Impurity pinning of discommensurations in charge-density waves

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The position of a discommensuration (DC) in a charge-density-wave state is pinned by impurities. This pinning gives rise to a barrier to the motion of a DC. If the pinning is weak the radius of curvature of the fluctuations in the position of the DC is large. Impurity pinning will enhance the hysteresis of a phase transition which requires the motion of DC, e.g., the stripe-hexagonal phase transition. As the temperature is raised, thermal fluctuations of the DC can cause a depinning transition of an isolated DC—a transition akin to the roughening transition of an interface. Depinning can also be achieved by external perturbations such as electric fields or inhomogeneous stresses.

I. INTRODUCTION

As the study of charge-density waves has evolved, the effect of the impurities has been a field of considerable interest. Some years ago, Sham and Patton,[1] and Imry and Ma[2] showed that impurities, which couple directly to the phase, or displacement, of the charge-density wave (CDW) destroy the long-range order of a CDW which is incommensurate with the underlying lattice and lead to an exponentially decaying correlation function. Experimentally, it is found that the range of the short-range order is quite large, indeed too large to be observed directly in conventional x-ray and neutron scattering. Fukuyama and Lee[3] in their analysis of the effect of impurities have given the criterion for the impurity pinning to be weak and the correlation length of the short-range order to be long. More detailed analyses of this problem have been made in one-dimensional systems.[4,5] As the boundary between the incommensurate and commensurate CDW is approached the nature of the problem changes. McMillan[6] has shown that as this boundary is approached the incommensurate CDW will consist of regions which are essentially commensurate separated by discommensurations (DC). These latter are narrow regions where the phase or displacement changes rapidly. It is the interaction of the DC with the impurities which is the subject of this work. This has been explored for one-dimensional systems by Fukuyama[7] and by Nakanishi.[8] Our interest will be in three-dimensional CDW where the DC are extended objects in two dimensions. In view of the ex-

perimental results in favor of weak pinning, we will concentrate on this regime.

The mathematical description of a DC within the phase-only approximation is that of a soliton. When the amplitude modulation of the DC is included the energy and interaction with neighboring DC is modified[9] but we shall assume that the impurity coupling is more important at low densities of DC. We will therefore only include phase modulation and the coupling of the impurity to the phase. For this coupling we take a local coupling between an impurity and the phase gradient of a CDW. The effective charge density in a DC varies as the phase gradient so that the model can be viewed as an interaction between the impurity and the charge density of the DC. We assume a short range for this interaction due to the screening by free carriers.

First in Sec. II we study the energy of a DC in one dimension in the presence of an impurity. Baeriswyl and Bishop[10] have shown that beyond a threshold value of the interaction strength the ground state has a DC at the impurity. Our interest here is in the interaction of a DC, which is present due to the incommensurability driving force and an impurity. We calculate the energy as a function of the separation and find the interaction energy has a range which is the width of DC. In Sec. III we examine the interaction with impurities in higher dimensions. For a three-dimensional CDW state the DC are extended objects in two dimensions. Individual DC are distorted by the presence of the impurities. The local radius of curvature of the DC is determined by balancing the

elastic forces against the interactions with the impurities. In Sec. IV we discuss the pinning at higher temperature where the entropy due to fluctuations in a DC can overcome the pinning by impurities and a transition, akin to the roughening transition of an interface,[11] occurs. Finally we discuss the response of a DC to perturbations and point out the possibility of observing the difference between DC pinned by impurities and DC which are thermally unpinned.

II. INTERACTION OF A DISCOMMENSURATION WITH AN IMPURITY IN ONE DIMENSION

We begin by discussing the interaction of a single discommensuration with an impurity in one dimension. We restrict our attention to the part of the free-energy density which depends on the phase, ϕ, of the order parameter only. The problem we wish to model is the interaction of a CDW with a charged impurity. Since the effective charge stored in a DC is related to the phase derivative[12] we take an interaction of the form[10]

$$F(\phi) = \int dx \left[\frac{1}{2} K \left(\frac{d\phi}{dx} \right)^2 + W(1 - \cos p\,\phi) - V(x) \frac{d\phi}{dx} \right] .$$

$$(2.1)$$

The first term represents the elastic energy associated with a deformation of the CDW, the second term the commensurate pinning potential of the background lattice, and the third term the electrostatic coupling of the effective charge stored in a DC or other phase disturbance to the impurity. In the last term we take a short-range potential, due to the screening by free carriers with a Gaussian form

$$V(x) = V_0 \exp(-x^2/2\sigma^2)/\pi^{1/2}\sigma .$$

$$(2.2)$$

The width, σ, generally is much shorter than the characteristic width of the DC, $\xi [= (K/p^2 W)^{1/2}]$, and for most purposes we use the simpler form

$$V(x) = V_0 \delta(x) .$$

$$(2.3)$$

We have chosen to take only the coupling of the impurity to the phase gradient rather than to the phase itself[3-5] for convenience. However both forms lead to a direct coupling between the impurity and the position of a DC,[8,10] which is all that matters in what follows. Taking the functional derivative of (2.1) with respect to $\phi(x)$ leads to

$$-K \frac{d^2\phi}{dx^2} + pW \sin p\,\phi + \frac{dV}{dx} = 0 .$$

$$(2.4)$$

The last term is singular in the limit $\sigma \to 0$. Integrating (2.4) through a small interval about the origin, $(-\epsilon, +\epsilon)$, the contribution of the second term is

negligible, and we obtain the result

$$\phi_{+\epsilon} - \phi_{-\epsilon} = V_0/K$$

$$(2.5)$$

with $\phi_{\pm\epsilon} \equiv \lim_{x\to 0\pm} \phi(x)$. The discontinuity in the phase at the impurity site is determined by the strength of the impurity potential V_0.

