Polarization Divergence in Driven Charge-Density Waves

T. Chen, L. Mihály,[a] and G. Grüner
Department of Physics and Solid State Science Center, University of California, Los Angeles, Los Angeles, California 90024
(Received 26 October 1987)

We report the first direct measurement of the polarization divergence associated with charge-density-wave depinning in $K_{0.3}MoO_3$ at low temperatures. The field-dependent remanent polarization diverges as $P(E)=P_0(1-E/E_T)^{-\gamma}$ with E_T the threshold field and the "critical exponent" $\gamma<1$. We discuss the novel effects associated with the polarization divergence and the inherent difficulties of the analysis.

PACS numbers: 77.30.+d, 72.15.Nj

In spite of a large body of experimental and theoretical work,[1-3] several aspects regarding the charge-density-wave (CDW) dynamics still remain elusive. The collective mode, driven by an applied dc electric field, develops into a current-carrying state above a well-defined threshold field E_T, signaled by nonlinear current-voltage characteristics.

The low-frequency dynamics is dominated by disorder,[4] brought about by competition between elastic energies associated with local distortions of CDW and local configurational energies due to the interaction of the collective mode with randomly positioned impurities. The Hamiltonian[3] is analogous to that of a random-field X-Y model, with the applied electric field analogous to an applied torque which leads to a phase-winding state. While a single–degree-of-freedom dynamics[5] leads to a sharp threshold field, it does not reproduce several essential features of the nonlinear conduction. One explanation for this is in terms of the quantum effects.[6] It has also been suggested[7] that randomness plays an important role in the nonlinear response of the collective mode. This leads to a semiqualitative agreement with the nonlinear behavior for electric fields $E>E_T$. In particular, the differential conductivity above E_T is given by[8]

$$dj/dE \sim (E-E_T)^{\alpha} \qquad (1)$$

with $\alpha=\frac{1}{2}$, in contrast to the single–degree-of-freedom model which leads to $\alpha=-\frac{1}{2}$. In Eq. (1) α can be regarded as a critical exponent, characterizing the depinning as a dynamical critical phenomenon. Below E_T, the relevant parameter within the framework of the single–degree-of-freedom description is the differential dielectric constant, which is given by

$$dP/dE \sim (E_T-E)^{\beta} \qquad (2)$$

with $\beta=-\frac{1}{2}$. The total polarization $P=2de$, where e is the electric charge and d is the displacement of the average coordinate of the collective mode, is finite and given by $P=2(\lambda/2)e=\lambda e$ with λ the period of the CDW. Various model calculations,[9,10] which take the many-degrees-of-freedom aspects of the dynamics into account, suggest a situation dramatically different from that which is predicted by the single–degree-of-freedom dynamics. In particular, at finite frequencies dP/dE does not diverge (although, strictly speaking, the dc value displays a divergence); instead the polarization P diverges as $E\rightarrow E_T$ from below. Moreover, numerical simulations on a simple model[11] are suggestive of a critical phenomenon, with a well-defined exponent characterizing $P(E)$.

In order to study the dynamics of this driven random-field system, we have performed detailed experiments in the model compound $K_{0.3}MoO_3$ at low temperatures. Recent experiments, performed in the liquid-He[4] temperature range,[12-17] show sharp nonlinear I-V characteristics with threshold field $E_T\simeq 10$–50 V/cm. X-ray measurements,[18] as well as the detailed study of the real-time oscillations in the nonlinear conduction regime,[16,17] indicate that the nonlinearity is due to the CDW conduction and is not a conventional breakdown effect. At low temperatures, where $K_{0.3}MoO_3$ is an insulator with a low-field resistivity $\rho>10^{12}$ Ω cm, direct measurement of the polarization can be made by a capacitive method. This is in contrast to the recent experiments[19] performed at higher temperatures where normal electrons may play important roles and can obscure the critical behavior which may occur.

Before discussing our experiments concerning the field-dependent polarization, in Fig. 1 we show the dc current-voltage characteristics obtained in nominally pure $K_{0.3}MoO_3$ at liquid-He[4] temperature. Similar I-V curves have been reported earlier.[14,15] In order to clarify whether the threshold field is due to the pinning by electrical contacts, or by impurities and/or lattice defects, we measured the length dependence of the threshold field E_T. The sample length along the b axis was shortened by fine polishing after each measurement of E_T. The contacts were prepared with copper electroplating. The threshold fields measured at 77 K range from 65 to 90 mV/cm for various samples. The inset of Fig. 1 displays V_T as a function of sample length L. The linear length dependence of V_T provides clear evidence that, with the contacts prepared by us, the nonlinearity associated with

