Classical versus quantum models of charge-density-wave depinning in quasi-one-dimensional metals

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A critique is given of classical models of depinning and transport by sliding charge-density waves (CDW) in quasi-one-dimensional metals. A considerable body of evidence supports a theory based on quantum tunneling over macroscopic distances, particularly on $NbSe_3$ and TaS_3. No sound prediction of the classical approach is consistent with these data. All evidence indicates that it is necessary to treat CDW metals as macroscopic quantum systems with quantum tunneling as an essential feature. Classical concepts are useful for memory, hysteresis, and low-frequency response in the pinned state below threshold. They can only be used for phenomena on length scales larger than the Lee-Rice domain length in the sliding regime above threshold.

I. INTRODUCTION

With the passage of more than eight years since the quantum tunneling model of charge-density-wave (CDW) depinning in quasi-one-dimensional (1D) metals was first proposed,[1-3] there is still no consensus as to the correct approach for describing these systems. Nearly all theorists who have worked on the problem have tried to account for the experimental results with semiclassical models. Classical models have dominated the thinking of experimentalists as well. Since I feel that this has been to the detriment of what should be an exciting field for study of macroscopic quantum phenomena, I have decided to give a critical assessment of the classical approach.

In classical models the CDW is regarded as a classical deformable object subject to random impurity pinning and viscous damping. No plausible classical model has been able to account for any aspect of CDW transport, dc, ac, or combined dc and ac. The only exceptions are phenomena that involve time variations of the phase with no changes in charge density. The latter include oscillations about the pinning frequency and transport in high fields at temperatures so low that there are few normal carriers present to help screen density fluctuations. No prediction has been made that subsequently has been verified experimentally. By plausible, I mean a model that uses classical variables that can be defined within the quantum limits of uncertainty and takes into account the important energies in the problem in at least an approximate way. Any approach that does not treat CDW metals as macroscopic quantum systems misses the essential physics of the problem.

Nearly all theories, classical and quantum alike, are based on the Fukuyama-Lee-Rice[4] (FLR) theory of weak impurity pinning in which there are many (typically $>10^3$) impurities in a phase-coherent domain. The CDW is defined[5] by a density variation $\rho_0+\rho_1\cos[2k_Fx+\theta(r,t)]$, where k_F is the Fermi wave vector in the direction of motion of the CDW and $\theta(r,t)$ is a phase that varies slowly in space and time. For uniform drift with a velocity v_d, $\theta=-2k_Fv_dt$.

A phase-coherent domain is one in which θ changes by no more than $\sim\pi/2$. A typical domain has a length $L_d\approx10^{-4}$ cm in the chain direction (the direction of propagation) and $\approx10^{-5}$ cm in transverse directions. It is convenient to define a chain as an area containing a charge $2e$ per wave length. The charge and current densities per chain are then

$$\rho_{\rm ch}=(e/\pi)(\partial\theta/\partial x),\quad J_{\rm ch}=-(e/\pi)(\partial\theta/\partial t)\ . \tag{1}$$

There are on the order of 10^5 parallel chains in a phase-coherent domain. This designation does not imply that the "chains" necessarily have any physical meaning, but is used as a way of counting the degrees of freedom. One could equally well use a $k_\perp$ value, with the number of discrete $k_\perp$ values equal to the number of chains per unit area.

The present discussion is limited to CDW metals at temperatures sufficient for complete screening of density fluctuations involved in transport by normal carriers. These include the most widely studied metals, $NbSe_3$ at nearly all temperatures and TaS_3 above about 110 K. It is only for such systems that a reasonably complete microscopic theory is available.

When random impurities are present, the phase $\theta_0(r)$ that minimizes the energy of a static CDW is an aperiodic fluctuation of average amplitude π and average period $2L_d$. The associated density functions arise from an aperiodic potential, due to fluctuations, in which the electrons move. The potential gives rise to a small pinning gap through which electrons must tunnel to accelerate the CDW. Experimentally, it is found that there is no conduction by moving CDW's unless the applied electric field exceeds a threshold value, E_T. Above E_T, the CDW current density can be expressed approximately in the form

$$j_{\rm CDW}=\sigma_b(E-E_T)\exp(-E_0/E)\ , \tag{2}$$

where $\sigma_b=n_e e^2\tau/M_F$. Here n_e is the electron density and M_F is the Fröhlich mass associated with ion motion that accompanies the moving CDW. The CDW may be regarded as a macroscopic occupation of a phonon of wave vector $2k_F$. When the CDW is moving with a drift velocity v_d, the phonons have an angular frequency $\omega_d=2k_F v_d$.