The binding energy of a DC is the difference between the energy of the DC at the origin and a DC placed well away from the impurity at the origin. Away from the origin the last term in (2.4) is negligible and the behavior of the phase is described by a soliton solution of the sine-Gordon equation (2.4). This has the form

$$\phi_s(x, X) = (4/p) \tan^{-1}[\exp(x - X)/\xi] .$$

$$(2.6)$$

The full solution of (2.4) is obtained by combining soliton solutions of the form (2.6) to satisfy the condition (2.5).

The ground state in which the DC is located at the origin is given by

$$\phi_G(x) = \begin{cases} \phi_s(x, X_G), & x < 0 \\ \phi_s(x, -X_G), & x > 0 , \end{cases}$$

$$(2.7)$$

where X_G is determined by

$$\phi_{-\epsilon} = \pi/p - V_0/2K = (4/p) \tan^{-1}[\exp(-X_G/\xi)] \quad (2.8)$$

and we have restricted our attention to the regime in which $\pi/p > V_0/2K$. A sketch of the phase is shown in Fig. 1(a). The energy of this solution, $E_{\text{g.s.}}$, can be broken into two parts

$$E_{\text{g.s.}} = 2 \int_{-\infty}^{-\epsilon} \left[\frac{1}{2} K \left(\frac{d\phi}{dx} \right)^2 \right.$$

$$\left. + W[1 - \cos p\,\phi(x)] \right] dx + \tilde{E}(\sigma) .$$

$$(2.9)$$

The second term represents the energy right at the origin and $\tilde{E}$ is singular as $\sigma \to 0$. However, it is common to all the configurational energies and cancels out of the binding energy.

Away from the origin, the last term in (2.4) is negligible so that we may multiply by $(d\phi/dx)$ and integrate to obtain

$$\left(\frac{d\phi}{dx} \right)^2 = \frac{2W}{K} (1 - \cos p\,\phi)$$

$$(2.10)$$

and when substituted in (2.9) we obtain

$$E_{\text{g.s.}} = 4W \int_0^{\phi_{-\epsilon}} \left[\frac{K}{2W} \right]^{1/2} (1 - \cos p\,\phi)^{1/2} d\phi + \tilde{E}(\sigma) ,$$

$$(2.11)$$

$$= 8(KW)^{1/2} p^{-1} [1 - \sin(pV_0/4K)] + \tilde{E}(\sigma) .$$
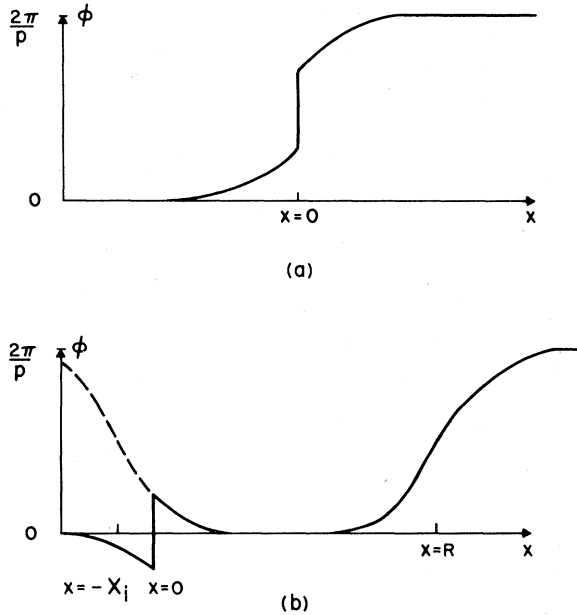
$$(2.12)$$

(a)

(b)

FIG. 1. (a) A discommensuration (DC) bound at an impurity site with a coupling which is proportional to the phase gradient of the charge-density wave. (b) The spatial dependence of the phase (solid line) when the DC is well separated from the impurity. The dashed line indicates the extrapolated DC, centered at $x = -X_i$.

When the separation R of the DC from the impurity is large, the phase has the form illustrated in Fig. 1(b). The phase at the origin is $\phi_{\pm\epsilon} = \pm V_0/2K$. The energy of this configuration in the limit $R \to \infty$, is

$$E_\infty = 8(KW)^{1/2}p^{-1}\{1 + [1 - \cos(pV_0/4K)]\} + \tilde{E}(\sigma) \ ,$$

$$(2.13)$$

where the first term is energy of an isolated DC. The last term is the energy associated with the phase variation near the origin.

The binding energy $-|E_B|$ is the energy difference between the two configurations;

$$-|E_B| = E_{\text{g.s.}} - E_\infty \ , \qquad (2.14)$$

$$= -8(KW)^{1/2}p^{-1}[1 + \sin(pV_0/4K)$$

$$- \cos(pV_0/4K)] \ . \qquad (2.15)$$

The form of the interaction between the impurity and the DC as function of the separation, R, can be obtained by examining the corrections to the two limits $R \to \infty$ and $R \to 0$. In the large-R limit the total energy $E(R)$ can be written as

$$E(R) = E_\infty + E_1(R) \ . \qquad (2.16)$$

The correction $E_1(R)$ is obtained by using $\phi(x) = \phi_s(x,R) + \phi_i(x)$ in (2.1), i.e., we superpose the two functions, $\phi_s(x,R)$, which describe the DC

