nonlinearity in the current-voltage characteristics beyond a threshold voltage V_T. Below V_T, which is a measure of the pinning produced by random impurities, the Ohmic current is carried by the normal electrons which constitute the uncondensed electrons in the case of an incompletely gapped system such as $NbSe_3$, or the electrons activated over the single-particle gap in the semiconductor, such as TaS_3. Above V_T, the CDW conducts as a parallel channel, resulting in the *I-V* characteristics given by

$$I=V/R_N+I_{CDW}(V,V_T)\,, \tag{1}$$

where R_N is the Ohmic part of the resistance due to the normal electrons and I_{CDW} is zero for $V<V_T$. This general description is further confirmed by the observation of a linear relationship between I_{CDW} and the narrow-band-noise (NBN) frequency ω_0. This identified ω_0 as the washboard frequency: $\omega_0=\mathbf{v}\cdot\mathbf{q}_0$, where $\mathbf{v}$ and $\mathbf{q}_0$ are, respectively, the velocity and the wave vector of the CDW. Although the general phenomenology is well established, the exact form of $I_{CDW}(V,V_T)$ for $V>V_T$ in Eq. (1) remains unsettled. This is a crucial issue, since it is the primary observation that led to the discovery of the sliding conductivity, which is, other than superconductivity, the only known collective electronic transport mechanism. A careful evaluation of this behavior provides a testing ground for the various theoretical scenarios of the sliding conduction.[2] Moreover, other systems, e.g., flux-lattice dynamics in type-II superconductors and two-fluid interface in random porous media,[3] are thought to be generic to this class of collective transport in disordered systems.[4]

In this paper we report results of an experiment that uses the generation of harmonics of a low-frequency and small-amplitude modulation signal to probe the nature of nonlinearity associated with the sliding conduction and determines the form of $I_{CDW}(V,V_T)$ near threshold. The results show that the current carried by the CDW obeys a scaling law:

$$I_{CDW}=[(V-V_T)/V_T]^\zeta\,, \tag{2}$$

with $\zeta=1.23\pm0.07$. This behavior appears to be consistent with the theoretical picture proposed by Fisher[5] that treats the depinning phenomena as a cooperative dynamical critical phenomenon involving many degrees of freedom of deformable CDW.

The experiments were performed on high-purity $NbSe_3$ crystals.[6] These samples were made from high-purity Nb with very low Ta content, generally thought to be the primary pinning center. The samples have a high value of residual resistivity ratio, $R\sim250$–300, and very small threshold field, $E_T\sim1$ mV/cm at 42 K. The dc *I-V* characteristics were determined in a four-probe configuration. The samples were characterized by different techniques to ensure the absence of filamentary conduction as described below. These samples also did not show any measurable switching or hysteretic behavior that is sometimes observed in some samples under poorly controlled conditions. The harmonic-generation experiment was performed by adding a small (6 μA) ac current source from an Hewlett-Packard model HP3562A dynamic signal analyzer at typically 100 Hz. The voltage output, also in the four-probe configuration, was amplified and the harmonic content was then analyzed by the same analyzer. The net driving current is given by $I=I_{dc}+I_{ac}\cos(\omega t)$, and the resulting voltage across the sample can then be obtained by a standard Taylor expansion in I_{ac}:

$$V=V_{dc}+(dV/dI)I_{ac}\cos(\omega t+\phi_1)+\tfrac{1}{2}(d^2V/dI^2)I_{ac}^2\cos^2(\omega t+\phi_2)+\cdots\,. \tag{3}$$

The Fourier transform of this signal, measured in the rms mode, consists of δ functions at $\omega_n=n\omega_0$ with strengths A_n given by

$$\begin{aligned} A_0&=V_{dc}+\tfrac{1}{4}(d^2V/dI^2)I_{ac}^2+\cdots\,,\\ A_1&=(dV/dI)I_{ac}+\tfrac{1}{6}(d^3V/dI^3)I_{ac}^3+\cdots\,,\\ A_2&=\tfrac{1}{4}(d^2V/dI^2)I_{ac}^2+\cdots\,,\\ A_3&=\tfrac{1}{18}(d^3V/dI^3)I_{ac}^3+\cdots\,. \end{aligned} \tag{4}$$

We note that the phase shifts ϕ_n in Eq. (3) are essentially zero and thus the absolute measurements are sufficient. Note that for every even (odd) harmonic, all higher-order even (odd) harmonics can, in principle, contribute. Therefore, the driving amplitude is kept so small that the higher harmonics do not produce any significant contributions. For $\omega\ll\omega_0$, as is the case here, the various derivatives of the I-V curves in Eqs. (3) and (4) are essentially their dc values. We have also measured the ac response of the system at various dc biases to ensure that there is no frequency dependence in the ac response in the frequency regime of interest, even for the smallest dc bias above threshold.

