

Mysteries in the Response of Density Waves at Low Temperatures

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Abstract. A survey of experiments on charge and spin density wave conductors at low temperatures reveals consistent behavior across disparate materials. This behavior illuminates the remarkable interplay between local and collective pinning and suggests a revised phase diagram for driven density wave systems.

1. INTRODUCTION

Many properties of charge- and spin-density-wave conductors show dramatic evolution upon cooling from T_P to low temperatures. This evolution follows from a complex and poorly understood interplay between collective pinning and local pinning [1-3], Coulomb interactions [4,5] and thermal fluctuations. At high temperatures, local strong pinning barriers are easily overcome and thermally excited single particles fully screen DW charge fluctuations. Observed properties are well described by collective pinning theory [6,7] and observed temperature variations are to a large extent set by simple fluctuation effects and by variations of microscopic parameters with the DW order parameter $\Delta(T)$. The large collective pinning barriers (typically $\gg 10^3 k_B T_P$) give rise to extremely slow thermal relaxation and essentially $T=0$ collective dynamics except near T_P , where thermal creep substantially rounds the depinning transition [7,8].

At low temperatures the characteristic time for thermal activation over local strong pinning barriers becomes substantial, and in fully gapped materials the single particle density becomes so small that screening of DW charges becomes ineffective. The dc velocity-field characteristic changes dramatically [9-11], the ac conductivity shows a low-frequency relaxation mode whose characteristic frequency decreases with decreasing temperature [12] and the thermal properties develop power-law relaxations and other features characteristic of a glass [13]. Many of these properties exhibit activated behavior with an activation energy comparable to the DW gap 2Δ . Unfortunately, this is the energy scale both for single particle excitations responsible for DW screening and for barriers associated with local strong pinning [3,14], and this makes separating screening and pinning contributions more difficult. Disagreements about many basic experimental facts have further complicated the task of developing a coherent theory.

Here we will give a critical review of the low temperature properties of density-wave conductors, focusing on the form of the velocity-field characteristic. We argue that all materials show strongly similar behavior to that observed in the prototypical CDW conductor NbSe_3 . We suggest a revised phase diagram for driven density waves in which, as argued by Larkin and Brazovskii [1], the interplay between local and collective pinning dominates and in which screening effects play a secondary role.

2. SURVEY OF EXPERIMENTS AT LOW TEMPERATURES

2.1 Partially-gapped CDW materials

We begin with NbSe₃, which can be prepared with extremely high crystalline quality and purity, is easy to contact electrically, and can show the basic phenomena of density wave transport far more cleanly than any other material. NbSe₃ is partially gapped and at low temperatures its single particle density remains metallic. Local strong pinning effects can thus be studied in the absence of descreening.

At high temperatures ($T > 2/3 T_p$), the CDW depins at a threshold field E_T and its velocity v increases continuously above it. E_T varies as n_i^2 in thin crystals and as n_i/t in thin crystals, where n_i is the impurity concentration and t is the crystal thickness [7]. E_T is thus identified as the threshold field for collective pinning. Conduction above E_T is only weakly temperature dependent and follows the variation of the Peierls gap near T_p . Depinning of an elastic CDW is a dynamic critical phenomenon [15,16] but the critical regime exists only for $E/E_T - 1 \ll 1$. In practice, finite size effects dominate the dynamics in this region for ordinary size crystals [7,17] and plasticity associated with current conversion at current contacts modifies the detailed behavior in the immediate vicinity of E_T [18].

As the temperature is decreased, E_T gradually increases and the velocity at fields a few times E_T rapidly decreases. At a second characteristic field E_T^* , the velocity increases steeply, and eventually this steep increase evolves at finite T into an abrupt hysteretic discontinuity or "switch" whose magnitude grows with further decrease in T . At low temperatures ($T < T_p/2$), the CDW still begins coherent motion at E_T but it moves very slowly ($\sim 10^1 - 10^4$ Å/s) until E_T^* at which the velocity increases by orders of magnitude to $> 10^8$ Å/s, towards values typical of the high-temperature regime [19,20]. Below E_T CDW creep is vanishingly small, with $v < 10^{-3}$ Å/s even at $E = 0.9E_T$ [21].

