

Electrodynamics of a Classical Charge Density Wave Model with Random Pinning

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Abstract

We have evidence that the classical random pinning model, if simulated numerically using a phase evolution scheme pioneered by Littlewood, gives dispersion relationships that are inconsistent with experimental values near threshold. These results argue for a revision of contemporary classical models of charge density wave transport phenomena. Classically, phase evolution equations are in essence driven harmonic oscillator models, with perturbing terms plus damping. These break down when we are adding more ‘energy’ into a measured sample via an applied electric field than is dissipated via a damping coefficient behavior in a phase evolution equation. We see the consequences of the breakdown of these phase evolution models in Charge Density Wave conductivity and dielectric functional graphs.

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1. Introduction

In 1986, Littlewood ¹ presented an innovative scheme which incorporates a classical phase pinning model of Fukuyama, Lee, and Rice ^{2 3} for the interaction of impurities in a one dimensional setting. We should note that Littlewood's scheme in numerical form bears striking semblance to the Sine- Gordon equation ⁴ for evolution of phase values along a one dimensional crystal. The impurity sites are randomly distributed in one dimension, and we have that the phase term $\phi(x)$ represents the local 'position' of a charge density wave which interacts via an interaction potential $V_j \delta(x - R_j) = V\delta(x - R_j)$ which is a short range interaction between the phase $\phi(x)$ and an impurity site R_j . V happens to be the strength of interaction at impurity sites. We set $V = 1$ and we interpret this statement as a simplification of an otherwise extremely complex distribution of varying impurity sites in quasi one dimensional metals. The variable R_j (denoting the position of impurities) is randomly chosen, but with an ordering requirement of $R_{j+1} > R_j$. Given this, Littlewood used an overdamped equation of motion as well as dimensionless units in order to give an evolution equation with a first order derivative of phase with respect to time, assuming that phase $\phi(x)$ responds 'instantly' to the effects of an extremely localized interaction of phase with each impurity site given by R_j . This last assumption permits us to integrate between impurity sites so as to come up with a first order in time evolution equation for the phase $\phi(x_j) = \phi_j$, where each

$x_j = cR_j$. The constant, c , is the impurity concentration, and assumes that we have a correlation length L so that we observe $cL^d \gg 1$, ie that we have weak impurity pinning(here, d is the dimensionality of the spatial integration). In our model, we set $d = 1.0$. The remainder of this article is to look at the consequences of taking a DFT (discrete Fourier transform) of current $J \approx \left\langle \dot{\phi} \right\rangle$ to obtain computed conductivity and dielectric values due to the evolution of charge density waves along a one dimensional crystal.

II: Showing the validity of the computer simulation ; initial tests of basic phase values

Several caveats are in order. First, we had to keep impurity sites from clustering too closely about the origin. . We could not allow them to be too close to each other. Otherwise we obtain wildly divergent computed numerical values for several computed physical quantities, especially, the derivative of phase with respect to time, leading to spurious results for conductivity even when the applied E field is $< E_{th}$. In fact, the scheme became so unstable if we had a lot of impurity sites near the origin that the derivative of phase with respect to time would blow up after only several dozen time steps from an initial time. In contrast with this instability, quite stable values of the derivative of phase with respect to time exist so long as the applied field to a quasi one dimensional metal sample (e.g.NbSe₃) was less than a threshold value. We observed stable output values for the time derivative of phase after one hundred thousand time steps. One of the easiest ways to show the effects of a threshold field being exceeded is to take a time average of phase, and to then

observe a E_{dc} (direct current) approximation, with constant E field being applied, and to observe after 500,000 steps how a constant value for time averaged phase was obtained for $E < E_{th}$. We obtain a continuously increasing phase value if $E > E_{th}$ that, comparatively speaking, diverged wildly from the equilibrium values one obtained after a certain number of time steps for $E < E_{th}$. Phase values reached a plateau value which would not deviate even after 500,00 time steps. This is how we obtained the stabilized average phase values as represented by Figure 1. When we do not have stability of an averaged value of phase over time, we are observing what happens when the applied electric field to an $NbSe_3$ sample exceeds a threshold value. Then, CDW is free to move. However, we cannot guarantee what sort of ‘movement’ happens in this case. We can either have a continuous evolution of CDW in this case, or an abrupt transition. However, having an abrupt transition can lead to a situation for conductivity for which the ‘current’ part of the discrete Fourier transform (DFT) representing conductivity can be either finite or infinite.

