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Molecular-dynamical studies of the depinning of charge-density waves

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It is shown that a pure, classical insulating many-body system will exhibit nonlinear threshold behavior of the form $(E - E_T)^{1/2}$, where E is the applied and E_T is the threshold electric field. Evidence is presented, based on molecular dynamics calculations on a model for sliding charge-density waves in an impure lattice, that the current versus field curve will become concave upward, but will still have a well defined threshold field in the thermodynamic limit in qualitative agreement with experiment. It is speculated that the low-frequency noise found by Fleming and Grimes is due to the depinning and motion of a hierarchy of soliton lattices of large lattice constant which appear in any incommensurate system.

I. INTRODUCTION

$NbSe_3$, which develops one charge-density wave below 144 K and a second below 59 K, has the unusual feature that it exhibits nonlinear conduction at fields as low as 1 eV/cm. This is believed to be due to the sliding of previously pinned charge-density waves.[1-6] Recent experiments by Fleming and Grimes indicate that the onset of nonlinear conduction occurs at a well defined threshold electric field and that the current near threshold can be fitted with

$$J = \sigma E + (E - E_T)\sigma_b e^{-E_0/(E-E_T)}, \tag{1}$$

where E_T is the threshold field, E is the field, and E_0, and σ_a, and σ_b are parameters.[6] This is consistent with Lee and Rice's theory of depinning of charge-density waves by weak electric fields.[7] Lee and Rice gave estimates for "breakaway" field. In previous work, Fukuyama and Lee,[8] Koehler and Lee,[9] and Weisz, Sokoloff, and Sacco[10] have discussed the linear conductivity of a charge-density wave in the presence of impurities. In this article, we are concerned with the nonlinear conduction due to the depinning of the charge-density wave by a dc electric field. In a previous article Sacco *et al.*[11] discussed the damping of a charge-density wave moving in an impure system and found the damping force to be a linear function of the drift velocity in three dimensions (i.e., a Drude form). This result, which should be applicable for larger fields at which the charge-density wave becomes completely free, would imply a current contribution due to the sliding charge-density which is linear in the applied electric field. In the present article, the intermediate nonlinear behavior, which occurs as the wave begins to break away, will be treated. Since threshold behavior at $T = 0$ for pure or single-particle systems[12,13,14] is usually of the form $(E - E_T)^{1/2}$, it is not easy to understand why in this system there should be a current versus E curve which is concave upwards near threshold. Maki[15] and Bardeen[16] independently proposed theories based on quantum-mechanical tunneling of solitons which give an $e^{-E_0/E}$ dependence of the nonlinear part of the conduction. In view of the data of Fleming and Grimes,[6] which gives a well defined threshold field at which nonlinear conduction begins, in qualitative agreement with the ideas of Lee and Rice,[7] a simple classical model of impurity depinning will be treated in this article.

In Sec. II, the threshold behavior of a pure, pinned system will be discussed. In Sec. III, molecular dynamical studies on a simple model for charge-density waves in an impure system will be presented. In Sec. IV, conclusions and speculations on the origin of the noise spectrum observed by Fleming and Grimes will be presented.

II. THRESHOLD BEHAVIOR FOR INFINITE PURE AND FINITE IMPURE SYSTEMS

It is well known that both a single particle in a sinusoidal potential and a chain of atoms connected by springs, situated in a sinusoidal potential of the same period, exhibit threshold behavior of the form $(E - E_T)^{1/2}$ in the overdamped limit at zero temperature.[12-14] At nonzero temperatures there is no longer a well defined threshold.[12-14] In the chain system this is because of the creation of soliton-antisoliton pairs at any nonzero temperature, which can conduct electric current. Of course, since for two- and three-dimensional systems solitonlike defects can extend across the crystal, it may not be possible for them to be thermally activated,[17] and hence, well defined threshold behavior may persist at nonzero temperature.