From the beginning,[6] the major problem has been to account for the Zener-like factor $P(E)=\exp(-E_0/E)$. The value of E_0 is about that expected for tunneling of individual electrons through a small pinning gap. However, the gap is orders of magnitude smaller than thermal energy. In 1979 I suggested[1(a)] that the problem could be resolved if the gap and Zener factor apply only to coherent tunneling of electrons that advance the CDW condensate in a large phase-coherent volume containing $\sim 10^8$ electrons or more. There is only one thermal degree of freedom for motion of the CDW in a phase-coherent volume. Only one quasiclassical variable can be used to define CDW motion in this volume.

In 1980, not long after the first measurements of ac conductivity were made below the microwave region, I suggested[1(b)] that effects of ac voltages, or combined dc and ac voltages, could be accounted for by application of Tucker's theory of photon-assisted tunneling (PAT theory). At the time, it was not known whether or not the threshold field reflected a gap that must be overcome as in normal-superconducting junctions. Later it was realized that the important driving field is not E but $E_{\rm eff}=E+E_{\rm pol}$, where the CDW polarization field $E_{\rm pol}=-E_T$ just above threshold. Below threshold, $E_{\rm eff}=0$. The PAT theory was successfully applied to a wide range of phenomena, including mixing, harmonic mixing, and harmonic generation.[7]

If valid, the theory implies that effects of quanta of megahertz radiation are being observed at temperatures above 100 K. Particularly since this is *a priori* implausible, it is important to see whether or not such data, as well as the dc I-V characteristic, can be explained with classical models. We shall show that all such attempts that claim success are flawed.

II. DESCRIPTION OF CDW MOTION WITH CANONICAL VARIABLES

Motion of the CDW in the chain direction x may be defined[3] by a variable $N(x)$ that gives the total wave-vector density of the electrons and macroscopically occupied phonons in units of $2\hbar k_F$. The momentum density is

$$P_m(x)=N(x)2\hbar k_F=n_e v_d(x)M_F\ , \tag{3}$$

where $n_e=N_{\rm ch}(2k_F/\pi)$ is the electron density, $N_{\rm ch}$ is the number of chains per unit area, v_d is the drift velocity of the CDW, and M_F is the Fröhlich mass. The mass M_F includes both that associated with ion motion of the macroscopic phonons and the band mass m of the electrons that accompany the moving CDW.

The variable conjugate to $\hbar N(x)$ in the Hamiltonian sense is the phase $\theta(x)$. For a freely moving CDW, the free-energy density is

$$F(x)=\frac{P_m^2}{2n_e M_F}+\frac{N_{\rm ch}\mu(x)}{\pi}\frac{\partial\theta}{\partial x}+\frac{N_{\rm ch}\hbar v_F}{4\pi}\left[\frac{\partial\theta}{\partial x}\right]^2 . \tag{4}$$

The first term on the right-hand side is the kinetic energy, the second is the potential energy of electrons in the field E, with $eE=-\partial\mu/\partial x$, and the third is the increase in Fermi energy (KE) of the electrons from fluctuations in electron density. The Hamiltonian equations of motion are

$$\hbar(\partial N/\partial t)=(N_{\rm ch}/\pi)(\partial\mu/\partial x)-(N_{\rm ch}\hbar v_F/2\pi)(\partial^2\theta/\partial x^2)\ , \tag{5a}$$

$$\partial\theta/\partial t=-2k_F v_d\ , \tag{5b}$$

leading to the equation for acceleration,

$$\partial^2\theta/\partial t^2=2k_F eE/M_F+c_0^2(\partial^2\theta/\partial x^2)\ , \tag{6}$$

where $c_0=(m/M_F)v_F$ is the phason velocity. The second term on the right-hand side is that arising from the polarization field, $E_{\rm pol}$. This equation was derived by a different method by Lee, Rice, and Anderson[8] for a freely moving CDW.

Random impurities give a phase-dependent energy of the form

$$H_{\rm imp}=V_0\sum_j\cos[\Psi_j^0+\theta(r_j,t)]\ , \tag{7}$$

where $\Psi_j^0=2k_F x_j$ is a random phase for the impurity at r_j and $\theta(r,t)$ is a slowly varying function of space and time that gives the density variations and motion of the CDW. The pinning energy is minimized by a static distortion $\theta_0(r)$ that varies on the scale of a phase-coherent domain, $L_d\approx 10^{-4}$ cm. The density fluctuations described by $\theta_0(r)$ give rise to an energy gap through which electrons must tunnel to accelerate the CDW when an electric field is applied. The gap is similar to that in an amorphous semiconductor except that there is a second distortion $\theta_0'(r)$ that makes the energy a minimum when the CDW is displaced by half a wavelength or π in phase.