at R, and $\phi_i(x)$. The latter is the functional form of the phase near the origin, given by

$$\phi_i(x) = -\phi_s(x,X_i), \quad x < 0$$

$$= \frac{2\pi}{p} - \phi_s(x,-X_i), \quad x > 0 \ , \qquad (2.17)$$

and X_i is determined by the relation

$$V_0/2K = (4/p) \tan^{-1}[\exp(-X_i/\xi)] \ . \qquad (2.18)$$

It is convenient to consider first the attractive interaction between the DC at R and the negative DC at $-X_i$ which results from extrapolating the form of $\phi_i(x)$ from $x > 0$ to $x < 0$. This extrapolation is shown as the dashed line in Fig. 1(b). This attractive interaction between two DC's is well known. The correction to this attractive interaction arises from the region $x < 0$. In this region $\phi_s(x,R) \ll 1$ and we can expand the total energy in powers of $\phi_s(x,R)$. It is straightforward to show that the correction from the region, $x < 0$ is $\sim\exp(-2R/\xi)$ and therefore smaller than the leading attractive interaction. As a result we obtain

$$E_1(R) = -32(KW)^{1/2}p^{-1}\exp(-R/\xi)$$

$$\times \tan(pV_0/8K) + O(\exp(-2R/\xi)) \ . \qquad (2.19)$$

In the opposite limit, as $R \to 0$, the displacement of the pinned soliton, changes the symmetric form (2.7) to an asymmetric form, $X_G \to X_G + R$ and $-X_G \to -X_G + R$. The result is a change in phases at the origin, $\phi_{\pm\epsilon}$, by an amount

$$\delta\phi = -4(R/\xi)[\cosh(X_G/\xi)]^{-1} \ . \qquad (2.20)$$

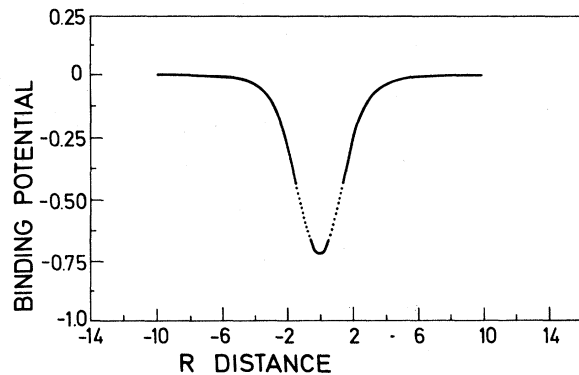The energy change that results in $E_{\text{g.s.}}$ is easily

FIG. 2. The potential energy of a DC interacting with an impurity which has a local coupling to the phase gradient. The energy is measured in units of the DC energy $[8p^{-1}(KW)^{1/2}]$ and the distance in the units of the DC width ($\xi = K^{1/2}/pW^{1/2}$). The solid lines are the large-R expansion (2.19) and small R (2.20). The dashed line is the interpolated form (2.21).

evaluated from (2.11) as $4(KW)^{1/2}p^{-1}$
$\times \sin(pV_0/4K)(\delta\phi)^2$.

Putting the two corrections together we obtain the interaction between an impurity and a DC as a function of the separation R for $R/\xi \ll 1$ and $R/\xi \gg 1$. As an example we show in Fig. 2 the behavior for a particular choice of the parameters. The attractive interaction is short range of width ξ and is exponentially damped at large distances. A useful parametrization is to take the form

$$E_1(R) = E_0/\cosh^2(\alpha R) \ . \tag{2.21}$$

This form gives a good interpolation between the large and small R limits as illustrated in Fig. 2. The bound states of this potential are known analytically.

III. INTERACTION OF A DISCOMMENSURATION WITH AN IMPURITY IN THREE DIMENSIONS

In the previous section we showed that an impurity can have a short-range attractive interaction with a DC. In higher dimensions the DC is a plane or a line which has a rigidity, and distortions in the position of the DC to maximize the energy gain from impurities will be opposed by the transverse rigidity of the DC. If we denote the position of the DC by $z_0(\vec{r})$, where $\vec{r}[\equiv(x,y)]$ is a vector perpendicular to the $\vec{Q}$ vector of the CDW, the elastic energy associated with such distortions can be written[13] as

$$E_{el} = \frac{1}{2} \int d\vec{r}\,dz K_\perp \left[\left(\frac{d\phi_s(z - z_0(\vec{r}))}{dx} \right)^2 \right.$$
$$\left. + \left(\frac{d\phi_s(z - z_0(\vec{r}))}{dy} \right)^2 \right] \tag{3.1}$$

$$= \frac{1}{2} \int dz K_\perp \left(\frac{d\phi_s}{dz} \right)^2 \int d\vec{r} \left(\frac{dz_0}{dx} \right)^2 + \left(\frac{dz_0}{dy} \right)^2 \tag{3.2}$$

$$\equiv \frac{1}{2}\tilde{K} \int d\vec{r} \left(\frac{dz_0}{dx} \right)^2 + \left(\frac{dz_0}{dy} \right)^2 \ , \tag{3.3}$$

where $\tilde{K} = 4K_0\xi^{-1}$.

The short-range impurity interaction obtained in Sec. II can be conveniently approximated by a square-well attractive potential. The model form of the total energy that results

$$E_T\{z_0\} = E_{el} - \tilde{V} \sum_i \Theta(z_0(\vec{r}_i) - z_i) \ , \tag{3.4}$$

where $\tilde{V} = |E_B|$ and $\Theta = 1$ if $|z| < \xi$ and 0 otherwise. The impurity positions $(\vec{r}_i, z_i)$ are assumed to be random.