Between E_T and E_T^* , the CDW velocity increases approximately exponentially with field and is temperature activated with an activation energy comparable to Δ . As discussed by Cicak in this volume, $v(E)$ in pure and doped crystals of varying thickness is extremely well described by

$$v(E) \propto (E - E_T) \exp[-T_0/T] \exp[\alpha(E - E_T)/T], \quad [1]$$

and is inconsistent with other proposed forms. Eq. 1 strongly resembles the Anderson-Kim formula for thermally assisted flux creep, with an extra $E - E_T$ prefactor included to account for the steep decrease in $v(E)$ at E_T .

Coherent oscillations (narrow band noise) corresponding to CDW motion within the entire crystal cross section (or at least a large fraction of it) are observed in this field range [20], indicating that this creep-like motion exhibits temporal order, just as in the high temperature sliding state. The magnitude of these oscillations is comparable to those observed at high temperatures but their harmonic content is much smaller, suggesting more uniform motion despite the extremely slow velocities. These oscillations at frequency $v(E)$ allow precise measurement of $v(E) = v(E) \times \lambda_c$ even though the single particle resistance R_s is field dependent and the single particle current $I_s = V/R_s$ is up to 10^9 times larger than the collective current I_c , which preclude the use of I-V measurements and the usual formula $v(E) \propto I_c = I - I_s = I - V/R_s$.

The detailed behavior at E_T^* is highly sample dependent. Experiments by Hall et al. [22] on NbSe₃ (which overlooked CDW conduction between E_T and E_T^*) focused on multiple "switches" observed over a narrow fractional field range in crystals that had been abused by Fe-doping, cleaving or quenching. Different crystal segments were reported to "switch" at different fields, and this was interpreted as evidence that switching was associated with phase slip at a handful of isolated defects. We find that samples with poor contacts or with thickness steps that produce inhomogeneous depinning and current flow can show multiple switches or broadened transitions with no switches. Although $v(E)$ clearly increases by orders of magnitude near E_T^* , its exact form is unclear since many mechanisms — both intrinsic and extrinsic — can produce bistability and hysteresis in the presence of a steep nonlinearity [5,23] or transform an intrinsically bistable $v(E)$ into a smooth transition.

Despite this sample-to-sample variability, our work on high quality crystals shows that the transition at E_T^* is clearly a bulk effect separating qualitatively different regimes of CDW motion. E_T^* increases

with decreasing crystal thickness and increasing impurity concentration, is independent of contact separation and position along the sample [19], and is not associated with major changes in longitudinal CDW strains. Most remarkably, Cicak has shown that at finite temperature the transition is associated with a characteristic velocity. Despite E_T^* 's large sample-to-sample variations, the velocity v_T^* just before the transition is given by $v_T^* = v(E \sim E_T^*) = v_0 \exp[-E_b/T]$, where $E_b \sim \Delta$ and v_0/λ_c corresponds to a washboard frequency $v_0 \sim 10^{10}$ Hz. This coincides with the estimated frequency of thermal activation over local strong pinning barriers, proving the bulk origin of the transition and providing insight into its mechanism.

2.2 Fully gapped CDW materials

In $K_{0.3}MoO_3$, $(TaSe_4)_2I$ and both monoclinic and orthorhombic TaS_3 , the single-particle density and conductivity are temperature activated and freeze out at low temperatures. The CDW becomes descreened and its elastic stiffness increases [24].

At $T < T_p/2$ in all of these materials, the current-field relation again exhibits two characteristic fields E_T and E_T^* [25-28]. Between E_T and E_T^* collective conduction is activated with an activation energy comparable to the CDW gap and scales with the single particle conductances [10]. At E_T^* the current increases by many orders of magnitude (>10 in blue bronze at $T=4$ K), and at high fields $E \gg E_T^*$ the conductivity approaches its value in the high-temperature regime. The behavior at E_T^* is extremely sample and contact quality dependent, and some samples show abrupt switching [11,29,30] while others show a steep but continuous increase in velocity [27,31,32]. Compared with $NbSe_3$, contacts are much more problematic and low-temperature conduction anisotropies are much larger. TaS_3 crystals generally have extremely large mosaic disorder and blue bronze crystals can have voids or inclusions of red bronze and other phases. As a result, current flow is generally much more inhomogeneous than in $NbSe_3$, as indicated by generally poor mode locking and coherent oscillation spectra and by the frequent observation of relaxation oscillations.