Usually, we used an applied E field which had both constant and oscillatory contributions. This applied E field is put into the following evolution equation ⁵ which is non dimensional due to the interaction strength V and applied electric field E having their dimensions ‘rescaled’ by variable changes to non dimensional constants. However, it is important to note that equation 2.1 uses a non uniform distribution of impurity sites which is where there is an interaction between phase and ions in a one dimensional setting. However, the $\Delta\phi_i$ term below represents the interaction between adjacent impurity sites and shows ‘compression’ (or

deformation) of the CDW ‘phase’, while assuming the impurity sites as given by $X_i = cR_i$ have a random distribution of R_i values, while having $R_i > R_{i-1}$.

$$\ddot{\phi}_i = \Delta^2 \phi_i + \frac{1}{2} E(X_{i+1} - X_i) + V \sin(\theta_i + \phi_i) \quad (2.1)$$

Equation 2.1 is due, in part to setting the acceleration term $\ddot{\phi}$ in the (uniform spacing for ‘impurities’) ‘sliding condition’ for CDW presented below equal to zero (called ‘deep damping’ due to importance of the $\frac{1}{\tau} \dot{\phi}$ term) while then, next, randomizing the position of impurity sites which is initially set equally spaced in equation 2. Furthermore, the θ_i expression in equation 2.1 is a ‘randomized force term’ which varies according to a random generation of numerical values between zero and 2π .. Furthermore although the sliding criteria for CDW mentioned in equation 2.2 below assumes no spatial compression (meaning the presence of CDW only, but of no soliton), we can specifically show a distinct spatial behavior for the ϕ ‘phases’ as generated by equation 1 above. We now refer to the uniform spacing between impurity sites equation for the evolution of phase values, by

$$\ddot{\phi} + \frac{1}{\tau} \dot{\phi} + \omega_0^2 \sin \phi = e \cdot \frac{Q}{M_F} \cdot E(t) \quad (2.2)$$

Equation 2.1 explicitly uses $\phi_i = \phi(X_i)$ where $X_i = c R_i$ and c represents impurity concentration for each impurity site on a one dimensional line. R_i represents each place on a one dimensional line for each impurity site and is a randomly set, monotonically increasing function for each i_{th} index which grows larger . We also used a discretized second derivative ^{6 7 8}

$$\Delta^2 \phi_i = \frac{\phi_{i+1} - \phi_i}{X_{i+1} - X_i} - \frac{\phi_i - \phi_{i-1}}{X_i - X_{i-1}} \quad (2.3)$$

If we look at the first end point of the impurity sites, this procedure leads to a rewrite of equation 2.3 which looks like ⁹

$$\Delta^2 \phi_1 = \frac{\phi_2 - \phi_1}{X_2 - X_1} - \frac{\phi_1 - \phi_N}{X_1 - (X_N - L)} \quad (2.4)$$

as well as the second endpoint of the impurity sites giving us

$$\Delta^2 \phi_N = \frac{\phi_1 - \phi_N}{X_1 - (X_N - L)} - \frac{\phi_N - \phi_{N-1}}{X_N - X_{N-1}} \quad (2.5)$$

where L is the grid length used in this simulation of CDW dynamics.