The total energy $E_T\{z_0\}$ is similar to the problem of an incommensurate CDW in a single $\vec{Q}$ state interacting with impurities which couple directly to the

phase or displacement of the CDW. This problem has been considered by several authors.[3-5,14] An important distinction is whether the impurity energy is much larger or smaller than the elastic energy associated with the distortions. The former case is the strong pinning limit in which the DC will distort to maximize its overlap with individual impurity potentials. The latter case is the peak pinning limit in which the radius of curvature of the DC remains large compared to ξ. We shall concentrate on this weak-pinning limit and make only a few remarks concerning the former limit.

In the weak-pinning limit (which can be either a stiff DC or a high impurity concentration), the distortions of the DC will possess a length scale L, a typical domain size over which the DC position $z_0(\vec{r})$ is nearly constant. Within a given domain, the average number of impurities interacting with the DC will be

$$\bar{N} = 2cL^2\xi \ , \tag{3.5}$$

where c is the impurity concentration. We shall see that the weak pinning limit corresponds to $\bar{N} \gg 1$, and in consequence the only contribution to the impurity pinning energy of a DC can come from statistical fluctuations in this number, which will be of order $\bar{N}^{1/2}$.

The amplitude of the DC distortion in the direction parallel to the $\vec{Q}$ vector of the CDW is characterized by a second length scale l. The larger the value of l, the more the DC can take advantage of the fluctuations in impurity density, but of course the greater is the penalty from the elastic energy (3.3). The values of the two length scales can now be estimated by minimizing the total energy with respect to the choice of l and L.

Bearing in mind the DC thickness 2ξ, we can make roughly $(l/2\xi)$ independent choices of the position of z_0 for the DC in each domain. The largest fluctuation in the number of impurities is expected to be approximately

$$\Delta N_{max} \simeq [\bar{N} \ln(l/2\xi)]^{1/2} \tag{3.6}$$

assuming a Gaussian distribution for ΔN. {Equation (3.6) is only is only strictly true if (l/ξ) is large which is not in fact the case [see Eq. (3.9)] but this will suffice to give us a rough estimate of l.} Hence the impurity pinning energy per unit area will be

$$E_{imp} \simeq -\tilde{V}\Delta N_{max}/L^2 \ . \tag{3.7}$$

Connecting the nearby domains together smoothly gives rise to an elastic energy

$$E_{el} \simeq \frac{1}{4}\tilde{K}(l/L)^2 \ . \tag{3.8}$$

Minimizing (3.7) and (3.8) with respect to l and L we find

$$(l/\xi) = U = 2e^{1/2} \tag{3.9}$$

and

$$L = \frac{1}{2}(U^2 \tilde{K}/\tilde{V})(\xi^3/2c)^{1/2} \; . \tag{3.10}$$

We see that the amplitude of the distortion is of the same order as the thickness of the DC, and that the average number of impurities in a domain is

$$\bar{N} = 2cL^2\xi = (U^2 \tilde{K} \xi^2 / 2\tilde{V})^2 \tag{3.11}$$

so that in the weak pinning limit ($\tilde{K}\xi^2 >> \tilde{V}$) we have $\tilde{N} >> 1$ as we suggested above.

Substituting (3.10) and (3.11) in (3.7) gives the impurity pinning energy per unit area

$$E_{\text{imp}} = -\frac{bc\tilde{V}^2}{\xi\tilde{K}} \; , \tag{3.12}$$

where the numerical constant $b = 8 \ln^{1/2}(U/2) U^{-2}$.

IV. ROUGHENING TRANSITION OF A DISCOMMENSURATION AT A FINITE TEMPERATURE

At finite temperature the thermal vibrations of the DC cause the pinning potential to be reduced. Eventually if the amplitudes of the thermal vibration are large enough the barrier against the motion of the discommensuration is reduced to effectively zero. At this point an isolated DC will have very large values of mean-square vibration amplitude or in other words the DC will be rough. At low temperatures however the DC is pinned in a definite position by the impurities. It is the transition between these two regimes—the roughening transition of an isolated DC which is the subject of this section. It is similar to the roughening transition of a discrete interface.[11]

The self-consistent harmonic phonon approximation was used by Okabe and Fukuyama[15] to describe the temperature dependence of the pinning in one dimension and it can be generalized to apply to the two-dimensional problem at hand. The DC at low temperature is pinned and energy spectrum of the small amplitude oscillations around the pinning position is given by

$$\omega^2(q) = \gamma_0^2 + v^2 q^2 \; , \tag{4.1}$$

where q is a wave vector in the reciprocal space (q_x, q_y). The velocity $v = (\tilde{K}/m)^{1/2}$ where m is the mass density of DC for motion in the z direction. The long-wavelength pinning frequency, γ_0, is determined by the average pinning potential energy (3.12) and the mass density, m;

$$\gamma_0^2 = \frac{bc\tilde{V}^2}{m\xi^3 \tilde{K}} \; . \tag{4.2}$$

The key assumption in the self-consistent phonon approximation[15] is that $\tilde{V}$ at finite temperature should

be replaced by

$$\tilde{V}(T) = \tilde{V}\exp[-\langle z^2(T)\rangle/2\xi^2] \; , \tag{4.3}$$

where $\langle z^2(T)\rangle$ is the mean-square amplitude of vibration. Using the well-known relation

$$\langle z^2(T)\rangle = \sum_q [n(\omega_q) + \frac{1}{2}](m\omega_q)^{-1} \; , \tag{4.4}$$

where $n(\omega_q)$ is the Bose function. Substituting (4.3) in (4.2) we obtain a self-consistent equation for the energy gap, $\gamma(T)$

$$\frac{\gamma^2(T)}{\gamma_0^2} = \exp\left[\frac{-z^2(T)}{\xi^2}\right] \; . \tag{4.5}$$