It will now be shown that the square-root thresh-

old behavior is much more general. Consider a classical pinned many-body system such as the present model which begins to conduct after a critical field E_T is reached. For $E<E_T$ the system will be in equilibrium in the applied electric field, but when E becomes equal to or greater than E_T, the equilibrium configuration becomes unstable. Thus, the system in the applied field must posess an unstable phonon mode for $E \geq E_T$. This phonon represents the intital motion of the system as it begins to slide in the applied field (i.e., the sliding mode). When E is slightly greater than E_T, the systems will spend a long time very close to the equilibrium configuration for E slightly below E_T. The reason for this is that for such values of E, the pinning forces almost cancel the effects of E. For this reason, we will consider small displacements (δu_j) of the atoms of the system from their equilibrium configuration for E just below E_T. We will solve the resulting equation of motion for the time required for the system to move far from this equilibrium state, at which point our small displacement assumption is no longer valid. We expect, however, that this "depinning time" will not depend significantly on the details of the motion when such large displacements are attained because the system will spend very little of its time at such large displacements. Once the system has broken loose it will at a later time become almost pinned again and the break-away process repeats itself. This is similar to "stick slip" friction. Here j labels both the particles and components of δu. The equation of motion to lowest order in the δu's is

$$m\delta\ddot{u}_j+\gamma\delta\dot{u}_j=-\sum_i C_{ji}\delta u_i+\sum_{lm} B_{jlm}\delta u_l\delta u_m+\cdots+\delta E, \tag{2}$$

where the C's and B's are constants, $\delta E=E-E_T$, and γ is the damping constant (e. $g.$, damping due to the effects discussed in Ref. 11). We will consider the extreme overdamped limit for simplicity. This same approximation will be made in the molecular dynamical calculations to follow. In doing so we are neglecting possible oscillatory motion of the system which may explain the noise spectrum in $NbSe_3$,[6] but the overall nonlinear conduction should still be described correctly. In this limit, the first term on the lefthand side of Eq. (2) is neglected compared to the second. Transforming to the normal coordinates of the system for the equilibrium configuration at $E=E_T$,

$$\delta u_j=\sum_0 U_{j\alpha}q_\alpha, \tag{3}$$

we get

$$\gamma\dot{q}_\alpha=\omega_\alpha^2 q_\alpha+\sum_{\alpha'\alpha''}\sum_{jlm} U_j^{\alpha*}U_l^{\alpha'}U_m^{\alpha''}B_{jlm}q_{\alpha'}q_{\alpha''}+\delta E', \tag{4}$$

where

$$\delta E'=\delta E\sum_j U_j^{\alpha*}.$$

By our assumption, one of the frequencies ω_α will be zero. Since for $|E-E_T|\ll E_T$ the system spends a long time near the $E=E_T$ equilibrium configuration, we may replace the stable modes q_α by their equilibrium values

$$q_\alpha=\delta E'/\omega_\alpha^2.$$

The anharmonic terms are neglected here because they are much smaller than the harmonic terms which are nonzero for these modes. Then if we eliminate these modes with the above relation, the equation of motion for q_{α_0}, where α_0 is the unstable mode, becomes

$$\gamma\dot{q}_{\alpha_0}=Fq_{\alpha_0}^2+D\delta E'q_{\alpha_0}+\delta E'. \tag{5}$$

Assuming that for small δE the conducting system spends most of its time stuck at points where it is almost pinned, the time t_c for the system to break loose is given by

$$t_c=\gamma\int_{-\infty}^{\infty}\frac{dq_{\alpha 0}}{Fq_{\alpha_0}^2+D\delta E'q_{\alpha_0}+\delta E'}\sim\frac{1}{\sqrt{\delta E}}. \tag{6}$$

Thus, the current $\sim 1/t_c\sim\sqrt{\delta E}$.[18] The above argument is correct for a finite system where there is a finite frequency spacing between the modes and will also be correct for an infinite, pure, charge-density-wave system which is pinned by commensurability. In a commensurate pure system, there exists a finite number of normal modes for each value of the wave vector (i.e., a finite number of bands). Since a uniform electric field only couples to the zero-wave-vector modes, the equation of motion reduces to Eq.(4) with the α's referring to the band states at zero wave vector. The above arguments follow and again give a square-root threshold behavior. For an infinite impure system, on the other hand, it is quite likely that there will exist many nearly-zero-frequency modes, and hence, our assumption that all modes except one are in equilibrium is probably not valid. The fact that the experiments give no such square-root critical behavior—in fact the current versus E curve is actually concave upwards instead of downwards[6]—may be an indication of the breakdown of the above theory.