Figure 1 shows the bias dependence of the fundamental and the first two harmonics. The amplitudes of the higher harmonics decrease rapidly as shown by the scales for each; this assures negligible contamination caused by the even higher harmonics. A_1 is essentially the differential resistance as is commonly measured for the CDW systems. However, the second and the third harmonics have not been reported before. Obviously, the higher harmonics represent the higher derivatives of the I-V curve. From these data one can easily obtain the quantities dI/dV ($=1/A_1$) and d^2I/dV^2 ($=-A_2/A_1^3$) through Eq. (4). Even without a detailed analysis we obtain considerable information from the qualitative behavior of the harmonics.

Note that, $dI/dV=1/R_N+dI_{\mathrm{CDW}}/dV$ and $d^2I/dV^2=d^2I_{\mathrm{CDW}}/dV^2$. Therefore, these results show that the first derivative goes down continuously to the conductance of the normal electrons, while the second derivative and all higher derivatives of I_{CDW} diverge at V_T as it is approached from above (barring a small rounded region very close to it). It also shows that the higher derivatives of the I-V curve decrease faster as V increases beyond V_T. These results suggest that if there is a scaling behavior, i.e., as in Eq. (2), then the exponent obeys the condition $1<\zeta<2$. Now we examine the scaling behavior in greater detail.

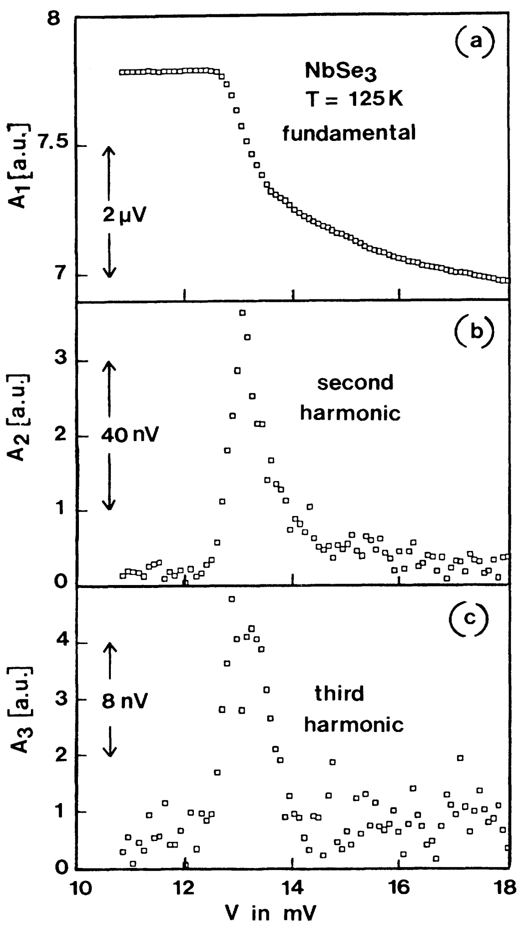

FIG. 1. Bias dependence of the fundamental and the two higher harmonics. Note that the two higher harmonics become very large at threshold. See the text for discussions.

Figure 2 shows the scaling behavior of I_{CDW} obtained from the dc I-V curve below the upper transition in the sample. The fit yields an excellent power-law form as in Eq. (2) with $\zeta=1.24\pm0.03$ in the range of reduced voltage ϵ $\{=(V-V_T)/V_T\}$ of $10^{-2}<\epsilon<1$; we cannot fit the data closer to V_T due to the rounding effects, as we shall describe below. Note that the information about the higher derivatives can also be obtained from the dc I-V curves by numerical differentiation. Figure 3 shows the apparent scaling behavior of the quantity dI_{CDW}/dV from (a) the dc I-V curve and (b) the strength of the fundamental in the harmonic-generation experiment. The exponents obtained from these are in excellent agreement with each other. Clearly, the latter yields considerably cleaner data. In the former, successive differentiations accumulate the errors and no quantitative estimates about the higher derivatives can be obtained from the I-V curve beyond the fact that the second derivative diverges at V_T. The harmonics, on the other hand, are much more precise and the scaling behavior of the higher derivatives can indeed be obtained from them. Figure 4 shows the scaling behavior of the second derivative (d^2I/dV^2) obtained from the harmonics; this quantity diverges as the threshold is approached from above.