One cannot arrive at an energy gap by treating the impurities as a perturbation in any order of perturbation theory. The analogous problem in superconductivity is that one cannot arrive at the superconducting ground state by treating the electron-phonon interaction and impurity scattering in perturbation theory. Similarly, one cannot treat low-temperature transport in a semiconductor with a perturbation expansion of the potential that gives the gap.

When the impurities are included in zero order, a tunneling step is required to add $2k_F$, or one macroscopic phonon, to the total wave vector of the CDW. This means that a Zener tunneling factor $\exp(-E_0/E)$ must be included at the elementary step of increasing N by one in a phase-coherent volume or of increasing the momentum of the CDW by $2\hbar k_F$.

It is convenient to include the Fermi energy from density fluctuations on the scale of L_d or less in a phase-dependent pinning energy $N_{\rm ch}V(r,\theta)$. Then θ varies only

on a scale larger than L_d. Changes in the density of the electrons from space variations of θ on a scale greater than L_d can be treated quasiclassically.

With $mv_d=\hbar q$, the equation for free acceleration, (6), becomes

$$\hbar\partial q/\partial t=e^*E\ , \tag{8}$$

where $e^*=(m/M_F)e$. The effect of weak impurity pinning is to add a polarization field

$$eE_{\rm pol}=-\pi\partial V/\partial\theta \tag{9}$$

to E. The semiclassical equation of motion is then

$$\hbar\partial q/\partial t=e^*[E+E_{\rm pol}(r)]\equiv e^*E_{\rm eff}\ . \tag{10}$$

Both q and E may be slowly varying functions of the space coordinates. The quantum equation is similar but includes the Zener factor

$$\hbar\partial q/\partial t=e^*E_{\rm eff}\exp(-E_0/E_{\rm eff})\ . \tag{11}$$

In both cases, the steady-state value for the drift velocity of the CDW is obtained by integrating over a short relaxation time. The quantum equation is

$$mv_d=e^*\tau E_{\rm eff}\exp(-E_0/E_{\rm eff})\ , \tag{12}$$

while the classical equation is similar except for the all-important Zener factor. In both cases, E and $E_{\rm pol}$ may, if desired, be regarded as space averages. At threshold, $E_{\rm pol}=-E_T$ and $E_{\rm eff}=0$. Above about $2E_T$, $E_{\rm eff}=E$. It is the expression with the Zener factor that fits the experimental data[7,9] (Fig. 1). The space average of $E_{\rm pol}$ gives the oscillating voltage (narrow-band noise) associated with dc current flow.

III. CRITIQUE OF CLASSICAL THEORIES

From the above analysis, the minimum classical dc current density should be $\sigma_b(E-E_T)$. How do the classical approaches account for substantially smaller values for fields of the order of E_0 and smaller? The initial approach of Sneddon, Cross, and Fisher[10] was a plausible one; it starts from a freely moving CDW and treats the impurities by perturbation theory. While plausible, it is, as discussed earlier, incorrect in its basic assumptions. The only prediction that could be compared with experiment is that at high fields the dc CDW current should be proportional to $E-cE^{1/2}$. Later, Fisher,[13] by treating threshold as a critical point, predicted that the current should vary as $(E-E_T)^{3/2}$ just above threshold. Neither of these predictions is in agreement with experiment[11,9].

Attempts to improve things by introducing internal degrees of freedom have been equally unsuccessful. By phase locking to an applied ac one can get a step in the dc $I-V$ characteristic on which the dc CDW current is precisely constant for a finite range of dc bias, and on which the current density is uniform throughout the entire specimen to a very high degree, in contrast with the predictions of many-degrees-of-freedom models.[9,11]

Much reliance has been put on computer calculations based on oversimplified models. Many of these take the phase at each impurity site, θ_j, as a classical variable, although this cannot be done within the quantum limits of uncertainty except on an energy scale at least 2 orders of