At high temperatures, $T >> \gamma$, the high-temperature limit can be taken for the Bose function (4.4) and we obtain

$$z^2(T) = \frac{T}{2\pi m} \int_0^Q q\,dq\,(\gamma^2 + v^2 q^2)^{-1} \tag{4.6}$$

$$= \frac{-T}{4\pi m v^2}\left[\ln\left(\frac{\gamma^2}{\gamma_0^2}\right) + \ln\left(\frac{\gamma_0^2}{\gamma^2 + v^2 Q^2}\right)\right] \; , \tag{4.7}$$

where the upper cutoff Q is of order the dimensions of the Brillouin zone. The self-consistent equation (4.5) is then reduced to the form

$$\frac{\gamma^2(T)}{\gamma_0^2} = A\left[\frac{\gamma^2(T)}{\gamma_0^2}\right]^\alpha \; , \tag{4.8}$$

with $A = [\gamma_0^2/(\gamma^2 + v^2 Q^2)]^\alpha << 1$ and

$$\alpha = \frac{T}{4\pi m v^2 \xi^2} = \frac{T}{4\pi\tilde{K}\xi^2} \; . \tag{4.9}$$

A self-consistent solution of Eq.(4.8) with $\gamma < \gamma_0$ only exists if the exponent $\alpha \lesssim 1$. The boundary $\alpha = 1$ determines a roughening temperature, T_r, given by

$$T_r = 4\pi\tilde{K}\xi^2 \; . \tag{4.10}$$

This is the temperature at which $\lim_{T \to T_r} z^2(T) \to \infty$.

This form of the roughening temperature is independent of the strength of the impurity potential, $\tilde{V}$, and depends only on the elastic coefficient. This form can be compared to the results of a diagrammatic analysis by Chui[16] and of renormalization-group calculations.[17] These results give the same criterion, (4.10), for the temperature at which a random-symmetry breaking field becomes irrelevant in a planar model. In the self-consistent harmonic phonon approximation the two problems—the positional pinning of a DC by impurities and the planar model in the presence of a random-symmetry breaking field—are identical and the above analysis carries through for the latter problem and gives the same answer as the renormalization-group analysis. The

transition temperature for the random problem is a factor of 2 lower than for an ordered symmetry breaking field. In the self-consistent harmonic phonon approximation this arises because the pinning frequency, $\gamma_0 \propto V^{1/2}$ where V is the strength of ordered symmetry breaking field, rather than the form $\gamma_0 \propto \tilde{V}$ in (4.2). As a result a factor of 2 carries through to give a transition temperature a factor of 2 higher than (4.10) in agreement with the results of the renormalization-group analysis.[18]

If $\tilde{V} \equiv 0$, the integral in (4.6) diverges at all temperatures and the DC is rough at all temperatures. If instead of the criterion for the roughening transition which occurs as $z \to \infty$, we take a criterion that $\langle z(T_r^1)^2 \rangle = \xi^2$ then T_r^1 depends on the strength and number of impurities. At this value of $\langle z^2 \rangle$ the effective activation barrier to move the DC will be small. From (4.7) we obtain

$$T_r^1 = T_r/[1 + 2\ln(vQ/\gamma_0)] \quad . \tag{4.11}$$

The reduction factor of T_r^1/T_r varies only logarithmically in the impurity strength and concentration so that even for low concentrations and weak impurity the temperature T_r^1 will still be quite high.

The above considerations apply to an isolated DC. In an incommensurate state, just above the commensurate-incommensurate transition there is a finite density of DC which are separated by an average length, Λ. In the limit that the commensurate transition is approached, $\Lambda \to \infty$, and the interaction between the DC falls off exponentially. Therefore in the limit of large Λ the pinning by impurities will always be more important than the interaction between DC. A simple criterion which will determine the region in which the impurities dominate is when the force on a DC from neighboring DC is not large enough to dislodge it from a pinned position. The interaction energy density of a DC with its neighbor at a distance Λ is similar to (2.19) and is given by

$$E_{\text{int}}(\Lambda) = 32(KW)^{1/2}p^{-1}\exp(-\Lambda/\xi) \quad . \tag{4.12}$$

Therefore, equating the change in the energy $E_{\text{int}}(\Lambda)$ on going from $\Lambda \to \Lambda - l$, with the pinning energy density gives a criterion

$$32(KW)^{1/2}p^{-1}\exp(-\Lambda/\xi)[\exp(l/\xi) - 1] < bc\tilde{V}^2/\tilde{K}\xi \tag{4.13}$$

for the region where the impurity pinning will dominate. The impurity-induced fluctuations in the curvature of the DC will affect the interaction between them. The average interaction energy of a roughened DC will be approximately

$$E_{\text{DC-DC}} \simeq 32(KW)^{1/2}p^{-1}\exp(-\Lambda/\xi)[1 + \tfrac{1}{2}\langle(z - \Lambda)^2\rangle/\xi^2] \simeq 32(KW)^{1/2}p^{-1}\exp(-\Lambda/\xi)(1 + \tfrac{1}{4}l^2/\xi^2) \quad . \tag{4.14}$$

The effect of including this extra term in the total energy is easily seen to be a small reduction in the deformation amplitude l, as might have been anticipated.

