Critical Divergence of the Transient Response of the Charge-Density Wave near the Depinning Threshold in $K_{0.3}MoO_3$

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When a charge-density-wave system is prepared in an unpolarized state the transient response to a step current shows a sharp divergence near the threshold for depinning. We interpret this singularity as a phase transition from the ordered pinned phase to the moving state.

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In the class of quasi one-dimensional charge-density-wave conductors[1] exemplified by $NbSe_3$, TaS_3, and $K_{0.3}MoO_3$, the charge-density-wave (CDW) condensate can be depinned by an applied electric field E. Despite intensive investigation, a consistent picture of the depinning process remains elusive. Among the numerous models which have been proposed, Fisher[2] has made the interesting suggestion that depinning is a phase transition in which the applied field E induces a transition from the pinned to the moving state. If the depinning is indeed a phase transition it should be possible to demonstrate critical divergence of the response of the system to applied fields which implies diverging length scales as the transition is approached. In this paper we report the observation of a sharp divergence in the transient response of the pinned CDW at a critical field E_c near the threshold field for depinning, E_T, in $K_{0.3}MoO_3$. The quantity measured is the total charge Q^* accumulated during the transient response of the system with the system initially in an *unpolarized* state. (Q^* is proportional to the irreversible polarization P^* of the CDW induced by E.) There is also strong evidence of increasing length scales as one approaches the transition.

In the pinned state the configuration Φ of the CDW is nominally determined by the impurity distribution and the elastic energy. However, Φ is especially sensitive to the recent history of the sample. The processes of thermal cycling or field cycling which force the system into special configurations have been studied in detail.[3-10] When a field strong enough to depin the CDW is applied and then removed the repinned CDW is in a singular state called[6] the "polarized" state. The polarization P^* is difficult to observe because of the presence of free carriers which screen out internal fields. Nonetheless, there exists a strong coupling[7] between the free-carrier density and Φ, such that the degree of polarization of the condensate is reflected in the weak-field resistance R. In the unpolarized state R lies on the perimeters (saturation curves) of the R vs T hysteresis loop, whereas in the fully polarized state R lies close to the midpoint. R may differ by as much as 20% between the two states in both $K_{0.3}MoO_3$ and TaS_3. Without recourse to R, it is possible to observe directly the build-up of P^* as a voltage lag in the transient response[11] of the system to a step current. Gill[10] first showed that when a current pulse of the opposite sign is applied to the polarized state a voltage lag is induced. This arises because P^* is forced to reverse itself and build up in the opposite direction. Gill's pulsed (sign) memory effect has been extensively studied[1,5,10] in the CDW compounds.

We have found that if the temperature T is changed by a finite amount (~ 10 K) the polarization built up in the repinned state is *destroyed*, leaving the sample in an unpolarized state at the new T. If a current pulse of the same sign is now applied a prominent voltage lag is observed because the polarization has to be built up anew. Hence, in contrast to Gill's effect, ΔT alone without field reversal is sufficient to induce a voltage-lag response. Figure 1(a) summarizes the process of recreating the unpolarized state in the phase diagram defined by R, E, and T. At 1 the sample is in the unpolarized state Φ_1. (R is on the cooling saturation curve.) Application of a pulse forces Φ to evolve to a new configuration Φ_2 or Φ_2', depending on the amplitude of the pulse. Reflecting this change, R increases irreversibly to 2 (or 2'). When the pulse is removed both R and Φ are frozen at these values (3 or 3'), indicating that the CDW is repinned in a configuration distinct from Φ_1. To recover the unpolarized state T is cycled to destroy the polarization induced by the pulse ($3 \rightarrow 4 \rightarrow 1$). Note that if a second pulse of the same magnitude E and sign is applied at 3, no voltage lag is observed since P^* appropriate to E already exists. In contrast, a prominent E-dependent voltage lag is always observed at 1.