III. NUMERICAL WORK ON IMPURE SYSTEMS

In order to study electric field depinning in impure charge-density-wave systems we have studied the impure discrete sine-Gordon chain or Frenkel-Kontorova[19] model via molecular dynamics. The equations of motion of our model may be written as

$$m\ddot{x}_j+\gamma\dot{x}_j=-\alpha(2x_j-x_{j+1}-x_{j-1})-\frac{2\pi}{a}V_0C_j\sin\frac{2\pi}{a}x_j+E \tag{7a}$$

for $1<j<N$ and

$$m\ddot{x}_1+\gamma\dot{x}_1=-\alpha(x_1-x_2+b)-\frac{2\pi}{a}V_0C_1\sin\frac{2\pi}{a}x_1+E\,, \tag{7b}$$

$$m\ddot{x}_N+\gamma\dot{x}_N=-\alpha(x_N-x_{N-1}-b)-\frac{2\pi}{a}V_0C_N\sin\frac{2\pi}{a}x_N+E\,, \tag{7c}$$

where x_j is the position of the jth atom, b and a are the lattice constants of chain and sinusoidal potential, respectively, α is the force constant of the interatomic bonds, γ is the damping constant, m the mass, E the applied field, and C_j =1 on a site containing an impurity and 0 otherwise. The locations of the nonzero C_j's are chosen randomly. This model has previously been applied as a model of an impure charge-density-wave system,[10] and is a discrete generalization of the phase models of Fukuyama and Lee.[8] The justification of including only interaction of the chain with impurities is that in the absence of the impurities, the pure incommensurate system is not pinned anyway.[20] In our model, the sinusoidal potential represents the charge-density wave, which is much more rigid than the lattice[10] and the chain represents the lattice. Our procedure is similar to previous calculations by the author on such a model.[21] The system is started in an equilibrium configuration for E just below E_T. This configuration is found by iterating the equilibrium equations [i.e., Eq. (7) with the left-hand side set equal to zero] for many choices of x_1 until the force on x_N vanishes.[20] The lowest energy configuration is taken. As pointed out in previous work,[20] it can become very difficult to determine the equilibrium configuration accurately for longer chains because the values of x_1 at which equilibrium occurs become more and more closely spaced as the system gets larger. As a consequence, the equilibrium configuration for longer chains was not found to the same accuracy as for the shorter chains. Nevertheless, the dynamical calculations should still be correct. The dynamical calculation is performed by taking the system initially in an equilibrium configuration, increasing E to a value above E_T, and solving Eq. (7) for the overdamped case (i.e., neglecting the first term on the left-hand side). This approximation probably overestimates the effect of the damping that is present and eliminates transient and oscillatory effects. Nevertheless, it should still be semiquantitatively correct, as discussed earlier. The ratio of the natural period of the chain to the sinusoidal potential period b/a was taken to be 100/77 (which is equivalent to 100/23, which is not far from the value for $NbSe_3$). Figures 1 and 2 show the results of chains containing 77 atoms, 20 of which are impurities, for two different distributions of the impurities. The figures show the mean drift velocity (found by finding the time for the center of mass of the chain to move one lattice constant of the sinusoidal potential) versus $E/\alpha a$. The impurity potential was taken to be $V_0=0.005\alpha a^2$. It should be noted that both the value of E_T and the shape of the curve near E_T depend on the impurity configuration. As the system approaches the thermodynamic limit, we would expect the dependence on impurity configuration to disappear since all possible local configurations are included in a large system. We have calculated the value of E_T for systems of ever increasing size with the impurity concentration fixed at the value 20/77, by solving the equilibrium equations for increasing values of E until E reaches a value at which equilibrium solutions can no longer be found. This is E_T. A plot of E_T as a function of N^{-1} where N is the number of atoms in the system is shown in Fig. 3. The curve appears to extrapolate to about $E_T=0.001\alpha a$. Although there are no data for very large N to rule out the possibility that the curve dips to zero when N^{-1} approaches zero, the next paragraph argues that such behavior will not occur.

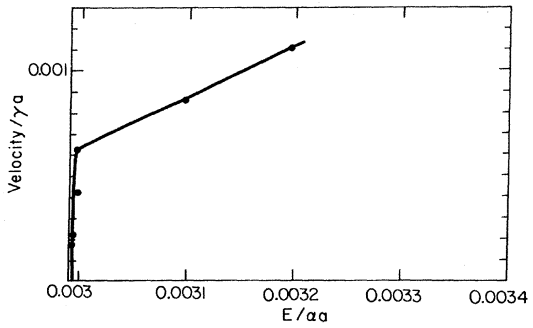

FIG. 1. Velocity of a 27-atom chain with 20 impurities relative to the sinusoidal potential as a function of the applied electric field is shown. Both variables are in dimensionless units.