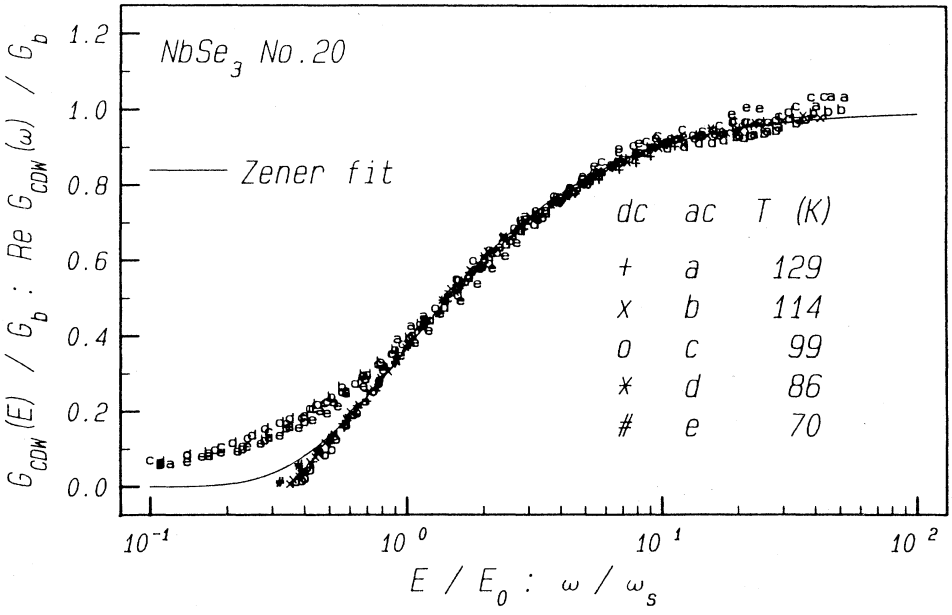

FIG. 1. Field and frequency dependence of conduction in $NbSe_3$ below T_1 (from Thorne, Ref. 9). The dc I-V characteristic, $G_{\rm CDW}(E)/G_b=I_{\rm CDW}(E)/G_bE$, fits the Zener expression $\exp(-E_0/E)$ for fields above about $2E_T$. As predicted by PAT theory, the ac conductance $G_{\rm CDW}(\omega)/G_b$ scales closely with the dc except near threshold and below. Classical concepts can be used to describe the ac conductance near and below threshold, but not above threshold where there is scaling between the ac and dc response.

magnitude larger than the actual energies in the problem. To define the phase at an impurity site within the quantum limits of uncertainty requires an energy per electron of the order $\hbar^2/2mL_i^2$, where L_i is the average distance between neighboring impurities. This is of the order of $(L_d/L_i)^2$, or greater than 100 times the pinning energy per electron. If each θ_j represents a classical degree of freedom, the thermal energy would be far greater than the pinning energy.

Sneddon[12] has used a model introduced by Fisher[13] with infinite-range interactions. The phase at each impurity $\theta_j=\theta(r_j,t)$ is taken to be a classical variable, with the θ_j satisfying equations of motion of the form

$$d\theta_j/dt=a(\theta_a-\theta_j)+b\sin(2k_Fx_j+\theta_j)-cE\ , \tag{13}$$

where $\theta_a=\langle\theta_j\rangle$ is the average phase and $2k_Fx_j$ is a random phase. No terms are introduced that represent the large energies involved when θ_i and θ_j differ and i and j are close together. The claim is made,[14] on the basis of computer calculations, not only to account for the gradual rise in $I_{dc}/(E-E_T)$ above threshold, but also for experiments on effects of combined ac and dc fields that have been interpreted in terms of PAT theory. Such claims are based on what I believe are unphysical and implausible models.

Takayama and Matsukawa[15,16] also claim to account for both the dc and ac characteristics, on the basis of a somewhat different model. It is similar to that of Sneddon in taking the θ_j as classical variables, but introduces terms in the equation of motion to represent elastic forces from space gradients proportional to $\nabla^2\theta$. The results appear to be reasonable for a classical theory when applied to a 1D chain. The ratio I/V rises rapidly above threshold to a constant value above about $2E_T$, as expected for $E_{\rm eff}$. Difficulties arise when they attempt to model a 3D system by adjusting the parameters of the 1D chain.

The dependence on dimensionality is not based on using true 2D and 3D models, but on reducing everything to calculations for a 1D chain. Dimensionality is taken into account in the reduced variables used for the chain. In the model, the frictional force from impurities is in 3D not proportional to the CDW velocity but to the square root of the velocity. Response to applied fields is limited by a frictional force proportional to the velocity. At high fields the latter gives the limiting current, but at low fields the impurity pinning force predominates and limits the current. The crossover field where the two are about equal corresponds to the depinning field E_0. In three dimensions the acceleration is not proportional to $E_{\rm eff}$, as in Eq. (10).