III. Computing the electrodynamics of quasi one dimensional materials (e.g. NbSe₃)

We should now look at the conductivity, as generated by both equations 2.1 and 2.3 (as well as 2.4 and 2.5). We are looking at a discrete Fourier transform (DFT) of the (left hand side) of equation 2.1 in such a way as to have

$$\text{Re } \sigma(\omega) \propto g1 \sum_n \left\langle \dot{\phi} \right\rangle_n \cos(\omega t_n) \cdot \Delta t \quad (3.1)$$

as well as

$$\text{Im } \sigma(\omega) \propto g1 \sum_n \left\langle \dot{\phi} \right\rangle_n \sin(\omega t_n) \cdot \Delta t \quad (3.2)$$

There is a serious issue which needs to be addressed, before proceeding with this development further. We can create a very inaccurate conductivity simulation if the $\dot{\phi}$ term in the conductivity is a higher order term in a first order expression for conductivity. As written, the derivative of phase used here is from a second order

Runge-Kutta simulation which was chosen for robustness of simulation . Having a higher order accurate simulation for the derivative of phase, as symbolically indicated above placed in what appears to be a first order calculation of conductivity would effectively negate the entire purpose of improved accuracy of taking the derivative of the phase calculation, as symbolically referred to in equation 2.1 . We must perform the DFT inside the Runge-Kutta subroutine initially chosen to analyze the left hand side of equation 2.1. accurately. This makes for a very slow calculation on a p.c., but yields accurate DFT results. Otherwise, round off error from the first order conductivity calculation dominates , negating the second order calculations used for the current calculation. This would create effects similar to those obtained if we restricted our calculations of equations 3.1 and 3.2 to first order Euler accuracy.

For the sake of including in both DC and AC contributions to an electric field, we can write

$$E = E_{dc} \tag{3.3}$$

and / or

$$E = E_{dc} + E_{ac}\sin(\omega \tau) \tag{3.4}$$

When these electric field values are put into both Equation 1.1 and either of the conductivity equations, we get striking conductivity graphs for both real and imaginary parts with the following qualitative features When the electric field applied to a sample is defined by equation 3.3, and we look at the conductivity, we have that there is a critical value for frequency in which the imaginary conductivity goes through an inflection point and decreases, whereas the real conductivity undergoes a decrease in its rate of increasing value and eventually levels off. Here,

ω_c is this critical value for frequency, which presupposes that the modulus of the applied electric field is below a threshold value, E_{th} . Also, ω_c is a critical value for frequency which is different for different materials.. We see both cases in Figure 2 as well as average phase values over time as given in Figure 1 Note, also, that if we look at Figure 3 that the solid line comes from evaluation of Equation .1 with a uniform spacing between impurity sites, and with the random phase set equal to zero . Then $\phi \approx \text{SIN}^{-1} (E_{dc} / E_{th})$ and ϕ has no time dependence. The squares represent what happen when everything is initially the same as when we plotted the solid line, but we have non uniform spacing between impurity sites. The graph due to the triangles results when we no longer keep the random phase equal to zero but other than that use identical conditions we used to graph the squares plot. We performed these simulations of equation 1.1 with the time derivative turned off in order to confirm that, indeed, we are getting expected behavior, and that our numerical simulation is actually performing well. When we look at Figure 1, we should note that just because we have a monotonically increasing phase (with nonuniform spacing between impurity sites and an electric field above a specific threshold value) that this does not imply that the CDW moves continuously or in jumps. Furthermore, we also have dielectric plots which are plotted against increasing frequency according to:

$$\text{Re } \varepsilon(\omega) = 4\pi \left(\frac{\text{Im } \sigma(\omega)}{\omega} \right) \quad (3.5)$$

as well as

$$\text{Im } \varepsilon(\omega) = 4\pi \left(\frac{\text{Re } \sigma(\omega)}{\omega} \right) \quad (3.6)$$

