Metastability of the Q Vector of Pinned Charge- and Spin-Density Waves

P. B. Littlewood and T. M. Rice[a]
Bell Laboratories, Murray Hill, New Jersey 07974
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The response of a one-dimensional charge-density wave weakly pinned by impurities to perturbations which change the Q vector is analyzed. From Monte Carlo calculations Q is found to lag behind the equilibrium value and the response is different for perturbations which move Q away from or towards equilibrium. Q changes by addition or removal of phase solitons and the metastability arises from the energy barriers for these processes. The behavior is similar to the pressure-induced hysteresis of Q observed in Cr.

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In an elegant set of experiments Ruesink, Perz, and Templeton[1] recently demonstrated that the $\vec{Q}$ vector of the spin-density-wave state in pure Cr can take on different values at low temperatures which depend on the history of the sample. When a hydrostatic pressure, P, was applied the $\vec{Q}$ vector changed linearly with increasing P with a value $d\ln Q/dP = -5\times10^{-7}$/bar but for decreasing P Q remained constant. The value of the $\vec{Q}$ vector at $P=0$ therefore depended on the maximum pressure reached during the cycle. This asymmetry between increasing and decreasing pressure was argued to be incompatible with an explanation based on impurity pinning.[1,2] In this Letter we show that this behavior is qualitatively similar to, and consistent with, that which arises from impurity pinning of an incommensurate density wave. We consider the simpler model of a charge-density wave (CDW) in one dimension weakly pinned by impurities. This model has been studied previously in some detail. In this Letter we analyze the response to a perturbation which changes the $\vec{Q}$ vector and show that if the CDW is a metastable state it responds differently to perturbations which drive it further from equilibrium than to those which drive it closer to equilibrium. Our assumption of an initial metastable state is based on the results of Fawcett, Griessen, and Vettier[3] who found that the $\vec{Q}$ vector in the spin-density-wave (SDW) state of Cr was temperature independent below a temperature $T_0 \approx 80$ K but varied with T for $T > T_0$. From these results we conclude that T_0 marks the onset of metastability and for $T < T_0$ the $\vec{Q}$ vector of the SDW does not have its equilibrium value. At $T < T_0$ then increasing P drives the $\vec{Q}$ vector further from its equilibrium value.

Consider a one-dimensional CDW in the presence of impurities which couple directly to the charge. Following Fukuyama and Lee[4] we ignore amplitude fluctuations and the energy has the form

$$E = \int dx\{\tfrac{1}{2}K[\nabla\varphi(x) - q]^2 + V\textstyle\sum_i \cos[Qx + \varphi(x)]\delta(x - R_i)\}, \quad (1)$$

where $\{R_i\}$ denote impurity positions, $\varphi(x)$ the phase (or displacement) of the CDW, V the impurity potential, and K the elastic constant. The case $q \equiv 0$ was considered by Fukuyama and Lee[4] who showed that the dimensionless parameter $\epsilon (= V/cK$; c, impurity concentration) determines the boundary between weak pinning ($\epsilon \ll 1$) and strong pinning ($\epsilon \gg 1$). In the weak-pinning regime $\varphi(x)$ varies on a length scale $\xi\ [= (2/\epsilon)^{2/3}c^{-1}]$ which is much larger than the impurity separation, c^{-1}, a result confirmed by the numerical work of Teranishi and Kubo.[5]

The choice $q \neq 0$ corresponds to a perturbation which changes the equilibrium value of Q to $Q+q$. The equilibrium state that results is obtained by replacing $\varphi(x)$ by $\bar{\varphi}_q(x) = qx + \varphi(x)$ and the form obtained for $\bar{\varphi}_q(x)$ is similar to that of $\varphi(x)$ when $q \equiv 0$. The length ξ and the binding energy of the CDW to the impurities, E_G, are independent of q. However, to pass from the equilibrium state $\varphi_{q=0}(x)$ to the state $\bar{\varphi}_q(x)$ requires a continuous deformation of the phase and there is an energy barrier between the two equilibrium states. A simple mixing of the two solutions gives a barrier proportional to E_G. To obtain a finite barrier one must make only local changes of the phase. To explore these changes we have made a series of Monte Carlo calculations in which the value of q was varied.

It is convenient to express the energy in terms

of the values of phases φ_i $[\equiv\varphi(R_i)]$, leading to

$$E = V\sum_i\{(\varphi_{i+1}-\varphi_i - qr_i)^2/2\epsilon r_i + \cos(QR_i+\varphi_i)\}, \quad (2)$$

where $r_i = (R_{i+1}-R_i)c$ and q is measured in units of c^{-1}.