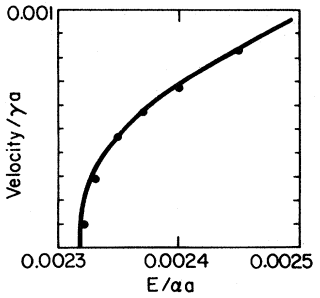

FIG. 2. Velocity of a second 77-atom chain with 20 impurities with a different configuration as a function of the applied field.

In any infinite impure system, there exists the probability of there being a large number N_1 of nonimpurity atoms in a row given by $p=(1-C)^{N_1}$, where C is the impurity concentration. The net force on each such region (of course, unopposed by pinning due to impurities) is N_1E. The mean number of atoms between such regions is $\sim 1/p = (1-C)^{-N_1}$. The unopposed force on the nonimpurity region must drag these atoms, which provide a pinning potential $\sim CV_0(1-C)^{-N_1}$. Thus, it is quite clear that the force due to the electric field on the impurity-free regions will never be large enough to "overwhelm" the pinning due to impurities located between such regions. This argument implies that there should be a well defined threshold field in the thermodynamic limit (i.e., no tailing off of the velocity versus E curve as E approaches zero).

We have also calculated E_T as a function for V_0. Our results are qualitatively similar to Fig. 5 in a paper by Teranishi and Kubo.[22] Therefore, they will not be reported here.

One way one might think of interpreting the velocity versus E behavior is to assume that our system consists of a collection of finite strands containing charge-density waves with a distribution in values of E_T due to variations in both the number and distribution of impurities in each

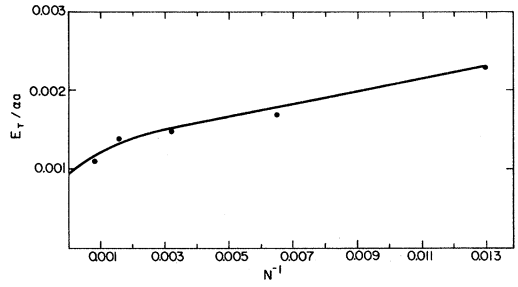

FIG. 3. Threshold field as a function of the reciprocal of the number of atoms in the chain.

segment. Assuming the $(E-E_T)^{1/2}$ behavior (and zero for $E<E_T$) for each strand and averaging over a rectangular distribution of E_T running from $E_{T_0}-\frac{1}{2}\Delta$ to $E_{T_0}+\frac{1}{2}\Delta$ gives

$$(E-E_{T_0}+\tfrac{1}{2}\Delta)^{3/2}$$

which has threshold at $E_{T_0}-\frac{1}{2}\Delta$ and is concave upwards, in qualitative agreement with experiment. This would correspond to the case where E_T can only vary over a small range. The lower end of that range becomes the new threshold field. If $E_{T_0}-\frac{1}{2}\Delta$ represents the infinite chain value of E_T this would qualitatively explain the experimental data. This interpretation is consistent with Fig. 3 which shows that the lower values of E_T do occur for the longer chains. Perhaps, a different probability distribution would give quantitative agreement with experiment. Another possibility is to assume an exponential probability distribution of charge-density-wave sizes, as assumed by Longcor and Portis,[23] and a dependence of E_T on chain length of the form

$$E_T = E_{T_1\alpha} + \Gamma/l\,,$$

where $E_{T_1\alpha}$ and Γ are constants and l is the length of a charge-density-wave segment. Figure 3 shows that this form is approximately valid for shorter chains. Longcor and Portis have shown that the above dependence of E_T on l combined with the exponential distribution of l gives the experimentally observed form for the nonlinear conductor shown in Eq. (1). The problem with these interpretations is that it is not clear that the system really consists of such extremely short (of the order of 100 atoms) strands. Thus, it would be useful to see if longer impure systems in an electric field can give the observed experimental nonlinear conductor shown in Eq.(1).

Since we saw in the last section that the arguments which predict square-root threshold behavior for a finite impure system might break down for an infinite system, it is worthwhile to consider longer chains, in order to see if the current versus field curve becomes concave upwards in the thermodynamic limit in agreement with experiment. Figures 4 and 5 show the drift velocity versus electric field for a 616- and a 924-atom chain with a 50% impurity concentration. The impurity potential for these runs was chosen to be about a tenth of the strength used for the runs shown in Figs. 1 and 2. The reason for this choice is that as the chain gets longer the number of possible equilibrium configurations increases rapidly. At 600 or 900 atoms we are already approaching the limit of accuracy