Yet another source of pinning is due to the discrete nature of the underlying lattice. This pinning of a DC is analogous to the Peierls force which pins the position of a real dislocation in a lattice.[19] The strength of this pinning should be similar to the Peierls pinning and $\propto \exp(-\xi/a_0)$ where ξ is the thickness of a DC and a_0 is the lattice constant. Therefore such pinning will be much smaller than impurity pinning in most cases.

V. PERTURBATIONS WHICH COUPLE DIRECTLY TO THE POSITION OF THE DISCOMMENSURATIONS

Finally we consider various perturbations which couple directly to the position of the DC. The observation of a threshold value for the motion of a DC in response to an applied perturbation would confirm the pinning by impurities.

The first example is an electric field. The charge density per unit area associated with a DC varies with the magnitude of the phase change[12] across the DC $(2\pi p^{-1})$. This leads to a coupling to an external field, E_Z, of the form

$$E_f = -2\pi p^{-1} e^* E_Z z \quad , \tag{5.1}$$

where the effective charge e^* is temperature and band-structure dependent.[20]

A second perturbation is one which couples to the value of the $\bar{Q}$ vector. An example is an external stress. First we consider a single-$\bar{Q}$ state. The free-energy functional that describes the coupling to the strain field $\eta(x)$ following Bak and Timonen[21] can be written

$$F_S\{\phi, P\} = \int dx \left[\tfrac{1}{2}K\left(\frac{d\phi}{dx} - \delta_0\right)^2 - \tfrac{1}{2}\delta_0^2 + W(1 - \cos p\phi) \right.$$

$$+ G\eta(x)\frac{d\phi}{dx}(x) + \tfrac{1}{2}C\eta^2(x)$$

$$\left. + \frac{D}{2}\left[\frac{d\eta(x)}{dx}\right]^2 - P\eta \right] \quad . \tag{5.2}$$

In this expression the fourth term is the local coupling between the strain and phase gradient. The fifth and sixth terms are the elastic and higher-order energies and the last term is the external pressure. This free energy functional has been studied by several groups, most recently, comprehensively by

Brocksch, Everts, and Müller-Krumbhaar.[22] Of interest to us here, is the question of the role of an external pressure P. If we write

$$\eta(x) = \eta_0 + \tilde{\eta}(x) \ , \tag{5.3}$$

where $\tilde{\eta}(x)$ is localized at the DC and η_0 is a constant strain caused by the external pressure, minimizing with respect to η_0 leads to

$$\eta_0 = P/C \tag{5.4}$$

and when substituted back in (5.2) this gives

$$F_S\{\phi,P\} = \int dx \left[\frac{1}{2}K \left(\frac{d\phi}{dx} - \delta_1 \right)^2 - \frac{1}{2}\delta_0^2 - \frac{P^2}{2C} \right. $$
$$+ W(1-\cos p\phi) + G\tilde{\eta}(x)\frac{d\phi(x)}{dx}$$
$$\left. + \frac{1}{2}C\tilde{\eta}^2(x) + \frac{D}{2}\left[\frac{d\tilde{\eta}}{dx}\right]^2 \right] \ . \tag{5.5}$$

The effect of the pressure is to shift the driving term due to the q-vector mismatch from δ_0 to

$$\delta_1 = \delta_0 - GK^{-1}\eta_0 = \delta_0 - GP/CK \ . \tag{5.6}$$

This acts to shift the chemical potential, or energy per unit area to add a DC

$$\epsilon = \epsilon^{(0)} + 2\pi GP/C \tag{5.7}$$

where $\epsilon^{(0)}$ is the value at $P=0$. If a uniform pressure or uniaxial stress is applied, the shift in the $\vec{Q}$ vector will shift the equilibrium spacing between the DC. The force on an individual DC will come from the interaction with the neighboring DC in the way discussed above. The energy change arises only through the change in δ and is no different from other perturbations, such as temperature which also acts to change δ.

A nonuniform strain, however, acts directly on the DC. The energy of an isolated DC now depends directly on its location through the dependence of δ on position. This leads to a term, $\propto 2\pi p^{-1} G\eta_0(x)$, in the energy of an isolated DC. The energy ϵ is position dependent and there will be a direct force on an isolated DC to move equal to the gradient of this energy. Thus a strain gradient acts in a similar fashion to an electric field to move an isolated DC.

In $2H$-TaSe$_2$, the CDW state has three coexisting CDW with the $\vec{Q}$ vectors lying along the directions of the three shortest basal-plane reciprocal-lattice vectors close to $\{\frac{1}{3}\vec{G}\}$. As the temperature is lowered the $\vec{Q}$ vectors approach and finally lock in to a commensurate phase with $\vec{Q} \equiv \frac{1}{3}\vec{G}$.[23] There are three orientations of the DC and experiment[24] shows that they lie parallel to the three $\vec{Q}$ vectors. As the commensurate phase is approached, a two-dimensional

pattern of DC develops. Bak et al.[25] have shown that there are two possible patterns. One is a honeycomb array of DC which preserves the hexagonal symmetry and the second is a stripe array of parallel DC. The repulsive interaction energy between the DC is lower in the hexagonal phase but in this phase there are lines of intersection of the DC which require either a positive or negative energy. Bak et al.[25] have shown that it is the sign of this intersection energy which is crucial since the number of intersections scales algebraically with the separation of DC. If the energy is repulsive then the stripe phase is the lowest-energy phase at the onset of incommensurability and the sequence of phases is commensurate, stripe, and hexagonal incommensurate. This sequence has been observed recently in $2H$-TaSe$_2$ at ambient pressure by Fleming et al.[24] When the temperature was lowered, the stripe phase was not observed. The region of hysteresis extends over a temperature interval ≈ 20 K. This hysteresis region however is greatly narrowed in the presence of small nonhydrostatic stresses.[26]