Recent I-V data for $K_{0.3}MoO_3$ at a single temperature suggest that $v(E)$ between E_T and E_T^* is well described by Eq. 1 [30]. Published fits for TaS_3 use different forms [31] but include the region near E_T^* where $v(E)$ increases steeply. Extracting $v(E)$ from I-V data is complicated because the CDW and single particle conductivities are comparable just above E_T , and because the single particle conductivity is field dependent due to interaction with CDW deformations and to nonlinear effects associated with descreening and carrier trapping at low temperatures. As in $NbSe_3$, *there is only one reliable method to determine the intrinsic form of $v(E,T)$* : the coherent oscillation frequency $v(E,T)$ must be measured in samples that show complete mode locking and/or a single sharp spectral fundamental whose ratio to the nonlinear current density v/j_c agrees with the value $1/n_c e \lambda_c$ expected for homogeneous current flow in the crystal cross section. At present no such data exists for the fully gapped materials.

At intermediate temperatures the velocity $v(E,T)$ remains temperature activated and the CDW conductance scales with the single particle conductance [10], but its field dependence is nonexponential [25,28]. For example, in blue bronze at temperatures between ~ 20 K and ~ 80 K, $j_c(E) \propto E^\alpha$ for $E_T < E < E_T^*$, with $\alpha \sim 1.1-1.5$ [25], similar to the weak field dependence observed in the high temperature regime. Contrary to early claims that temperature activated conduction extended to the high field limit [10], subsequent experiments have indicated a high-field crossover to a weakly temperature dependent high velocity sliding regime [25,28] (which becomes difficult to reach at higher temperatures due to Ohmic heating). More detailed measurements are needed, but $NbSe_3$ appears to show qualitatively similar behavior between ~ 30 and ~ 45 K.

The response of fully gapped materials does show one important difference from $NbSe_3$. In blue bronze E_T strongly decreases with decreasing T [10,25] and can no longer be measured at $T=4$ K. This is qualitatively consistent with the identification of E_T as the threshold for collective pinning — which varies inversely with CDW elastic constant — and with elastic stiffening due to descreening. In o- TaS_3 and m- TaS_3 the behavior is more complicated: E_T decreases monotonically and then shows a sharp maximum near 70 K before decreasing again at low temperatures [10,27]. E_T 's low temperature decrease is consistent with elastic stiffening but the origin of E_T 's maximum (which has not been observed in any other

density wave material) has not been satisfactorily explained. In some samples E_T 's maximum is comparable to the low-temperature E_T^* , and this has led to confusion in the identification of E_T and E_T^* .

Despite this difference, what is most striking is the overall similarity in the response of fully gapped and partially gapped materials. Fully gapped materials show no extra features and no extra characteristic fields even though they undergo low-temperature descreening. It is possible that different processes in partially and fully gapped produce qualitatively similar behavior, or that for some reason screening by ungapped carriers in partially gapped NbSe_3 is ineffective so that descreening by gapped carriers becomes important at low temperatures [19,33] (and yet does not increase the collective pinning threshold E_T). We believe the simplest explanation for similar behavior is that strong local pinning effects dominate in all materials and that descreening effects play a secondary role.

2.3 Other density wave materials

Strikingly similar behavior is observed in sliding spin density wave conductors. I-V measurements on $(\text{TMTSF})_2\text{PF}_6$ show two characteristic fields E_T and E_T^* with temperature activated nonlinear conduction between them and a steep increase near E_T^* to a high conductivity state. The effects of descreening are (as expected) less evident and E_T in the TMTSF salts can increase or decrease at low temperature, depending on the anion [34]. The steep increase in conductivity near E_T^* was fit with a Zener form and interpreted as evidence for a classical to quantum crossover [35], sparking a flurry of theoretical activity. The behavior near E_T^* must be extremely sample and contact quality dependent, and yet the most basic experimental checks were not reported. Consequently, the Zener fit is meaningless, consistent with the nonsensical values of the fit parameters.

Recently, many of the hallmarks of density wave transport including nonlinear conduction with two characteristic fields have been observed in the ladder compound $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ [36]. The persistence of collective effects to high temperatures (>500 K) suggests an unconventional density wave mechanism and provides additional evidence for the ubiquity of these phenomena in sliding density wave materials.

3. PHASE DIAGRAM OF DRIVEN DENSITY WAVES

Here we will briefly summarize our current interpretation of these results in terms of a phase diagram for driven density waves. Our interpretation is strongly motivated by the ideas of Larkin and Brazovskii, Abe, Zaitsev-Zotov and others. A more detailed discussion will be given elsewhere.