It is in fact possible to obtain solutions to (2) by direct integration from one end,[6] but this procedure is highly unstable with respect to a small change in the boundary conditions.[6] We have made numerical calculations on a chain of 200 randomly distributed impurities with $\epsilon = 0.1$ and $Q/c = 100$. It was not possible to use smaller values of ϵ because of the large number of iterations required when q is changed.

In our zero-temperature Monte Carlo procedure we vary the phase φ_i at each site in turn to within a chosen accuracy $\Delta\varphi$ to minimize the energy. $\Delta\varphi$ is decreased after a stable state is reached to check for convergence. A typical run to determine the ground state took about 5000 iterations per impurity with a final $\Delta\varphi$ of 10^{-4} rad. The procedure is rapidly convergent, only small adjustments of the phase occurring after the first 500 iterations. The ground-state energy E_G was $-0.258V$ per impurity, which agrees well with the Fukuyama-Lee[4] formula $E_G = -0.75(\epsilon/2)^{1/3}V$. Interestingly, different initial configurations do not always iterate to the same final state. If the starting phases have a gradient such that $\varphi_{200} - \varphi_1 > 2\pi$, the final (metastable) solution has roughly the same average gradient and an energy $E > E_G$. However, all the metastable solutions we have found differ only *locally* from the ground state as they can be written as a series of 2π phase solitons superimposed on $\varphi_{g.s.}$. These metastable solutions play an important role when q is varied, which we now explore.

The parameter q was monotonically increased in intervals of 0.1 from $q = 0$ to 1.8 and at each value of q the $\{\varphi_i\}$ are varied one at a time, to lower the energy until convergence is obtained. In Fig. 1 we plot the phase gradient $\langle\nabla\varphi\rangle$ determined by the end values of the phases versus q. If the CDW stayed in equilibrium the value of $\langle\nabla\varphi\rangle$ should equal q as shown by the dashed line in Fig. 1. Instead $\langle\nabla\varphi\rangle$ lags behind substantially as shown by the line $OACEG$ in Fig. 1. Clearly these states are metastable. In the inset the energy is shown. The equilibrium ground-state energy is independent of q and the rise in the energy shows that the system is in a metastable excited state.

We have also made a series of calculations (the

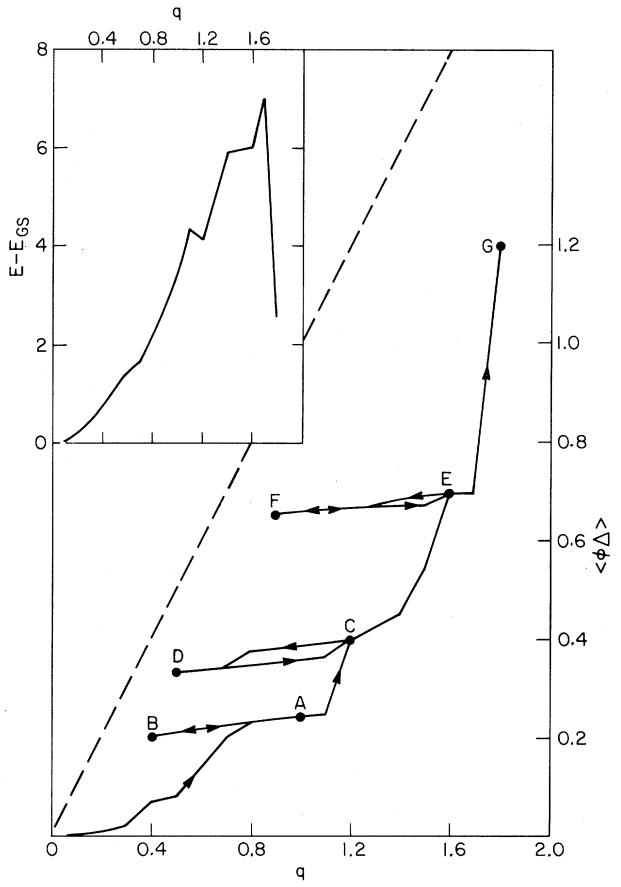

FIG. 1. The average phase gradient $\langle\nabla\varphi\rangle$ obtained from Monte Carlo calculations starting from the ground-state solution at $q = 0$. The arrows denote the direction in which q was changed, and the dashed line is the expected value of $\langle\nabla\varphi\rangle$ if the system remained in equilibrium. In the inset is plotted the energy per impurity (in volts) of the metastable solutions as q is increased monotonically from zero to 1.8.