It is therefore of interest to examine the effect of a uniaxial stress on the DC in $2H$-TaSe$_2$. There are three $\vec{Q}$ vectors and three orientations of the DC in this compound. These are illustrated in Fig. 3. The relevant term in the energy as shown above is the elastic energy—the first term in (5.2). This elastic energy can be written as a sum of phase gradients for each of the three $\vec{Q}$ vectors; integrated over the basal plane

$$F_{\text{el}}\{\phi,S\} = \sum_i \int dx^2 \frac{1}{2}K_1[\vec{m}_1^{(i)} \cdot (\vec{\nabla}\phi_i - \vec{q}^{(i)})]^2$$
$$+ \frac{1}{2}K_2[\vec{m}_2^{(i)} \cdot (\vec{\nabla}\phi_i - \vec{q}^{(i)})]^2 \ . \tag{5.8}$$

The phases $\phi_i (i=1,3)$ are associated with the three CDW and the elastic energy is minimized by the choice of $\vec{Q}$ vectors which are $\vec{q}^{(i)}$ away from the commensurate positions, $\vec{G}_i/3$. The two-dimensional vectors $\vec{m}_j^{(i)}$ are the principal directions for the expansion around the minima. In the absence of an external stress the system is hexagonal and vectors $\vec{q}^{(i)}$ lie along the directions determined by the reciprocal-lattice vectors $\{\vec{G}\}$. If we denote by $\vec{n}_1^{(i)}$ unit vectors along these directions and $\vec{n}_2^{(i)}$ unit vectors perpendicular (see Fig. 3) then the vectors $\vec{m}_j^{(i)} \equiv \vec{n}_j^{(i)}$ all (i,j) in the absence of stress. In the presence of a uniaxial stress $\vec{S}$ applied along a direction $\|\vec{G}$, the vectors $\vec{q}^{(i)}$ and $\vec{m}_j^{(i)}$ change. If $\vec{S} \| \vec{n}_1^{(i)}$ then

$$\vec{q}^{(1)}(\vec{S}) = (q_0 + \delta_1)\vec{n}_1^{(1)}; \quad \vec{m}_j^{(1)} = \vec{n}_j^{(1)} j = 1,2 \ , \tag{5.9}$$

with $\delta_1 \propto S$. The other vectors however may rotate under the stress and

$$\vec{q}^{(2)}(\vec{S}) = (q_0 + \delta_2)\vec{n}_1^{(2)} + \delta_3\vec{n}_2^{(2)} \ ;$$
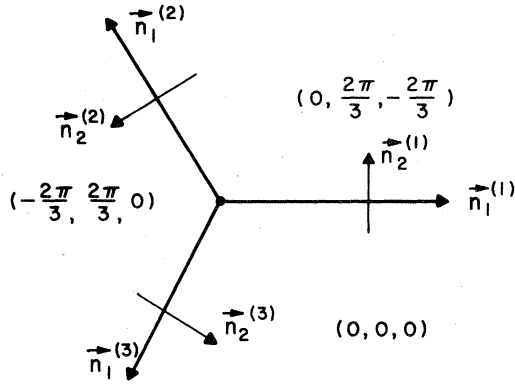$$\vec{q}^{(3)}(\vec{S}) = (q_0 + \delta_2)\vec{n}_1^{(3)} - \delta_3\vec{n}_2^{(3)} \ , \tag{5.10}$$

FIG. 3. The three orientations of the three lines of DC in $2H$-TaSe$_2$. The vectors $\vec{n}_1^{(i)}$, $\vec{n}_2^{(i)}$ are shown. Also the phase change across each DC is obtained by subtracting the phases (ϕ_1, ϕ_2, ϕ_3) of each of the CDW.

where $\vec{q}^{(2)}$ and $\vec{q}^{(3)}$ are related under reflection about the x axis. The principal directions are given by

$$\vec{m}_1^{(2)} = \vec{n}_1^{(2)} - s n_2^{(2)}; \quad \vec{m}_2^{(2)} = \vec{n}_2^{(2)} + s \vec{n}_1^{(2)},$$
$$\vec{m}_1^{(3)} = \vec{n}_1^{(3)} + s \vec{n}_2^{(3)}; \quad \vec{m}_2^{(3)} = \vec{n}_1^{(3)} - s \vec{n}_2^{(3)}, \quad (5.11)$$

where $s \propto S$. Again we have made use of reflection symmetry about the x axis to relate $\vec{m}_i^{(2)}$ and $\vec{m}_i^{(3)}$. Consider a type-I DC whose elastic energy ϵ_1 is given by

$$\epsilon_1 = \frac{1}{2} \sum_{i=2,3} \int d_x^2 \sum_{j=1,2} K_j (\vec{m}_j^{(i)} \cdot \vec{\nabla} \phi_i)^2$$
$$- 2(\vec{m}_j^{(i)} \cdot \vec{q}^{(i)})(\vec{m}_j^{(i)} \cdot \vec{\nabla} \phi_i) \ . \quad (5.12)$$