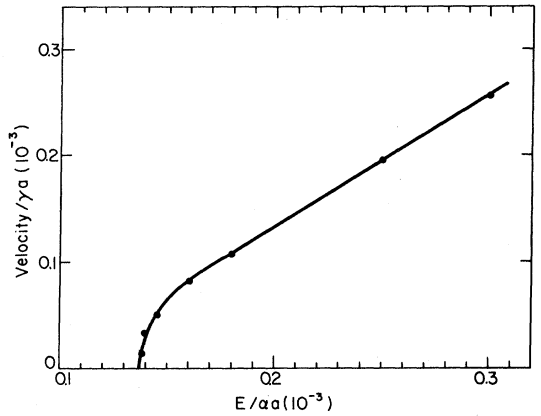

FIG. 4. Chain velocity versus field for a 616-atom system with 308 impurities.

of the computer in trying to solve the equilibrium equations. This is because the number of possible dislocation configurations increases rapidly as the size of the system increases. Decreasing the strength of the sinusoidal potential increases the size of a dislocation and hence fewer dislocations can exist in the chain implying lower multiplicity of the number of equilibrium states. Increased impurity concentration (up to 50%) also helps reduce the multiplicity because it moves us closer to the weak impurity limit.[7,8,20] The 616-atom system exhibits well defined threshold behavior, as predicted in Sec. II for any finite system. There appears to be a more gradual falloff of the current for E closer to E_T, however, than occured for the smaller systems. The threshold field found from the dynamics is $0.000138\alpha a$, whereas the threshold field found by solving the equilibrium equations was only $0.00012\alpha a$, which illustrates the difficulty in finding the equilibrium configuration for such large systems. The dynamical calculation, however, appears to be reasonably behaved. The 924-atom system shows a definite tendency toward the drift velocity versus field curve turning from concave downwards to concave upwards as the size of the system increases (it will show the square-root behavior only right near the threshold field). We have not done calculations on larger systems because the runs near threshold already require huge amounts of computer time for the 924-atom system.

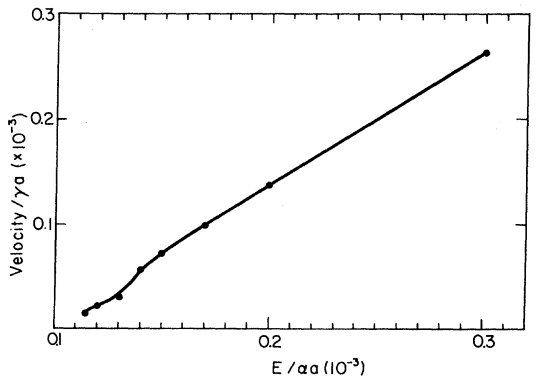

FIG. 5. Chain velocity versus field for a 924-atom system with 462 impurity atoms.

In order to illustrate how the breakaway behavior occurs physically, we have printed out a 19-atom section of a 77-atom chain for several times in Fig. 6 for a field just above threshold. It can be seen that most of the time the system is almost pinned in one place. At a certain time, the impurity atoms in the chain jump rapidly by a lattice constant of the sinusoidal potential to the next near-equilibrium configuration. Usually one impurity starts to shift before the others and pulls the others along. In Fig. 6, the impurity atom initially at $x/a = 57$ appears to jump to about 58 before the other two atoms shown start to jump.

Recently Teranishi and Kubo[22] have reported similar calculations to ours. They study finite temperature effects, which we do not consider, but we have considered much longer chains of atoms. We have concentrated on studying the threshold behavior of the current as a function of electric field, whereas they do not say much about this aspect of the problem. We disagree, however, with their prediction that the threshold

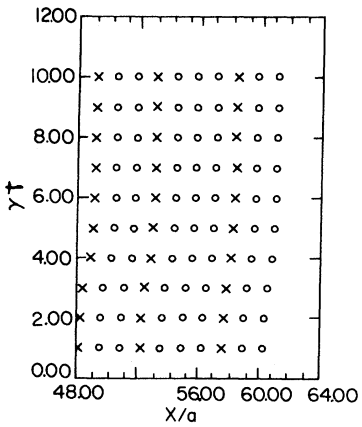

FIG. 6. Atomic positions of a 10-atom section of a 77-atom chain with 20 impurities in a field of $0.00295\alpha a$ ($E_T = 0.00292\alpha a$). The circles are ordinary atoms and the X's are impurity atoms.

field should approach zero in the thermodynamic limit. Teranishi and Kubo's argument is based on the fact that the mean distance between impurities diverges logarithmically with the size of the system. This is because any average over the interimpurity spacing is weighted by the occasional occurrence of very large regions which are free of impurities. Such regions could strongly weight the average. Thus, the average spacing of impurities will not be typical of interimpurity spacing. As has been argued earlier in this article, the fact that such an atypical region in the system can break loose says nothing about whether or not the whole system becomes depinned. A correct calculation of the depinning field must proceed from Eq. (3.7) in Ref. 19 and not Eq. (3.3). It is difficult to understand the assertion that the conductivity becomes infinite for $E_f > E_{fc}$ because it was shown in Ref. 11 that there is damping even at $T=0$, which implies noninfinite electrical conduction. They do not, however, give details of how they arrive at these conclusions.