The accumulated polarization charge Q^* delivered by the pulse is determined as follows. In response to the step-function current the voltage is observed to increase abruptly for about 80% of the final amplitude before saturating slowly for the remaining portion [Fig. 1(b)]. The transient pulse is preamplified and captured on a digital oscilloscope. Expressing the total instantaneous current as $I - V'[G_0 + G_S(V')] + dQ^*/dt$ we compute Q^* by integrating the transient current:

$$Q^*(V,t_p) = \int_0^{t_p} \{1 - V'(t)[G_0 + G_S(V'(t))]\}\,dt, \quad (1)$$

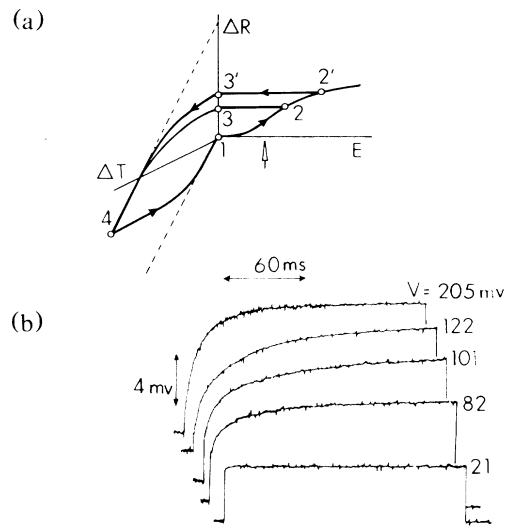

FIG. 1. (a) The phase diagram in R-E-T space for the configurations of the pinned CDW. A current pulse is applied at 1 and the charge Q^* accumulated during $1 \rightarrow 2$ is measured. The path $3 \rightarrow 4 \rightarrow 1$ is used to erase the polarization. Broken lines indicate the cooling and warming saturation curves. A divergence in Q^* occurs at E_C (open arrow). (b) Recorder traces of the voltage response to a step current in the unpolarized state of $K_{0.3}MoO_3$ at 80.0 K. A bridge is used to suppress the zero level of each pulse.

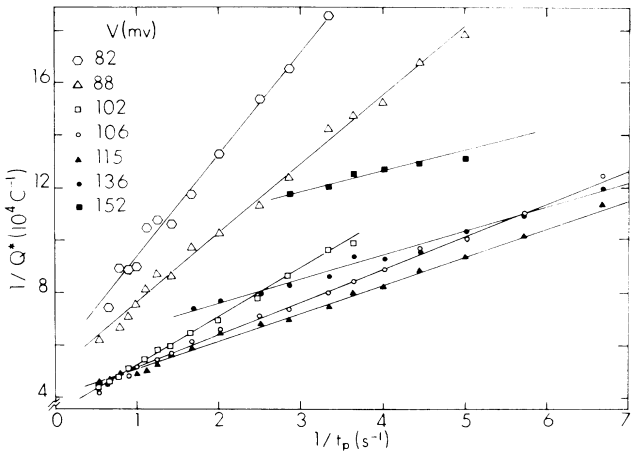

FIG. 2. The dependence of $1/Q^*$ calculated from Eq. (1) on the cutoff t_p. By plotting $1/Q^*$ vs $1/t_p$ we find that the variation is linear for $t_p > 0.1$ s. The data are taken on $K_{0.3}MoO_3$ (sample size $6 \times 1 \times 0.5$ mm^3) at 80.0 K.

where G_0 (G_S) is the Ohmic (CDW) conductance, t_p is the pulse width, $V'(t)$ is the instantaneous voltage at time t, and $V = V'(t_p)$. We emphasize that because Q^* is irreversibly accumulated these are single-shot measurements. If t_p is chosen to be very long (1.8 s) a second pulse with identical parameters will show a Q^* that is essentially zero. (The contribution of the reversible part[11] dP/dt makes a negligible contribution to Q^*.) To measure Q^* for another value of V the unpolarized state has to be recreated by warming up the sample by 30 K and slowly recooling to the same T. For each pulse we have systematically studied the dependence of the integral in Eq. (1) on the cutoff t_p to demonstrate that a finite Q^* exists in the $t_p \rightarrow \infty$ limit, and to explore how this limit is approached. It is convenient to display the approach to the limit by plotting $1/Q^*$ vs $1/t_p$ for each value of E (Fig. 2). At all fields $1/Q^*$ varies linearly with $1/t_p$ at long times. The intercepts of the straight lines with the vertical axis (corresponding to the $t_p \rightarrow \infty$ limit) indicate that as V decreases from 152 to 106 mV, $Q^*(V,\infty)$ increases rapidly. As V decreases further from 102 to 82 mV, $Q^*(V,\infty)$ turns around and decreases rapidly. Thus, $Q^*(V,\infty)$ goes through a sharp maximum as V crosses $V_c = 100.5$ mV. Similar peaking of Q^* evaluated at finite t_p is discernible from Fig. 2. However, because of the crossing of the straight lines the maxima occur at different V's for different t_p's.