In all these fits, the value of the critical exponent depends crucially on the choice of V_T. In order to circumvent this ambiguity, we can plot the quantities dI_{CDW}/dV and d^2I_{CDW}/dV^2 versus each other. This plot, shown in

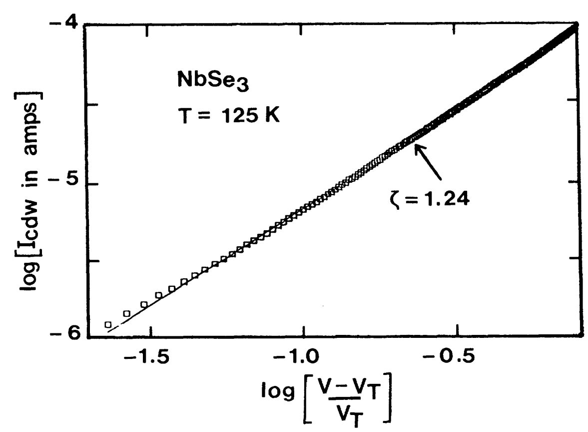

FIG. 2. Scaling plot of I_{CDW} vs reduced field obtained from the dc I-V curves below the upper transition in $NbSe_3$. The quoted value of the exponent is obtained from the straight-line fit shown in the figure.

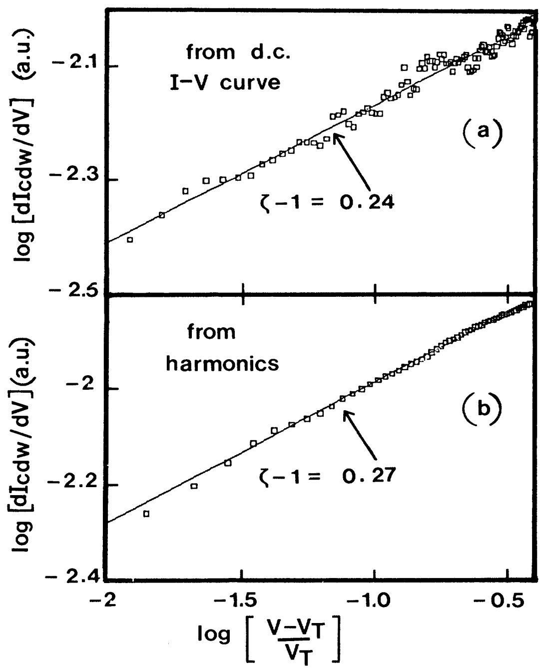

FIG. 3. Scaling plot of dI_{CDW}/dV from (a) the dc I-V curve and (b) from the harmonic at $T=125$ K. The exponent is $\zeta-1$. The quoted exponents refer to the straight lines shown in the figures.

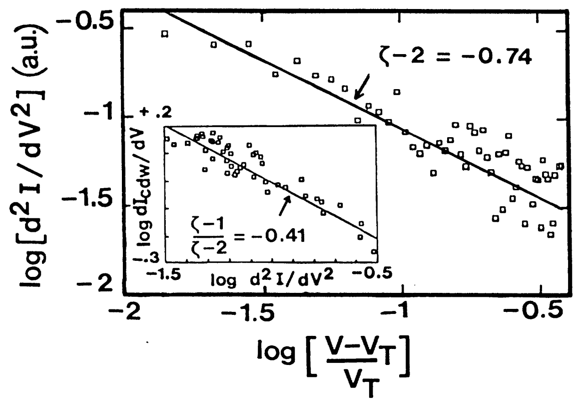

FIG. 4. Scaling plot of d^2I/dV^2 vs reduced field at $T=125$ K; the exponent is $\zeta-2$. Inset: A scaling plot of dI_{CDW}/dV vs d^2I/dV^2; the exponent is $(\zeta-1)/(\zeta-2)$. Both quantities are evaluated from the strength of the harmonics. The exponents refer to the straight lines shown in the figures.

the inset of Fig. 4, yields a value of $(\zeta-1)/(\zeta-2)$ equal to -0.41 ± 0.10, consistent with the value of ζ evaluated above. Although the error in this fit is considerably greater, it eliminates the need for a precise evaluation of V_T and provides unambiguous bounds on the exponent.

We have also made measurements such as reported above in several samples in both the upper and the lower CDW states in this material, between 120 and 135 K for the upper state and between 40 and 48 K for the lower state, i.e., the temperature range over which typical measurements are made in this system. At the lower CDW state, rounding due to a finite modulation amplitude could not be completely eliminated due to the smallness of V_T. Nevertheless, we find no evidence of any systematic T dependence of the scaling exponent over the range mentioned above. Combining all these data we arrive at the value of $\zeta=1.23\pm0.07$ for this material. The error referred to is not that of a single fit but of the overall variation at different temperatures; it also includes the ambiguities of locating the threshold, particularly below the lower CDW transition.