We find that if we re - scale dielectric measurements versus an applied electric field by resetting $\varepsilon / \varepsilon_{initial}$ in place of just ε versus \mathbf{E} field (applied to an experimental sample) that as the frequency ω gets much smaller than ω_c we observe increasingly non linear dielectric behavior as the E field approaches E_{th} . Curiously enough in this situation, we observe an almost linear line plot dependence of dielectric values on the \mathbf{E} field if $\omega \approx \omega_c$ almost up to where the applied electric field has a magnitude $\mathbf{E} \approx \mathbf{E}_{th}$. This is striking, because when we have an applied electric field with a magnitude at or just above \mathbf{E}_{th} we observe the dielectric value with singular behavior. It appears at first glance that low frequency behavior of the dielectric is much more continuous in low frequency regions as \mathbf{E} approaches E_{th} than in the higher frequency situations. This is shown in the almost flat graph of when the frequency divided by ω_c is either .75 or one in figure 4a. On the other hand, figure 4b has wildly divergent plots as frequency drops to $.3 \omega_c$. Figure 5 provides a direct comparison of several dielectric plot results Even when an electric field being applied to a quasi one dimensional material (e.g.NbSe₃) has a modulus value below a threshold field value , the results shown in the following dielectric plots do not have experimental verification.

What is not shown in these figures is the singularity blow up in dielectric response as the applied electric field reaches the so called threshold value. Interestingly enough , Figures 4a,4b, and 5 say that the non linearity in the

response actually increases as $\frac{\omega}{\omega_c} \rightarrow 1$. The abrupt transition to an ‘infinite’ dielectric value actually becomes more pronounced as $\frac{\omega}{\omega_c} \rightarrow 1$, which is unusual to say the least.. The conductivity is also very unusual. We have in Figure 2 a demonstration of what we can simulate for conductivity when simulating results with applied electric fields that are below a threshold value. Below a threshold electric field value (which is the modulus of the applied electric field to a sample) the agreement with classical results is adequate. This abruptly changes as one passes the applied electric field value. The resultant blow up was something which could not be easily simulated, since we have dispersion relationships (both dielectric plots and conductivity) showing infinite values at electric field values precisely at the calculated threshold . However, in all of this, we still have a question: Why does a classical model of conductivity perform well when an electric field is applied to , say, an NbSe₃ crystal in low temperatures, and then perform so poorly in regions in which we pass a threshold value for the applied electric field to the NbSe₃ sample? An obvious first approximate answer to all of this is to consider dissipative effects, or lack thereof, as contributing to unwanted surge in calculated conductivity values when $E \geq E_{th}$ and appearing to signify almost discontinuous behavior in the conductivity and dielectric calculations when $E \approx E_{th}$.

We shall endeavor to set forth a formulation of how to evaluate a current expression in NbSe₃ which helps us avoid the unphysical behavior we obtained numerically when we had infinite conductivity values when the applied electric field exceeded a threshold value, thereby contravening known experimental results..

Clearly, a model of phase evolution which is, in essence, a driven harmonic oscillator with dissipation removing only a part of ‘energy’ placed within a material sample by an applied electric field is only partly right.

IV: Conclusions

The importance of these simulation results lies in showing how classical models for phase evolution over time are insufficient to take in account all of the physics which is pertinent when we have an electric field which is applied to an material like a NbSe₃ sample that exceeds in magnitude a threshold value. In particular, the weird dielectric function behavior observed just *below* the threshold values of an applied electric field as seen in figures four and five raise issues which are related to a defect in the classical model. A driven damped harmonic oscillator can exhibit blow up behavior when an incoming physical driving force exceeds the effect created by damping. In effect, more energy is placed in the system than is lost, and that this makes blow up behavior inevitable, given sufficiently strong electric fields applied to a quasi one dimensional material (e.g. NbSe₃). What we have observed is evidence that a model which works reasonably well for low strength electric fields but is in need of major revisions in order to take into account experimental measurements seen in laboratory conditions. We have an alternative model based upon quantum mechanical tunneling through pinning gaps which will be presented in a later publication which avoids some of the unphysical behavior observed in this classical simulation of dispersion relationships .