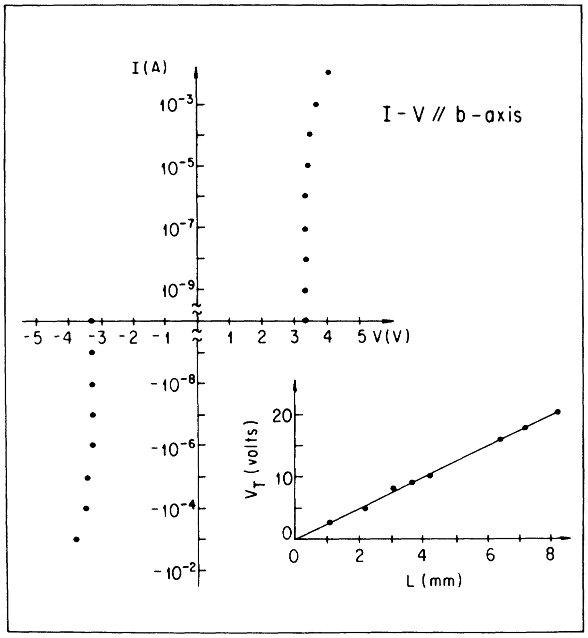

FIG. 1. Nonlinear current-voltage characteristics of $K_{0.3}MoO_3$ measured at the liquid-He^4 temperature with constant dc current configuration. Inset: The threshold voltage V_T as a function of the sample length L. The line gives the threshold field $E_T = 24$ V/cm.

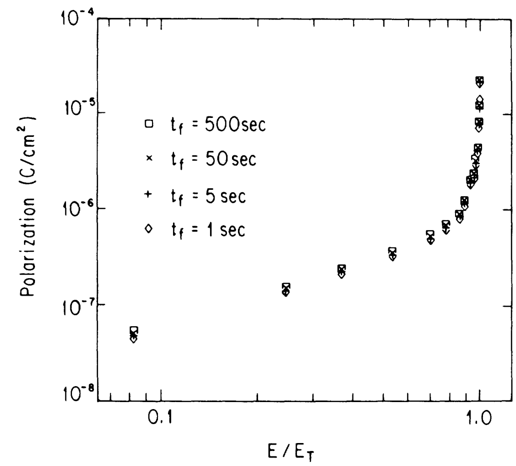

FIG. 2. Polarization $P(E)$ measured at $T=5$ K vs E/E_T. Different symbols represent the values of $P(E)$ measured at different t_f, the times elapsed after the field is applied.

the depinning of CDW's is a bulk phenomenon.

At low temperature, the polarization $P(E)$ below E_T can be measured directly by the measurement of the surface charge which is proportional to the bulk polarization. As the polarization may depend on the initial state as well as the detailed time dependence of the applied electric field $E(t)$, a well-defined protocol has to be followed during the experiment. The initial state was established as follows. After each measurement, the *I-V* curve was again measured with the applied voltage increased above the threshold voltage V_T several times. Specimens conditioned with such a procedure displayed symmetric and reproducible response to voltage steps of both polarities. Procedures similar to the demagnetization of ferromagnets, i.e., applying a sequence of binary pulses with decreasing amplitude, led to essentially the same polarization. Bipolar voltage steps were also applied and resulted in slightly different time dependence of the response for a given amplitude V_0. The time dependence of the polarization was measured by monitoring of the charge accumulation as a function of time after the voltage step $V(t) = V_0\theta(t-t_0)$ was applied. The measurement was repeated with increased V_0. The polarization $P(E)$, measured at different times t_f (assuming $t_0=0$), is shown in Fig. 2 as a function of E/E_T with $E=V_0/L$.

The time dependence of the polarization can also be evaluated and in general we have found that $P(t)$ can be well described by the so-called stretched-exponential form[14,20,21]

$$P(t) = P_0[1-\exp(-(t/\tau_0)^{\zeta})] \quad (3)$$

with $\zeta < 1$ over a broad time domain, between 0.1 and 100 s. The time dependence was, however, not measurable for times exceeding approximately 100 s, as is also evident from Fig. 2. Consequently, we regard P measured at $t_f = 500$ s as close to the remanent polarization which corresponds to the saturation polarization in the $t_f \to \infty$ limit. This point will also be discussed later.