Now we compare the experimental results with the predictions of the various theoretical models. The classical single-particle model[7] predicts the following form for the CDW current near threshold:

$$I_{\mathrm{CDW}}=[(V-V_T)/V_T]^{1/2}. \tag{5}$$

It is obvious that the experimental results are qualitatively inconsistent with this prediction since it implies that the first derivative of the CDW current diverges at threshold. Since our results are obtained in very pure and well-characterized samples, we believe that the failure of this model is not related to problems of sample quality, but rather a real inadequacy of the single-particle approach to the CDW dynamics, as has been suggested previously.[2]

The quantum tunneling model[8] has also been used to describe the field dependence of I_{CDW} at large dc biases, although it has not been tested adequately near threshold. This model predicts

$$I_{\mathrm{CDW}}=(V-V_T)\exp(-V_0/V). \tag{6}$$

In other words, a discontinuity in the first derivative is expected at threshold, in addition to the exponent being unity, neither of which is observed in any of our samples. Although an incorrect value of the exponent can be obtained by choosing V_T incorrectly, the harmonics show clearly whether or not the exponent is consistent with observed behavior. We conclude that, at least near threshold, the quantum tunneling model too is inconsistent with the data.

The deformable CDW model with many internal degrees of freedom has been analyzed by Fisher[5] using a mean-field model. This model predicts a scaling form as in Eq. (2) with a mean-field exponent $\zeta=\frac{3}{2}$. No prediction is available for spatial dimensionality $d=3$. However, we note that the measured exponent is within the bounds provided by this model. An exponent around unity has been found[9] theoretically for one-dimensional models. At this time, the theoretical expectations for $d=3$ lies between 1 and $\frac{3}{2}$, assuming (a) monotonicity of the exponent with increasing d and (b) that the (yet unknown) upper critical dimension is greater than 3. Therefore, we conclude that the measured exponent is not inconsistent with the deformable CDW model.

Recently, a different classical model has been proposed by Tucker, Lyons, and Gammie[10] that assumes that the pinning is strong near the impurity site. Far away from the impurity, the average CDW phase is correlated as in the standard weak-pinning scenario. At this time, no predictions are available for this model on the behavior of I_{CDW} near threshold. It would be important to know to what extent the behavior near threshold is modified in this picture over that given by the previous classical deformable model.

We now discuss some experimental problems. We have investigated the origin of the small rounding near V_T seen in Fig. 1. We have determined that this rounding is about 5 times larger than what would be expected for the finite modulation amplitude. A finite-size effect is expected to cause a rounding when ξ_v, the velocity-velocity correlation length in the classical deformable-medium model,[5] approaches the sample dimensions. We do not observe a single-particle behavior expected in this regime[11] and thus rule out this explanation too. Another possibility is that there is some thermal rounding at finite temperatures, as in, e.g., Josephson tunneling.[12] If this is the origin of rounding, it has important implications for other CDW materials such as in o-TaS_3 where the rounding is much more pronounced.[13] In all cases, we believe the peak in the second derivative of the *I-V* curve is a good measure of the location of V_T. I_{CDW} due to rounding in $NbSe_3$ is more than 2 orders of magnitude smaller than I_{CDW} at $V=2V_T$. Thus it does not produce any significant error in our evaluation of the exponent so long as the data above the rounded region are used in the analysis. A second, and more difficult, experimental problem is to eliminate the effects of filamentary conduction, where the sample consists of several distinct segments and the current distribution is highly nonuniform. We find that no reliable analysis can be made in samples where it occurs. Several methods have been employed to characterize the sample to ensure that no filamentary conduction occurs in the sample.[14]

To conclude, we have obtained a precise experimental determination of the scaling behavior associated with the conduction via the CDW condensate in high-purity samples of $NbSe_3$ which show no detectable filamentary conduction. Because of the aforementioned rounding effects, one tends to overestimate V_T and thus underestimate ζ. Therefore, the smaller values are, in general, more reliable. However, there are indeed samples that show different qualitative behaviors. In some samples, dV/dI has a downward wing at threshold[15] that cannot be described by the form reported here. Samples that show switching or hysteretic behavior[16] are also exceptions. More work is needed to understand these classes of samples, the conditions under which they occur, and whether or not they represent a different qualitative situation. We have also found evidence of a rounding effect caused by yet unknown sources. $NbSe_3$ shows by far the smallest rounding effects. This can be a serious problem with other CDW systems. It is likely that the apparent variation of the critical exponent in previous studies is caused by including the data from the rounded region.[2] The methods described here would be useful in determining if the depinning behavior is all CDW systems can be described by one universal critical exponent. It also remains to be seen to what extent this theoretical scenario[4] describes similar transport phenomena in other disordered systems, as mentioned above.

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[14]The presence of filamentary conduction is checked (1) by the occurrence of multiple thresholds in the *I-V* curves and multiple NBN fundamentals, (2) by the presence of secondary maxima in the second-harmonic amplitude, and (3) most reliably by the appearance of interference features in a model-locking experiment, when $\omega_0/\omega_{ext}=p/q$, p and q being integers [see S. Bhattacharya *et al.*, Phys. Rev. B **38**, 7177 (1988)] that *cannot* be indexed by one set of p/q values.

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