portions of the line in Fig. 1 labeled AB, CD, and EF) to demonstrate the effect of cycling the driving wave vector q. For example, on increasing q from zero to 1.0 we follow the line OA. If q is then decreased to 0.4, $\langle\nabla\varphi\rangle$ moves along AB, with the value of $\langle\nabla\varphi\rangle$ nearly constant. The asymmetry between increasing or decreasing q, i.e., moving away from or towards equilibrium, is striking, and the value of $\langle\nabla\varphi\rangle$ depends on the highest value of q previously applied. Also on increasing q along BA $\langle\nabla\varphi\rangle$ remains almost constant until we have reached the highest value of q applied previously. Both these effects are similar to the effects of pressure cycling in Cr at low temperature.

To understand the origin of this hysteresis we have plotted out the change in the phase $\delta_q\varphi(x)$,

which occurs as q increases. In Figs. 2(a) and 2(b), $\delta_q \varphi(x)$ is plotted for a series of q values starting at $q = 0$ (equilibrium ground state) and increasing q to 0.7. In all cases $\delta_q \varphi(x)$ is the phase change measured from the starting equilibrium phase. Changes in the phase occur through the introduction at the sample ends of phase solitons with a phase change 2π. The involvement of solitons in changing the phase can be seen even more clearly in Fig. 2(c) which shows the phase over a short section of the chain. We begin by finding an equilibrium metastable state for $q = 1.7$ (the dashed line in the figure) and we then increase q to 1.8. The remaining curves are "snapshots" of $\delta_q \varphi(x)$ taken as the Monte Carlo solution progresses and show how solitons are launched across pinning barriers.

This behavior can be derived as follows. If we expand around the ground state $\varphi_0(x)$ of Eq. (1), and write $\varphi(x) = \varphi_0(x) + \psi(x)$, then we obtain

$$E - E_G = V\{\int dx (2\epsilon)^{-1}(\nabla\psi - q)^2 + \sum_i [\cos(QR_i + \varphi_0(R_i))\cos(\psi(R_i) - 1) - \sin(QR_i + \varphi_0(R_i))(\sin\psi(R_i) - \psi(R_i))]\}. \quad (3)$$

If we approximate the sums by averages (valid if $\nabla\psi \ll \xi^{-1}$) then

$$E - E_G \simeq V\int dx [(2\epsilon)^{-1}(\nabla\psi - q)^2 + (c\xi)^{-1/2}(1 - \cos\psi)]. \quad (4)$$

Equation (4) has the usual soliton solutions with width $\xi/\sqrt{2}$ and energy $8\sqrt{2}V/\epsilon\xi$. Since the width is $\sim\xi$ the continuum approximation is clearly approximate but is nonetheless a useful aid to understanding. For example, this approximation has barriers to introduction and removal of solitons at the ends. The barrier to introduction of a soliton vanishes only when $q = 2\sqrt{2}/\xi$ and to removal of a soliton vanishes only when $q = 0$. Thus there is hysteresis to be expected at such a transition in an ordered system—a result first derived by Frank and van der Merwe.[7] Clearly from Fig. 2 such a barrier at the ends is present in our disordered systems. There are additional barriers in the disordered system to the movement of the solitons due to fluctuations in the impurity potential. Therefore as solitons are introduced at the ends the repulsive interaction with the solitons already present acts to drive them through the impurity barriers (see Fig. 2). Our numerical results give a distribution of impurity barriers ranging from zero to $8\sqrt{2}V/\epsilon\xi$.