Figure Captions

- FIG 1 Average phase $\langle\phi\rangle$ plotted against time (for E_{dc}) with $\langle\phi\rangle$ stabilizing if $E_{dc} < E_{th}$ and $\langle\phi\rangle$ monotonically increasing if $E_{dc} > E_{th}$.
- FIG 2 This is conductivity in the case when one has only an electric field E_{dc} with a magnitude less than E_{th}
- FIG 3 Average phase vs. E_{dc} when E_{ac} is set equal to zero.
- FIG 4 Comparison of scaled dielectric values when one has signal frequency $\omega \leq \omega_c$ i.e. near a critical value ω_c . Figure 4a is for high frequency plots, and figure 4b is low frequency plots
- FIG 5 This, above, is a direct comparison of plots, which does highlights the divergence from linearity occurring as frequency drops. The dielectric is infinite valued when $E=E_{th}$

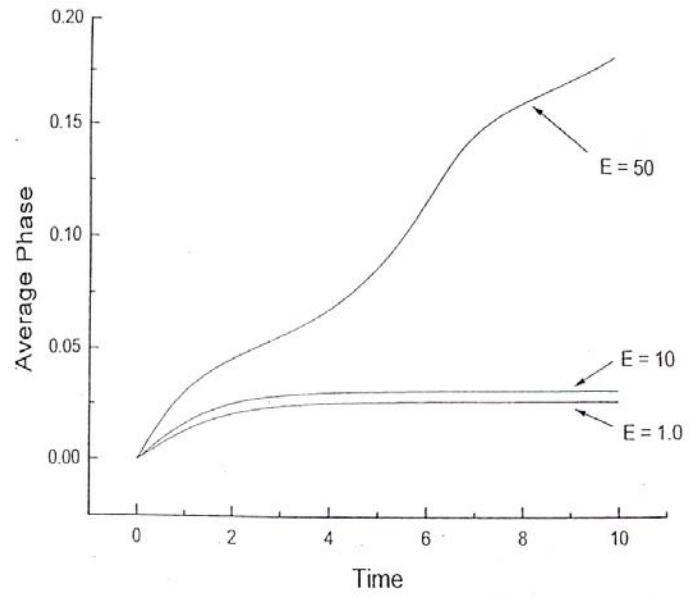


Fig. 1

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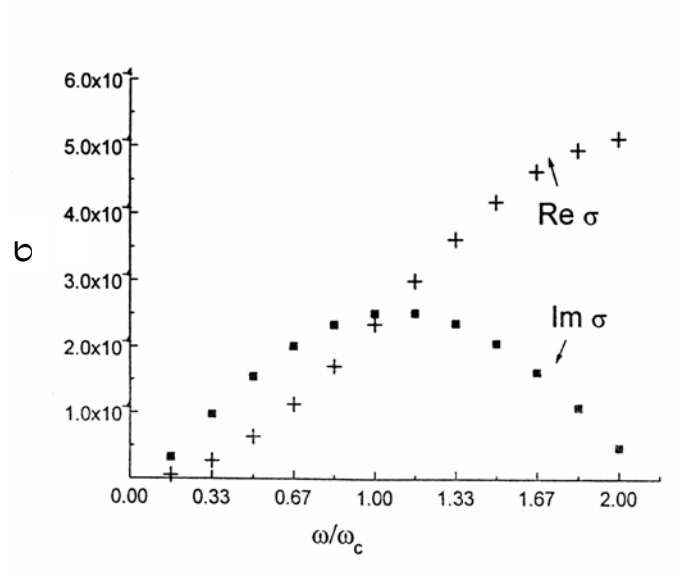


Fig 2

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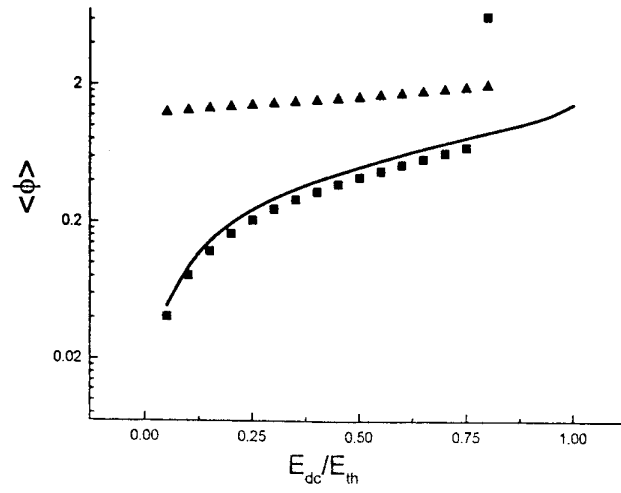