I believe that an error may have been made in the modeling. Only fluctuation effects should depend on dimension. Equation (1) should apply to a classical model. The polarization field $E_{\rm pol}$ can depend on θ but not on the CDW velocity, as has been verified experimentally.[11] At the crossover field E_0 the drift frequency $\omega_d=\partial\theta/\partial t$ is typically $\sim10^8$ rad/sec. In a relaxation time $\tau\approx10^{-11}$ sec, θ changes by only a small fraction ($\sim10^{-3}$) of a radian. The drift velocity v_d at a field E_0 can have no effect on the relaxation rate. The force from impurity pinning can arise only from the θ-dependent polarization energy $V(\theta)$, not the CDW velocity. The maximum polarization force is given by the threshold field, E_T. A reasonable modeling would adjust the density and strength of impurities so as to make the Lee-Rice length, L_d, the same in one dimension as in three dimensions. There would then be no difference in response between 3D and 1D systems.

Matsukawa[16] also claims to derive the equations of PAT theory with a classical model and thus account for results of applied ac voltages and combined ac and dc voltages in the same way as is done by the quantum-tunneling approach. For example, for an applied field $E=E_{dc}+E_{ac}\cos(\omega t)$, the equations of motion in the absence of impurities are integrated over a relaxation time to derive an expression for the ac current:

$$2\alpha\omega I^0_{ac}=E_{ac}[I^0_{dc}(E_{dc}+\alpha\omega)-I^0_{dc}(E_{dc}-\alpha\omega)]\ , \tag{14}$$

where α is a scaling factor inversely proportioned to τ.

This equation is of course valid. In the absence of impurities, I^0_{dc} is linear in the field, $I^0_{dc}=\sigma E_{dc}$ so that $I^0_{dc}=\sigma E_{ac}$. He uses the same equations when impurities *are* present, with I^0_{dc} replaced by the nonlinear I_{dc} as derived by the analysis criticized earlier. A perturbation expansion also is used to derive I_{dc}. In a correct classical theory, E_{dc} would be replaced by $E_{\rm eff}$, and the ac conductivity would be the same as the dc. In PAT theory, the same equations are derived, but with the nonlinear tunneling probability (given by the Zener factor) included.

The expression derived by Matsukawa for α is almost identical to that of the quantum approach. He finds $\alpha=\hbar/2e^*v_F\tau$, but derives it in a form in which $\hbar$ does not appear explicitly from the semiclassical equations of motion. This form can be obtained by multiplying numerator and denominator by k_F and replacing $\hbar k_F$ by the equivalent mv_F. The first form is more physical since it relates an energy drop in a field with the quantum $\hbar\omega$.

Littlewood and Varma[17] have given a classical description of CDW dynamics in which different regions respond on different frequency and length scales. These regions are coupled together. The theory is reasonable when applied to such phenomena as hysteresis, memory, and low-frequency ac response when the applied voltages are below or in the vicinity of threshold. In this region effects of random disorder of the impurities on length scales $> L_d$ are all important and their discussion is pretty much model independent. There is no Zener tunneling below threshold.

In the sliding regime above threshold, Littlewood and Varma rederive results obtained earlier by Fisher:[13] In the classical model the dc current should increase above threshold as $(E-E_T)^{3/2}$ and at high fields, $I_{dc}=\sigma_bE-cE^{1/2}$. They also find $I_{ac}(\omega)=I_{ac}(\omega)-c\omega^{1/2}$ for ω large. None of these predictions are consistent with experiment.

They derive the infinite-range equations of motion (13) as a first approximation of a perturbation expansion of the impurity interactions. These have the difficulties discussed earlier.

There are regions where classical models are appropriate and quantum tunneling does not play a significant

role. One is pure phase oscillations about the pinning frequency where the phase changes in time but not in space. Another is depinning at high fields at temperatures so low that there are two few normal carriers to screen density variations of the CDW.[18] In such depinning the phase again changes only in time.

It is remarkable how well single coordinate models based on the quantum approach fit the data for a wide variety of phenomena in spite of the random disorder of impurities known to be present. Computer calculations should shed light on this if $E_{\rm eff}$ is multiplied by the tunneling probability. Such effects are most important near threshold and below.

I conclude that no sound prediction of classical CDW motion in the charge-compensated sliding regime is borne out by experiment. Experimenters and theorists alike should think of CDW motion as a beautiful example of macroscopic quantum mechanics, with many analogies to superconductivity.

Note added. A number of articles on CDW transport, both experimental and theoretical, have appeared since the manuscript was first submitted. No attempt has been made to cite these since nothing has appeared that would alter the criticisms made. Tucker[19] has proposed to account for some CDW phenomena on the basis of a model with strong pinning. The phase-dependent energy per impurity is of the order of thermal energy at the Peierls transition temperature. For a Lee-Rice domain of length $L_d \sim 10^{-4}$ cm there would have to be of the order of 10^4 such impurities in a phase-coherent volume in order to account for the pinning energy, well within the weak-coupling limit.

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