This pattern of behavior is confirmed by plotting Q^* vs E in Fig. 3 for fixed cutoff times t_p. The $t_p \rightarrow \infty$ curve forms an envelope for the finite-t_p data. For shorter and shorter probing times the maximum in Q^* occurs at successively higher fields. (This systematic shift in the peak requires the lines in Fig. 2 to cross, as noted above.) Although the divergence in Fig. 3 suggests that Q^* becomes ∞ near 100 mV, there are practical reasons that this cannot be realized with our technique. First, the slow rise in V' at the leading edge of the pulse at short times [Fig. 1(b)] imposes a finite resolution on V. Near V_c we estimate this resolution to be 4%. Second, the diverging time scales implied by Fig. 3 require the integration to be continued to longer and longer times as V approaches V_c from either direction. Sample heating and digitization noise impose an upper bound on the longest t_p that can be used.

The accumulated Q^* measured from the transient current is clearly related to the nonlinear polarizability of the condensate. Thus, it is natural to interpret the divergence in Q^* as reflecting a change in the polarization of the condensate. The singular behavior implied by the power-law falloff alone is strong evidence for a field-induced phase transition at constant T. An analysis of the t_p dependence of the data further supports the notion of diverging length scales implied by a phase transition. The linear plots in Fig. 2 show that $1/Q^*(V,t_p) = 1/Q^*(V,\infty) + b(V)/t_p$ where b is the slope at each value of V. If we assume that the growth of the irreversible polarization within the sample is diffusive we may associate a length scale l to the probing time t_p via $l^2(t_p) = Dt_p$ where D is a diffusion constant, Thus, the accumulated charge assumes the Lorentzian form

$$Q^*(V,t_p) = Q^*(V,\infty)(1+\xi^2/l^2)^{-1}, \quad (2)$$

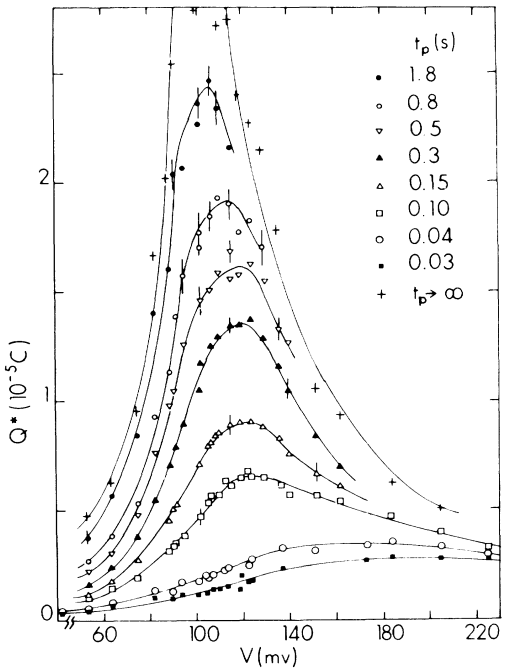

FIG. 3. The variation of $Q^*(V,t_p)$ with V at fixed t_p in $K_{0.3}MoO_3$ at 80.0 K. The solid lines are guides to the eye. The $t_p \to \infty$ limit (crosses) is obtained by extrapolation of the straight lines in Fig. 2 to the vertical axis.

where the correlation length $\xi = [bDQ^*(V,\infty)]^{1/2}$. As V_C is approached from the high-field side ξ grows approximately as $\sqrt{Q^*(V,\infty)}$. (b is insensitive to V.) In the region within 30 mV of the critical field this growth appears to escalate. By rewriting Eq. (2) as

$$\xi^2 = Dt_p\{[Q^*(V,\infty)/Q^*(V,t_p)] - 1\}$$

and examining the variation of the ratio $Q^*(V,\infty)/Q^*(V,t_p)$ vs $V - V_C$ in Fig. 3 we see that the ratio, and hence ξ^2, increase dramatically.

It is instructive to compare the data in Fig. 2 with typical neutron-scattering data near a magnetic phase transition. The comparison suggests that the applied field in the CDW system is probing a characteristic length scale proportional to $1/\sqrt{t_p}$ which diverges as E approaches E_C. Moreover, a plot of $1/I$ vs q^2 in neutron-scattering experiments gives a straight line with an intercept $1/\chi_0$ in the Ornstein-Zernike regime.[12] This is to be compared with the plots of $1/Q^*$ vs $1/t_p$ in Fig. 2 and Eq. (2). Thus, in the picture suggested by the comparison, the growth of the peaks in Q^* for various probing times is consistent with an increasing length scale probed by the current pulse as the critical field is approached. At E_C the length scale diverges to macroscopic values and depinning of the whole condensate proceeds.