It is clear from Fig. 2 that the depinning is accompanied by a polarization which diverges as $E \to E_T$ from below, a phenomenon drastically different from the single-degree-of-freedom dynamics of this driven system. It is instructive to compare the measured maximum polarization with P^*, expected from a rigid displacement of the collective mode by one-half wavelength, the condition for depinning of a completely rigid CDW condensate. With $n_1 = 5\times 10^{14}$ cm^{-2} for the number of conducting chains per unit area perpendicular to the b axis, the surface charge corresponding to a rigid CDW displacement by $\lambda/2$ (note that there is $2e$ charge per chain per wavelength λ) is given by

$$P^* = en_1 = 1.6\times 10^{-19}\times 5\times 10^{14}\ \mathrm{C/cm^{-2}}$$
$$= 8\times 10^{-5}\ \mathrm{C/cm^2}. \quad (4)$$

The measured maximum polarization P_{max}, from Fig. 2, is 2.4×10^{-5} C/cm^2, close to P^*. Whether this is a coincidence or P^* is a fundamental limit remains to be seen.

More detailed analysis of the apparent polarization divergence shown in Fig. 2 reveals that the field dependence of $P(E)$, as $E \to E_T$ from below, can be described by

$$P(E) = P_0(1-E/E_T)^{-\gamma}, \quad (5)$$

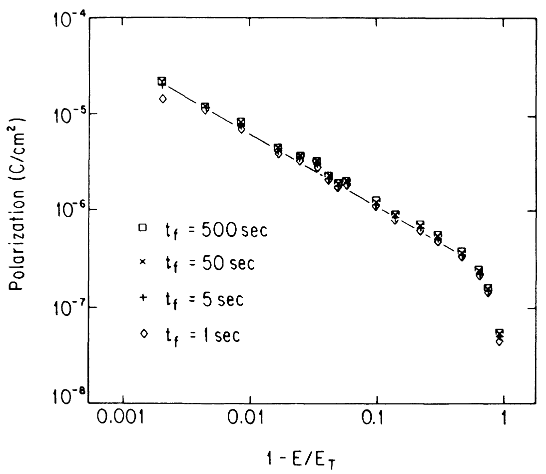

FIG. 3. Polarization $P(E)$ measured at $T=5$ K vs $1-E/E_T$. Different symbols represent the values of $P(E)$ measured at different t_f, the time elapsed after the field is applied. The solid line gives $P(E)=\mathrm{const}\times(1-E/E_T)^{-\gamma}$ with the exponent $\gamma=0.7$.

with the exponent γ characterizing the critical divergence. In Fig. 3, the field dependence of the polarization $P(E)$, measured at different times t_f, is displayed as a function of $1-E/E_T$. The full line in Fig. 3, which fits the data in the electric field range of $0.45E_T<E<0.998E_T$, gives a "critical exponent" $\gamma=0.7$. Here the threshold field E_T is determined from the *I-V* characteristics. It is also evident from the figure that, within the framework of our analysis, the time dependence of $P(E)$, for $t_f>10$ s, does not play a significant role on the critical behavior. On the other hand, we note that because of the inhomogeneities in the specimens, the threshold field may have a slight distribution even within a single specimen. In this case the threshold field E_T, at which nonlinear conduction occurs, signals the depinning of certain sections where the pinning is the weakest and consequently E_T is the smallest. We have, therefore, also attempted to fit Eq. (5) to the experimental results by keeping E_T as an adjustable parameter, but no significant variation of γ was found.

We now discuss the implications of our experiments and some questions relevant to the dynamics of driven random systems. First, the divergent polarization is the clear signature of a many-degrees-of-freedom system, in contrast to the single-degree-of-freedom treatment of the dynamics which leads to finite polarization at $E=E_T$. As discussed elsewhere,[22] the polarization also remains after the electric field is switched off, and this remanent polarization is another bit of evidence for the importance of many metastable states, which arises as the consequence of the disorder. Both a simple one-dimensional (1D) model[11] and more realistic treatments of the depinning[8] lead to a divergent polarization which reflects the development of dynamically coherent domains with domain size $D\rightarrow\infty$ at E_T. Moreover, within the framework of the 1D description, a critical behavior can be recovered only by solutions which go beyond the mean-field treatment of the depinning process. The exponent $\gamma=3$, however, is in sharp contrast to our findings.

