Vortex-Array Model for Charge-Density-Wave Conduction Noise

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It is proposed that ac voltage (v_{ac}) oscillations observed in sliding charge-density-wave systems are caused by phase vortices at the sample ends. The model's prediction that v_{ac} is length independent is verified experimentally. The long-range phase coherence implied by thermal-gradient experiments is consistent with the vortex model but not with models based on impurities.

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One of the most puzzling features of electronic transport in the series of compounds that demonstrate charge-density-wave (CDW) conduction is the appearance of well defined voltage oscillations¹ when the dc electric field exceeds a threshold value $E_{\mathbf{T}}$. The fundamental frequency of this oscillation has been shown² to be proportional to the current carried by the moving CDW. This phenomenon has been seen³ in NbSe₃, TaS_3 , $(TaSe_4)_2I$, $(NbSe_4)_{10/3}I$, and the bronzes $K_{0.3}MoO_3$ and $Rb_{0.3}MoO_3$, all of which display nonlinear *I-V* curves associated with CDW depinning. Most of the theories⁴⁻⁶ proposed for the ac voltage invoke the impurities as the source of the current oscillations through their interaction with the moving CDW. The simplest such model is the impurity washboard model of Grüner, Zawadowski, and Chaikin.⁴

In the static situation the weakly pinned condensate distorts its phase to maximize the pinning energy gained from the impurities. The average length of the domains within which the phase is roughly uniform is the Fukuyama-Lee-Rice $(FLR)^7$ length λ_{FLR} (10-50 μ m in NbSe₃). It is well established experimentally that the threshold field is determined by the impurity concentration. However, the situation when the CDW moves is less clear. In particular we shall present evidence showing that the phase coherence length (when sliding) is much longer than λ_{FLR} and infer that the impurities play no major role in the (narrow-band) noise generation. (A perplexing problem with domain structures in the sliding CDW is the question of how oscillations in adjacent domains couple to provide the sharp spectra seen. Also the question of what happens at walls separating domains with different velocities is unresolved.)

We propose a model for the CDW noise based on the following assumptions: (I) An array of vortices is created when the CDW phase undergoes an abrupt change such as at the sample ends. (II) Regardless of the phase configuration in the

static (pinned) condensate, when sliding occurs the phase coherence may extend over macroscopic distances approaching the sample length in pure samples. The creation and annihilation of the phase vortices at the sample ends leads to a modulation of the current which is observed as the ac noise. Various theories^{8,9} have explored the effect of internal degrees on the dynamics of the moving CDW. In our model we may regard the conduction noise as a manifestation of the internal degrees of freedom expressed as phase dislocations in inhomogeneous situations. The vortex array insofar as it is an energetically favorable way for the system to overcome phase disruptions is expected to appear at places where the internal electric field or the sample cross section undergoes an abrupt change. Assumption II is an experimental inference. It implies that when sliding occurs the region over which the CDW drift velocity is uniform extends over the whole sample *despite* the existence of inhomogeneous field and temperature distributions. Although II is not derivable from I, the two are not contradictory. On the other hand II is irreconciliable with the existence of a coherent periodic potential generated by random impurities. Further, if II is valid the existence of FLR domains is irrelevant to the dynamics of moving CDW's.

Contact probes have been shown to perturb seriously the motion of the CDW. For example, in a recent experiment¹⁰ it was shown that voltage probes attached with silver paint partition the sample into three segments that oscillate independently. It was proposed that at low E the portion of the sample covered by the paint remains pinned while the uncovered portion slides. This creates a surface S of phase discontinuity between the sliding and nonsliding regions. To allow the phase at S to slip by 2π each time the CDW advances by a wavelength λ_{CDW} one may envision the order parameter being driven to zero at S. However, a more economical way for the phase slip to be realized is by the creation and annihilation of phase vortices at S. Figure 1 shows in sequence the removal of a vortex line (drawn as an edge dislocation in the superlattice) and the advance of the CDW wave front in the left portion of the sample. Each time the bulk CDW phase advances by 2π a vortex sweeps across S, thereby removing the phase discontinuity. Calling the separation of the vortices l_v we have the relationship

$$v_{D} / \lambda_{CDW} = v_{s} / l_{v} = f, \qquad (1)$$

where v_D is the drift velocity of the CDW, v_s is the vortex velocity, and f is the observed fundamental frequency. Equation (1) ensures that the phase-slip rate is equal to the bulk phase-winding rate. It also predicts that the rate at which vortices are annihilated at the sample sides is equal to the so-called washboard frequency and suggests in our model that the observed conduction noise is due to the motion of the vortices.