The most important observations and conclusions are as follows:

1. The lower characteristic field E_T smoothly evolves from high to low temperatures in all materials except TaS_3 . Based on E_T 's size and impurity dependence in NbSe_3 and its different temperature dependences in partially and fully gapped materials we identify it as the threshold field arising from collective pinning.
2. Below E_T the collectively pinned DW undergoes thermal creep and the depinning transition at E_T is thermally rounded, but these effects are significant only near T_p . The characteristic energy per collectively pinned FLR domain is $\sim 10^3$ - 10^5 $k_B T_p$, and these gigantic barriers produce an extraordinarily slow, glassy response below E_T at $T < T_p/3$.
3. At low temperatures the functional form of $v(E,T)$ between E_T and E_T^* indicates that DW motion is limited by thermal activation over finite barriers having a size comparable to the DW gap. These barriers arise from strong local pinning that produces metastability in the local energy-phase relation and a local pinning force per impurity that adds to the collective pinning force.
4. Coherent oscillations accompany DW motion in this "creep-like" or "thermally-assisted flow" regime. The coexistence of temporal order and creep is possible because of the wide separation of length and energy scales for local and collective pinning. This separation allows averaging of stochastic thermally activated local motion to a friction force that modulates the essentially $T=0$ collective dynamics on the FLR length. This friction leads to a more uniform advance of the DW phase just

above E_T , eliminating the periodic jumps forward responsible for the large harmonic content observed at higher temperatures.

5. At higher temperatures increased thermal activation over barriers renders barrier reduction by the electric field irrelevant, and conduction between E_T and E_T^* crosses over to a temperature activated, weakly field dependent regime.
6. At high temperatures all signatures of E_T^* vanish and roughly temperature independent sliding begins at E_T . At very low temperatures the activated motion between E_T and E_T^* freezes out, and roughly temperature independent sliding begins at E_T^* . In the $T \rightarrow 0$ limit $E_T^*(0)$ is the true threshold for motion. It may be determined by the total pinning force, equal to the sum of the collective pinning force and the much larger strong local pinning force $\propto n_i$, although dynamical effects may produce bistability and large hysteresis.
7. At low but finite temperatures, an apparently first-order transition occurs near E_T^* to a high velocity sliding state when the DW washboard frequency reaches a critical frequency $v_T^* = v_0 \exp[-E_b/T]$. v_T^* is roughly independent of crystal thickness and impurity type and is comparable to the characteristic frequency of activation over the barriers responsible for the form of $v(E,T)$ between E_T and E_T^* . At $T=0$ the phase boundary given by $v_T^*(T)$ must intersect with $E_T^*(0)$. At finite T E_T^* is the field at which v reaches v_T^* . E_T^* is only weakly temperature dependent because both the critical frequency $v_T^*(T)$ and the washboard frequency at a given field above E_T are temperature activated with comparable activation energies. E_T^* 's size dependence follows from the size dependence of E_T , which sets the field at which a given washboard frequency is reached.

4. SOME QUESTIONS

Although a consistent phenomenological picture of low-temperature density wave transport is emerging, we are still far from having a detailed microscopic description. We conclude with a few of the many basic unanswered questions:

1. What is the correct theory of $v(E,T)$ between E_T and E_T^* ? What are the relevant barriers, and how are they related to impurity type and concentration? As pointed out by Cicak in this volume, the notion of one impurity one - barrier within the simple Anderson-Kim picture fails to account for the value of the field exponent α in Equation 1. At typical impurity concentrations of 100-1000 ppm the mean impurity separation is only $\sim 35-80$ Å and so the assumption of independent impurities may break down.
2. What is the nature of the transition at v_T^* ? Simple arguments based on thermal activation over local barriers might give a crossover, but the apparent first-order character of the transition suggests nontrivial dynamical effects are important.
3. What role does single particle freeze-out and descreening play in the response of fully gapped density waves? Descreening has been invoked to explain both $v(E,T)$ and the temperature activated low-frequency dielectric response $\epsilon(\omega,T)$. Can the dielectric response also be understood in terms of strong local pinning [1]?
4. How generic is the behavior discussed here to driven disordered systems? Calculations based on phenomenological models of DWs, vortex lattices and Wigner crystals [16] so far do not predict the most important features of the DW phase diagram because they ignore the coexistence of contributions from local and collective pinning [1].

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