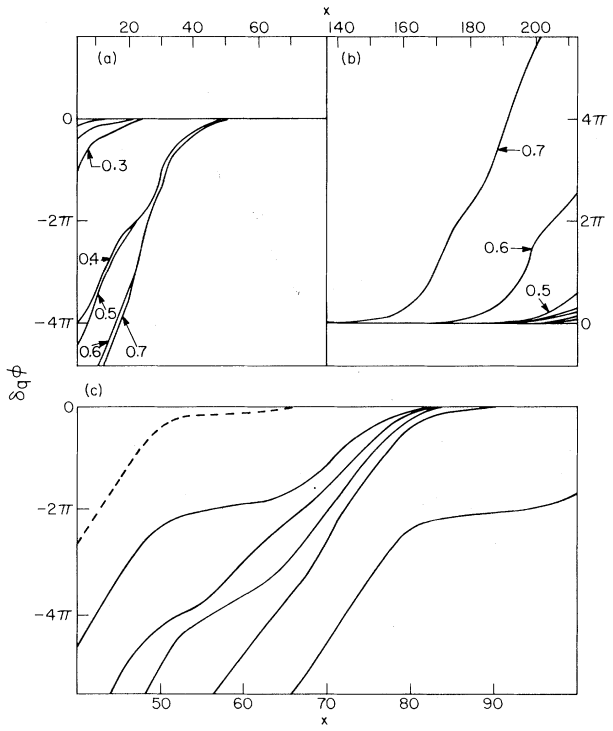

FIG. 2. The change in phase $\delta_q \varphi(x)$, measured from the equilibrium ground-state phase at $q = 0$, as q is changed. (a), (b) The phases of the metastable solutions at the two ends of the chain labeled by the values of q. (c) Progress of the solution with increasing Monte Carlo time, starting from the metastable solution for $q = 1.7$ (dashed curve) and increasing q to 1.8.

There are important differences between the one-dimensional CDW model and a three-dimensional SDW. First if the dominant impurity coupling is to a CDW harmonic[8] then the argument of the cosine term in Eq. (1) is multiplied by a factor of 2. The result will be that solitons will involve a phase change of π rather than 2π which reduces the soliton energy but does not change the qualitative results. In three dimensions the solitons are extended two-dimensional planar objects which have a transverse rigidity. They are still pinned at impurities but the pinning energy is reduced because of the transverse rigidity.[9] Nucleation of phase solitons at the ends must now proceed in a finite radius and is easier at a curved surface. However even in this case there are finite barriers to adding or removing solitons at equilibrium as in one dimension.

Our problem is directly analogous to the decay of supercurrents in superconductors through the introduction of phase slip. Homogeneous nucleation of phase-slip centers can occur via fluctuations in the magnitude of the order parameter,[10,11] but is very unlikely except at temperatures close to the transition temperature; however, heterogeneous nucleation at strong pinning sites is possible.

Whether or not these metastable states are long lived on a macroscopic time scale depends on a comparison of the pinning barrier with kT. In three dimensions, the pinning energy per domain is approximately $4\pi K\xi^2 = kT_1$ (Ref. 9) and for any reasonable choice of our parameters $T_1 \sim T_F \gg T_0$, which is the temperature at which a sharp onset of metastability was observed.[3] We suggest that T_0 makes a glass transition temperature below which the SDW is in a metastable state with a nonequilibrium value of Q, in which case the result $T_1 \gg T_0$ is expected. The probability of a local fluctuation in energy sufficient to depin a soliton is $\sim\exp(-T_1/T)$, which is vanishingly small. However, the fundamental attempt frequency is very large, so that on physical time scales depinning will be seen at $T \ll T_1$.

In summary we have shown that a one-dimensional model of impurity pinning has a behavior qualitatively similar to the hysteresis of the $\vec{Q}$ vector on pressure cycling in Cr. While the kinetic processes leading to phase slip may be different in one and three dimensions we believe that the general feature that such phase slip occurs through local nonlinear phase changes with barriers for the introduction, removal, and movement of the phase-slip centers persists between one and three dimensions.

[a]Present address: Theoretische Physik, ETH-Hönggerberg, CH-8093 Zurich, Switzerland.

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Spin-Lattice Relaxation via Phasons in a Multisoliton Lattice

R. Blinc, F. Milia,[a] V. Rutar, and S. Žumer

J. Stefan Institute and Department of Physics, E. Kardelj University of Ljubljana, 61001 Ljubljana, Yugoslavia

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The change in the phason spectrum on going from the "incommensurate plane wave" to the "multisoliton lattice" modulation limit has been observed via phason-induced ^{87}Rb spin-lattice relaxation in the incommensurate phase of Rb_2ZnCl_4. The observed temperature dependence of T_1^{-1} close to the "lock-in" transition reflects the temperature dependence of the "acousticlike" branch of the phason spectrum in a multisoliton lattice.

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Structurally incommensurate (I) systems are characterized by the appearance of a lattice distortion

$$u(z) = u_0 \cos\phi(z) \tag{1}$$

with a periodicity $\phi(z)$ which is an irrational fraction of the periodicity of the underlying lattice.[1] In the plane wave (IPW) modulation limit $\phi(z)$ is a linear function of the coordinate z in the direction of the modulation:

$$\phi(z) = q_s z + \phi_0, \quad \phi_0 = \text{const}, \tag{2}$$