As a starting point we consider the phase Hamiltonian

$$H = \left(\frac{1}{4}N_0 v_F^2\right) \int d^3x \, (\nabla \varphi)^2 + \int d^3x \, V(\varphi), \qquad (2)$$

where N_0 is the three-dimensional density of states per spin and v_F the Fermi velocity. Inserting the vortex configuration $\varphi = \tan^{-1}(y/x)$ into Eq. (2) (x along the direction of v_D ; y along v_s), we find that the free energy of an isolated vortex equals $\frac{1}{2}\pi N_0 v_F^2 \ln(\lambda_{FLR}/\xi)$, where ξ is the coherence length (approximately 10 nm in NbSe₃). It is clear that creating an array of vortices at S compared with driving the order parameter to zero results in an energy gain since the ratio of



FIG. 1. Schematic drawing of the role of phase vortices (drawn as edge dislocations) in causing phase slip near paint contacts. The open (solid) arrow in (a) indicates the direction of v_D (v_s). The hatched stripes identify two CDW wave fronts that are brought closer together each time a vortex runs the width of the sample [(b)-(d)]. the free energies $\Delta F_v / \Delta F_s = (\pi \xi / l_v) \ln(\lambda_{FLR} / \xi)$ is always less than 1.

In order to calculate the effect of the vortices on the CDW current density $J_{\rm CDW}$ we use the equation

$$J_{\rm CDW} = n_s e \, Q^{-1} \, \partial \varphi / \partial t \,, \tag{3}$$

where $n_s e$ is the condensed charge density and Q is the CDW wave vector. Integrating Eq. (3) around a loop enclosing the vortex array we get that the current difference between the two sides of the array is

$$\Delta J = J_{\rm CDW}({\rm left}) - J_{\rm CDW}({\rm right})$$

$$= -2\pi n_s e Q^{-1} \sum_i \partial y_i / \partial t, \qquad (4)$$

where we have assumed that the array is described by

 $\varphi(x, y, t) = \sum_{i} \tan^{-1} \{ [y - y_{i}(t)] / x \}.$

In general any phase modulation at the frequency f will give rise to an ac current by Eq. (3). We favor the specific mechanism whereby the velocity of the vortices is periodically modulated by the image force near the sample sides. A second mechanism whereby the existence of vortices near the sample ends can modulate the voltage has been proposed by Gill.¹¹ The vortices can modulate the scattering of the *free* carriers and change the Ohmic resistance. Such a possibility may be operative in orthorhombic TaS₃ where the resistance is nonmetallic.

In usual experimental arrangements the current is held fixed by active feedback and the oscillations in the voltage are observed on an oscilloscope or a spectrum analyzer. Under these conditions the current oscillations in Eq. (4) are converted into an oscillating chemical potential difference across the voltage leads by an effective resistance $R_{\rm eff}$ associated with the contact geometry. We calculate R_{eff} to be of order $(\sigma w)^{-1}$ where σ is the Ohmic conductivity and w the sample width. Thus the ac voltage amplitude is independent of sample length. This is consistent with associating the ac source with the ends of a two-probe sample or the ends of the uncovered segments in a four-probe sample. (A similar conclusion holds if the noise is due to modulation of the free-carrier resistance since the source of modulation is associated with the ends.) To test this hypothesis we have made measurements of the ac amplitude in two-probed samples as a function of the sample length l. Starting with a long sample the ac noise was captured by a spectrum analyzer and computer averaged over twenty runs and the area under the fundamental frequency was integrated. The length was then shortened by decreasing the exposed segment between silver paint pads and the measurement repeated. Studies on eight samples each shortened four times on the average showed no variation of the ac voltage with l (Fig. 2) although fluctuations are large. (The ac signal fluctuates by a factor of 2 with a time scale of 2 to 3 sec, hence the need for electronic averaging; averaging by eye or by xy recorder trace is inadequate.) We have extended the study to a sample 7.8 mm long chosen for the spectral purity of its ac voltage and found no decrease of the area under the full spectrum when the length was shortened six times to a final value of 0.13 mm (a factor of 60). These results contradict a recent report¹² that the ac voltage increases with length as \sqrt{l} .