As noted above, the evidence for a transition is derived from single-shot measurements, with the system initially in the unpolarized (pinned) phase. It is not possible by monitoring Q^* alone to detect the transition moving in the other direction (E decreasing) or starting from the repinned state. After E is decreased in the last two cases P^* remains frozen at the maximum value attained ($2 \to 3$ in Fig. 1). This remanence precludes study of changes in P^* for subsequent pulses. Nevertheless, if the peak in Q^* signals a transition there should be a cusp in the thermodynamic response of the system at E_C. We have found that in all samples investigated the second derivative d^2V/dI^2 shows a sharp peak precisely at E_C. The coincidence of the peak in d^2V/dI^2 with E_C suggests to us that E_C (rather than E_T which is 20% to 30% smaller) is the field at which depinning of the whole condensate occurs. (If we operationally define E_T as the *onset* of nonlinearity in dV/dI we find that E_T is usually ill defined in $K_{0.3}MoO_3$ below 60 K.) The sharp feature in d^2V/dI^2 is observable for decreasing E, and also starting from the repinned state.

In the disordered phase the system has a simple structure in the free-energy vs Φ landscape. In contrast, if the transition is crossed by decrease in E the system is trapped in a deep valley ("repinned valley") selected by the applied field itself [$2 \to 3$ in Fig. 1(a)]. Our technique for measuring Q^* in fact affords a way to detect decay of the CDW polarization associated with escape from the repinned valley. First the sample is polarized by application of a short depinning pulse. By waiting a time t_d before applying a second pulse we readily determine if P^* has decayed. The longest time for which we have found no evidence of decay is 60 h (at fixed T). From this evidence we deduce that as E is reduced below E_C the system remains trapped in a deep valley from which escape is unlikely on laboratory time scales. Depinning from this polarized state should be qualitatively different from depinning from the unpolarized state. The present technique is sensitive only to the transition occurring in the latter state. Data very similar to Figs. 2 and 3 have been obtained at 80 K in three samples of $K_{0.3}MoO_3$. In one sample we also verified that the divergence is qualitatively similar on both the cooling and warming curves. The magnitude of Q^* at E_C is similar in the three samples when normalized to the volume.[13] This strongly suggests that the polarization is a bulk effect rather than a simple charging that occurs at the ends of the sample. In all three samples E_C is 20% to 30% larger than E_T (the onset of nonlinear dV/dI). Independent of this work, Chen and co-workers[14] have observed a diverging P^* in $K_{0.3}MoO_3$ at 4.2 K near the onset of nonlinear conduction. It should be noted that their onset occurs at fields (26 V/cm) $\simeq$300 times larger than our E_C.

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search for a kink at V_C in the dc response. This research is partially supported by the Department of Physics at Princeton University.

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[11]Transients due to P^* should be distinguished from transients caused by the *linear* dielectric response (Ref. 8). The latter are reversible and occur on much shorter time scales (10 μs at 80 K). In response to a train of pulses the linear dielectric response produces transients at both the leading and trailing edges of each pulse (Ref. 9 and 10). On the other hand P^* does not decay unless T is changed. Hence, once the CDW is polarized by a single pulse the sample will show no voltage-lag response to all subsequent pulses with the same magnitude and sign (disregarding the weak transients caused by the linear response.)

[12]For a review see Jens Als-Nielsen, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 5a, p. 87.

[13]The sizes of the three samples (current along the longest dimension) are $6\times1\times0.5$, $2\times2\times0.2$, and $2.2\times0.7\times0.15$ mm^3. $E_C=166$, 86, and 182 mV/cm, respectively. The maximum Q^*/volume = 10, 12.5, and 13 μC/mm^3. (The volumes are in the ratio 13:3.5:1.) All show a sharp peak in d^2V/dI^2 at V_C.

[14]Ting Chen, L. Mihály and G. Gruner, unpublished; L. Mihály and G. Gruner, in *Nonlinearity in Condensed Matter,* edited by A. R. Bishop, D. K. Campbell, P. Kumar, and S. E. Trullinger, Springer Series in Solid State Sciences Vol. 69 (Springer-Verlag, Berlin, 1987), p. 308.