These terms can be evaluated to lowest order in S

$$\epsilon_1 = \frac{1}{2} \int d_x^2 \left[\frac{3}{4} K_1 + \frac{1}{4} K_2 + \frac{\sqrt{3}}{2} s (K_1 - K_2) \right]$$
$$\times \left[\left(\frac{d\phi_2}{dy} \right)^2 + \left(\frac{d\phi_3}{dy} \right)^2 \right]$$
$$- K_1 \left(\frac{\sqrt{3}}{2} + \frac{s}{2} \right) \left(\frac{d\phi_2}{dy} - \frac{d\phi_3}{dy} \right) (q_0 + \delta_2)$$
$$- K_2 (-\frac{1}{2}) \left(\frac{d\phi_2}{dy} - \frac{d\phi_3}{dy} \right) (\delta_3 + sq_0) \quad (5.13)$$

and this reduces to an elastic energy per unit area for the DC

$$\epsilon_1 = [3K_1 + K_2 + 2\sqrt{3} s (K_1 - K_2)] \xi^{-1}$$
$$- \frac{2\pi}{\sqrt{3}} K_1 \left[q_0 + \frac{sq_0}{\sqrt{3}} + \delta_2 - (\delta_3 + sq_0) \frac{K_2}{\sqrt{3} K_1} \right] \ . $$
$$(5.14)$$

Similarly we find after some algebra

$$\epsilon_2 = \epsilon_3 = [3K_1 + K_2 - \sqrt{3} s (K_1 - K_2)] \xi^{-1}$$
$$- \frac{2\pi K_1}{\sqrt{3}} \left[q_0 - \frac{sq_0}{2\sqrt{3}} + \frac{1}{2} (\delta_1 + \delta_2) \right.$$
$$\left. + \frac{1}{2} (\delta_3 + sq_0) \frac{K_2}{\sqrt{3} K_1} \right] \ . \quad (5.15)$$

The result is a splitting, linear in the strain, of the energy of type-I DC, which is parallel to a uniaxial strain direction, from the other two types. This splitting was derived within the generalization of the strain-dependent free energy of Bak and Timonen[21] in which the coupling occurs through the elastic energy terms. In principal the commensurability terms can also be different between the type-I DC and the types II and III modifying the energy splitting.

The consequence of the linear splitting is to favor the stripe phase over the hexagonal phase. By suitably orienting the DC or stripes in the stripe phase, the energy is lowered relative to the hexagonal phase which has equal densities of all three DC. The energy difference is linear in the uniaxial strain S and proportional to the DC density. As such it can dominate close to the commensurate-incommensurate transition and cause the I phase to be a stripe phase at its onset irrespective of the sign of the intersection energy.

A nonuniform uniaxial strain can have a large effect on the hysteresis associated with stripe-hexagonal phase transition. Individual DC experience a force proportional to the strain gradient. If the force is large enough at $T < T_r$, the DC will move. For general strain patterns, the different DC can be driven in different directions, towards or away from the regions of largest strain. As a result such inhomogeneous strains can act to greatly reduce the hysteresis at the stripe-hexagonal transition.

VI. CONCLUSIONS

In this paper we have discussed the effects of impurities in the host crystal on dilute concentrations of DC. The relevance of our results to experimental systems will depend on the values of the various parameters which enter the theory. Unfortunately we do not have good *a priori* values of these parameters and we must depend on experiment as a guide.

The system which has been most studied experimentally is $2H$-TaSe$_2$. The scattering experiments[23,24] were briefly reviewed in the previous section. These experiments and recent NMR experiments support the existence of an array of parallel DC in the striped phase of $2H$-TaSe$_2$ (Ref. 24) which occurs on warming in the temperature interval $90 \leq T \leq 110$ K. In this region the period of the stripes, and therefore

the spacing between DC, rises rapidly initially and then is essentially constant. Since the separation between DC is of order 300 Å it is not *a priori* obvious which side of the criterion (4.13) is obeyed. Several years ago, Barmatz and collaborators[27] studied the low-frequency ($\sim$3 kHz) vibration of a plate of $2H$-TaSe$_2$. In these vibrations a nonuniform stress is set up and as discussed in Sec. V this will drive a motion of the DC relative to the host crystal. However, Barmatz *et al.*[27] did not observe an extra damping in this temperature interval—a result which indicates that the DC array is pinned to the lattice and moves rigidly with it at these low frequencies. There is no evidence for a change in the damping with temperature and one is forced to conclude that the pinning frequency of the array of DC is much higher than the driving frequency ($\sim$3 kHz).

Surprisingly Barmatz *et al.*[27] observed a large temperature-dependent damping at $T \leq 90$ K — the region in which Fleming *et al.*[24] observe only the commensurate structure by x-ray scattering. An explanation is that there remains a small but finite density of DC which are now behaving as isolated DC and as a result the thermal vibrations are large. The average pinning frequency $\gamma(T)$ drops exponentially with increasing temperature [see (4.5)] which causes an exponential rise in the damping at low frequencies. This hypothesis explains the strong temperature

rise in damping that Barmatz *et al.* observed in the commensurate phase. There is a sudden disappearance in the damping at $T \approx 90$ K on heating, and at the same temperature Fleming *et al.* observe the $\vec{Q}$ vector move away from the commensurate value. This could signal the crossover transition from isolated DC to an array of DC with a simultaneous reduction in $\langle z^2 \rangle$ and a rise in the pinning frequency. These suggestions can be tested, hopefully, by electron microscope and high-resolution x-ray experiments.

In conclusion, we have examined the pinning of individual DC by impurities and discussed the temperature dependence of the pinning and the possibility of thermal "depinning." Further experiments on $2H$-TaSe$_2$ to test our results are suggested.

ACKNOWLEDGMENTS

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