To distinguish this model from those based on the impurity potential we expect that in the pure limit the noise amplitude will be undiminished whereas in impurity models it will ultimately vanish. In addition we expect that in a closedloop sample (with no breaks) no noise should be observed. The appearance of the noise is ultimately related to the destruction of the periodic condensate when it is driven through an interface separating dissimilar regions. The vortex model is also to be distinguished from theories which associate the periodic noise with shot noise generated at sample ends by the arrival of charged periodic solitons.¹³ Quite apart from the difficulty of seeing shot noise in metals, the sample ends are simply not flat enough to resolve spatial



FIG. 2. The ac voltage v_{ac} at 1 MHz for six samples of NbSe₃ vs the sample length l, showing that v_{ac} is lindependent. For each sample the length is shortened four times and the measured v_{ac} is normalized to the value at the longest length. Each data point is the integrated area under the fundamental f, averaged over twenty spectrum analyzer sweeps.

periodicities of the order of 5 nm. The vortices (in analogy with dislocations) do not carry charge although they can modify J_{CDW} through periodic modulations of the CDW phase. On the basis of the value of λ_{FLR} the size of each vortex is of the order of 10 μ m. In contrast to the soliton lattice picture the vortex is a macroscopic singularity moving in a direction transverse to J_{CDW} .

In a second experiment we have observed the effect of a thermal gradient $\Delta T = T_2 - T_1$ on the noise spectrum (Fig. 3). For $\triangle T$ as large as 6 K the fundamental maintains its intrinsic width of 5–10 kHz in NbSe₃ despite large variations in Eand E_T inside the sample.¹⁴ This experiment implies that rather than breaking up into segments sliding at different velocities the condensate slides at two distinct velocities (see below) even though the field E is, say, increasing along l and, simultaneously, the threshold field is decreasing along l (because of ΔT). The energy cost of supporting a broad distribution of v_p (through vortex creation or local condensate destruction) is so prohibitive that the condensate maintains a strictly uniform v_{p} . Under sliding conditions the length scale over which this phase coherence is maintained can reach 4 mm and is clearly distinct from the FLR length which only applies to the static situation. A more important feature of the data in Fig. 3 is the appearance of a *new* fundamental (labeled A) which moves rapidly with increasing ΔT . This new line can be unambiguously



FIG. 3. The effect of thermal gradient on the ac spectrum in a high-purity two-probe sample of NbSe₃. As the thermal gradient increases the fundamental (lines 0) shows no enhanced broadening (lines 1 and 2 are harmonics). The line A which increases rapidly in f is identified with the hot end of the sample (see text). The arrow at 0.67 MHz is a marker. In all traces the current is kept at the same value.

identified with the hot end of the sample because it reverses direction (moves to lower f) when T_2 exceeds the temperature at which E_T is a minimum, and vanishes when T_2 exceeds the transition temperature. The old line (labeled 0) remains roughly unshifted.

The phase rigidity poses serious difficulties for models based on impurities.⁴⁻⁶ The thermal gradient experiment implies the following. Regardless of the phase configuration of the pinned CDW (whether broken up into domains or uniform), once sliding occurs v_D is uniform even in the presence of field and temperature inhomogeneities. This uniformity, which is experimentally indistinguishable from the homogeneous case (i.e., no enhanced line broadening), implies that phase rigidity is maintained throughout the sample. Thus the contribution from all impurities should wash out to zero,⁸ and no current oscillations should be observed. Impurity models which invoke the existence of domains in the sliding state would predict that within each domain $v_{\rm p}$ would assume the value appropriate to the local value of $E - E_T$; the spectrum would broaden into a continuous band in contradiction to Fig. 3.

In contrast the phase rigidity presents no difficulty for models in which the voltage oscillations are associated with boundary conditions at the sample ends. In a sense each end acts as a well localized pinning center. In the vortex model the phase-slip rate at each end is determined by the local temperature of that end. Thus the appearance of two sharp frequencies in a gradient is to be expected. We interpret the appearance of the new line (and its behavior with respect to T_2) as irrefutable evidence that the origin of the sharp oscillations is at the sample boundaries.

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