IV. CONCLUSIONS AND SPECULATIONS ON THE OBSERVED LOW-FREQUENCY NOISE SPECTRUM

It has been shown that in the overdamped limit all threshold behavior in a charge-density-wave system pinned by commensurability should be of the square-root type, which does not agree with the results of Fleming and Grimes.[6] Molecular dynamical calculations on relatively long impure systems (i.e., pinned by impurities) show a tendency for the threshold behavior (which for finite system is also of the square-root type) to become concave upwards in the thermodynamic limit, in agreement with experiment. An average over the threshold field for short systems gives threshold behavior qualitatively like experiment if the distribution in E_T has a low-E_T cutoff. In all calculations reported, impurity concentrations as high as 30% to 50% were used in order to give good statistics and to cut down on the multiplicity of possible equilibrium states. Although the experimental impurity concentrations are much lower, our results should still be qualitatively correct.

Two macroscopic surfaces sliding with respect to one another should also exhibit a similar threshold transition from static to dynamical friction. Although the actual location of the threshold force will be impossible to determine because neither surface is periodic (as is true in our model), once the system begins to slide, similar threshold behavior should be observed (in which the system remains stuck in several almost pinned positions for a long time and then breaks loose). This is the phenomena of "stick-slip friction."

One of the most puzzling features of the data of Fleming and Grimes[6] is the appearance of low-frequency noise once the threshold field for nonlinear conduction has been exceeded. Although this is a phenomenon not explainable by the model calculations presented in this article, it is interesting to speculate on the possible cause of such behavior. Indeed, the observed fundamental frequency implies a characteristic length in the system of about 1μm, such that the charge-density-wave drift velocity divided by this length gives the frequency of the noise. This length is longer than the wavelength or any other length in the system. Actually, it is not difficult to understand where such a length might come from in an incommensurate system. It is well known that an incommensurate system will distort so as to form a structure in which the system is nearly commensurate in most places, with intermittent domain walls or solitons (where the system is incommensurate), which form a periodic lattice.[24] The wave vector of the soliton lattice in $NbSe_3$ will be given by $4\vec{q} - \vec{b}^*$, where $\vec{q}$ is the wave vector of the charge-density wave and $\vec{b}^*$ the nearest reciprocal-lattice vector to $4\vec{q}$. As the charge-density wave, and hence the soliton lattice, slides past the impurities, it will give oscillations of period equal to the lattice constant of the soliton lattice divided by the drift velocity. Using the experimental values of q, this gives a frequency which is over an order of magnitude too large.

Theodorou and Rice[25] have shown, however, that there exists a whole hierarchy of soliton lattices of wave vector $n\vec{q} - \vec{b}_m^*$ corresponding to each integer n such that $n\vec{q} \approx \vec{b}_m^*$, where b_m^* is some reciprocal-lattice vector of the cyrstal lattice. Each such soliton lattice will, if Fourier transformed, contain the above fundamental wave vector and all its harmonics. Thus, each such soliton lattice will result in a small amplitude periodic oscillation in the current, including all possible harmonics. This is in qualitative agreement with Fleming and Grimes,[3] who find low-frequency oscillations in the current with a hierarchy of frequencies and all their harmonics. This explanation is certainly reasonable in the large field limit, at which the charge-density wave has completely broken loose and is moving at nearly constant drift velocity. It is difficult to understand on the basis of this model, however, why each new fundamental frequency appears at zero frequency at a critical value of the applied field. This can only be understood if the breakaway of the charge-density wave occurs as a re-

sult of each member of the hierarchy of soliton lattices breaking loose separately as the field is increased. It would be expected, however, that the soliton lattices corresponding to higher-order commensurate "lock-ins" will break away first. These, however, happen to have the larger lattice constants and the lower amplitudes. Clearly, further theoretical work is needed to understand this phenomenon. More accurate measurements of the charge-density wave vector are also needed for testing these ideas.

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