There are several fundamental questions which make a detailed comparison between theory and experiment impossible at present. In contrast to the response of a regular or single-degree-of-freedom system, here the response depends (a) on the initial state of the system before the electric field is applied, and (b) on the detailed time dependence of the field. As we have discussed earlier, two different procedures used to establish the initial state led to similar results for $P(t)$ for a given field amplitude E. In contrast to the relative insensitivity of the system as to how the initial state was established, we have found important differences in $P(E)$ with the application of various time-dependent fields. For example, bipolar steps lead to time dependences and also to $P(E)$ different from that of unipolar steps, and also a progressive steplike increase of E_0 leads to $P(E)$ which is different from the polarizations obtained by driving the system out of the initial "depolarized" state. These aspects will be treated in detail in a forthcoming publication. In addition to these questions (which can in principle be answered by experiments), the long-time phenomena lead to complications which may make a description in terms of critical behavior extremely difficult if not impossible. As discussed earlier, the time dependence of $P(E)$ can, for short times, be adequately described by the so-called "stretched exponential" expression. Equation (3) can, therefore, be used to evaluate both τ_0 and P_0 as functions of the applied electric field. Such a procedure also leads to a divergent P_0, but with a critical exponent γ which is between 2 and 3. While this does not affect our analysis based on long-time behavior (see, for example, Fig. 2 and Fig. 3), Eq. (3) suggests that, as τ_0 may be field dependent, $P(E)$ is ill defined. At the same time, Eq. (3) is, most probably, an empirical approximation of the observed time dependences, and is not expected to be correct for rather short or long time scales. Consequently, it cannot be used to evaluate the remanent polarization $P(E)$ and $\tau_0(E)$.

In conclusion, we have observed an electric-field-dependent polarization in $K_{0.3}MoO_3$ at low temperatures, which diverges as $E\rightarrow E_T$ from below with E_T the threshold field for nonlinear conduction. This divergence can be described in terms of a critical behavior with a critical exponent $\gamma=0.7$. Although we have attempted to describe $P(E)$ in terms of a critical divergence, we have also pointed out the uncertainties of this analysis. Finally, we note that a critical divergence in the transient response was also inferred from measurements of the polarization currents at higher temperatures.[19] Aside from the obvious differences related to the different nonlinear characteristics, there are important differences between the two types of measurements. In our experiments, because of large background resistance, the polarization is directly measured, while in experiments utilizing transient currents, the normal-electron contributions are important and may also lead to a divergence in the accumulated charge,[23] comparable to the effect reported in Ref. 19. Further investigation is clearly required to clarify the relation between the critical behavior of the transient response and of the polarization.

Useful discussions with G. Mihály and P. Littlewood are gratefully acknowledged. This research was supported by the National Science Foundation Grant No. DMR 86-72022.

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[a]Permanent address: Central Research Institute of Physics, Budapest, Hungary.

[1]G. Grüner and A. Zettl, Phys. Rep. **119**, 117 (1985).

[2]*Charge Density Waves in Solids,* edited by Gy. Hutiray and G. Sólyom, Lecture Notes in Physics Vol. 217 (Springer-Verlag, Berlin, 1985).

[3]*Electronic Properties of Inorganic Quasi-One-Dimensional Compounds,* edited by P. Monceau (Reidel, Dordrecht, 1985), Vol. 2, p. 139.

[4]H. Fukuyama, J. Phys. Soc. Jpn. **41**, 513 (1976).

[5]G. Gruner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. **46**, 511 (1981).

[6]J. Bardeen, Phys. Rev. Lett. **45**, 1978 (1980).

[7]H. Fukuyama and P. A. Lee, Phys. Rev. B **17**, 535 (1978).

[8]D. S. Fisher, Phys. Rev. B **31**, 1396 (1985).

[9]P. B. Littlewood and C. M. Varma, Phys. Rev. B. **36**, 480 (1987).

[10]P. B. Littlewood, Phys. Rev. B **33**, 6694 (1986).

[11]L. Mihály, M. Crommie, and G. Grüner, Europhysics Lett. **4**, 103 (1987).

[12]W. Fogle and J. H. Perlstein, Phys. Rev. B **6**, 1402 (1972).

[13]A. Maeda, T. Furuyama, and S. Tanaka, Solid State Commun. **55**, 951 (1985); A. Maeda, T. Furuyama, K. Uchinokura, and S. Tanaka, Physica (Amsterdam) **143B**, 123 (1986).

[14]L. Mihály and G. X. Tessema, Phys. Rev. B **33**, 5858 (1986).

[15]G. X. Tessema and L. Mihály, Phys. Rev. B **35**, 7680 (1987).

[16]G. Mihály and P. Beauchêne, Solid State Commun. **63**, 911 (1987).

[17]G. Mihály, P. Beauchêne, and J. Marcus, unpublished.

[18]L. Mihály, K. B. Lee, and P. W. Stephens, Phys. Rev. B **36**, 1793 (1987).

[19]Z. Z. Wang and N. P. Ong, Phys. Rev. Lett. **58**, 2375 (1987).

[20]R. Kohlrausch, Ann. Phys. (Leipzig) **12**, 393 (1947).

[21]G. Kriza and G. Mihály, Phys. Rev. Lett. **56**, 2529 (1986).

[22]L. Mihály and G. Grüner, 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.

[23]G. Mihály, Phys. Rev. Lett. **60**, 470 (1988) (this issue).