Fig 3

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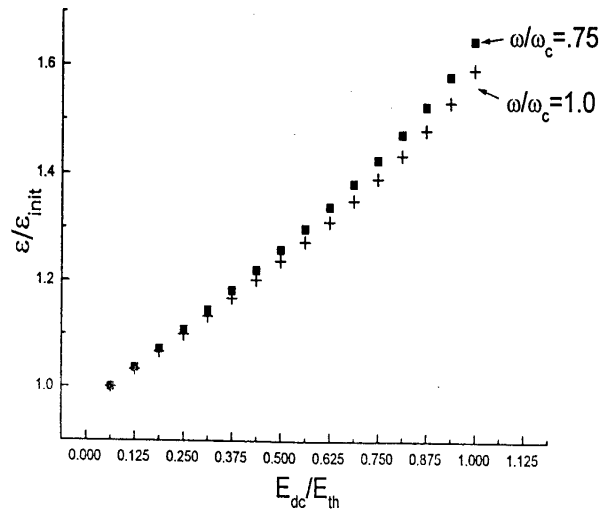


Fig 4a

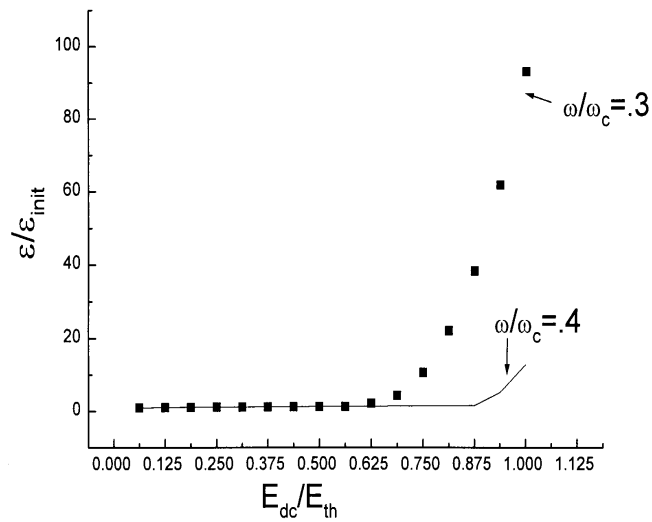


Fig 4b

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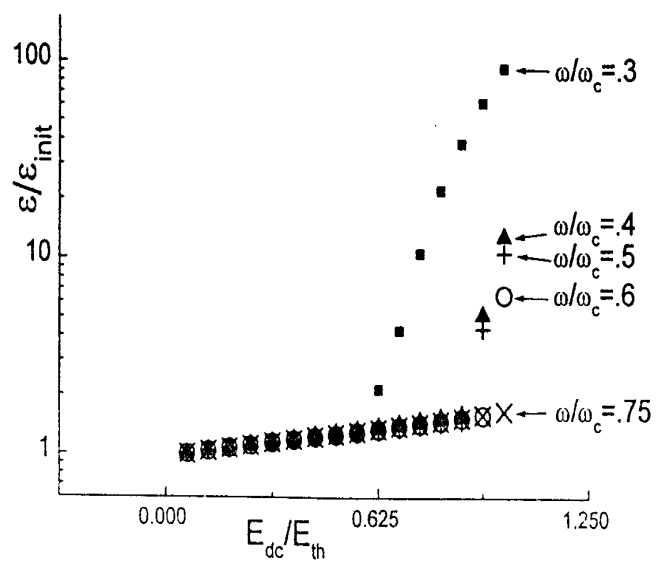


Fig 5

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