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Dislocation and impurity effects in smectic-A liquid crystals*

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The effects of dislocations and impurities on the macroscopic elastic properties of smectic-A liquid crystals are discussed. The first conclusion is that smectics behave like linear elastic media only so long as the stresses are smaller than some critical value that is analogous to the critical velocity of a superfluid. Below the critical stress, smectics can store elastic energy without flowing and consequently without any dissipative processes in analogy with the fact that, below a critical velocity, superfluids store kinetic energy without any dissipation. For most practical samples the critical smectic stress is that value for which pinned dislocation will grow unstable; however, for ideal samples, initially free of dislocations, the critical value is determined by the condition of unstable growth of thermally generated dislocation loops. In the linear elastic region both dislocations and impurities modify the macroscopic elastic properties such that the effective elastic constant is smaller than the value for an ideal sample. This is a sort of diaelasticity and can be discussed in the same way as diamagnetism. Impurities are shown to act as sources of stress fields analogous to the way magnetic dipoles and magnetic monopoles are sources of magnetic fields. The result is to predict long-range elastic interactions between impurities in smectic systems. Since biological systems like chloroplasts and retinal rods have lamellarlike structures that are similar to the smectic structure, there is the possibility that long-range elastic interactions may play some role in biological function.

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I. INTRODUCTION

In a previous paper Pershan developed theoretical expressions for the stress-strain fields surrounding simple dislocations in smectic-A liquid crystals and for the forces on dislocations due to macroscopic stresses in the material.[1] These results were conveniently discussed in terms of the Peach-Koehler analogy between the magnetic forces on electric currents and the stress-induced forces on dislocation lines.[2] With this analogy in mind the suggestion was made that dislocations in smectics might have effects on the stress fields in smectics analogous to the effects that electric currents in metals have on applied magnetic fields. That is, one might expect a "diaelasticity" in smectics that is analogous to the diamagnetism of metals. The purpose of this paper is to develop these ideas further, presenting some theoretical predictions for possible effects.

If dislocation lines can be considered as analogous to electric current lines, then dislocation loops can be treated analogously to electric current loops which are formally equivalent to magnetic dipoles. Thus, the initial point is that for the purposes of discussing macroscopic stress-strain relations in smectics, it will be more convenient to deal with dislocation moment densities rather than the dislocation densities. This is analogous to the macroscopic magnetic properties of solids in which case one defines the magnetization in terms of the first moment of the microscopic electron currents.

The second point is that certain types of impurities induce stress-strain fields that are identical in the far field with those of a dislocation loop. Thus, if dislocation lines are analogous to the orbital motions of free electrons, these impurities are analogous to the permanent magnetic moments of bound electrons (i.e., paramagnetic ions) or electron spins. The elastic properties of smectic liquid crystals are influenced by the presence of both dislocations and impurities analogously to the manner in which magnetic properties of metals are influenced by both free and bound electron currents. The far fields of more general types of impurities also contain terms more appropriately analogous to magnetic monopoles and these also influence the macroscopic elasticity of smectics. Parenthetically one should note that macroscopic elastic effects of focal conic defects can also be discussed in terms of the analogies presented here.

The third point to be made is that the analogy to magnetism in normal materials is only profitable in smectics subjected to low stresses. Above some critical stress one expects dislocation moments to become unstable, resulting in macroscopic plastic deformation or flow of the smectic. The existence of such a critical stress is a direct consequence of a separate analogy between smectics and superfluids that was pointed out by de Gennes.[3] In Table I we list the various analogies that will be useful. The analogy between the smectic liquid crystal and magnetism was discussed in detail by Pershan.[1] It is just a special case of the result originally derived by Peach and Koehler for crystals.[2] The analogy between vortices in classical fluids and electric

TABLE I. Useful analogies for discussing smectics.

Smectic	Superfluid He	Superconductor	Magnetism
Dislocation density l	Vortex density Ω	Flux density B	Electric current density J
Stress field $\nabla \cdot \phi = 0$	Superfluid current (momentum) $\nabla \cdot g_s = 0$	Superfluid current $\nabla \cdot J_s = 0$	Magnetic flux $\nabla \cdot B = 0$
Strain field $\nabla \times m = l$	Superfluid velocity $\nabla \times v_s = \Omega$	Vector potential $\nabla \times A = B$	Magnetic field $\nabla \times H = (4\pi/c)J$
Peach–Koehler force (Ref. 2)	Magnus force (Refs. 4—6)	Lorentz force (Refs. 7 and 8)	Lorentz force

currents in magnetism is well known.[4] The extension to superfluids seems to have occurred in a number of places.[5-8] In de Gennes's development of the analogy between smectics and superfluids, he emphasized the analogy to superconductors and pointed out that dislocations exist in smectics only to the extent smectics are analogous to type-II superconductors. The critical smectic strain could thus be discussed in analogy with critical currents in type-II superconductors. In many respects, however, superfluid helium behaves like an extreme example of a type-II superconductor and we find it slightly more convenient to discuss the critical smectic strain in analogy with critical currents in superfluid helium.

To illustrate the analogy as it is relevant to the present discussion, consider that below some critical value of current density superfluid helium can sustain a permanent supercurrent with no dissipation. There is stored kinetic energy in the superfluid, but no dissipative processes. Below the critical stress a smectic sustains stored elastic energy with no plastic flow, that is, with no dissipation. Above the critical current vortices are generated in superfluid helium and their motion in the force field arising from the superfluid current (i.e., Magnus effect) gives rise to dissipative processes that are analogous to the dissipative plastic deformation of a smectic sample associated with the motions of dislocation lines. Vortex generation in pure superfluid helium away from any boundaries has been discussed theoretically in terms of a nucleation and growth process similar to the condensation of drops in a supersaturated vapor.[9-11] At some critical velocity that is an intrinsic property of the material, independent of sample dimensions, thermally generated vortex loops are predicted to grow unstably and pass out the sides of the sample. Similarly, if a pure smectic is strained by, for example, homogeneously compressing the layers, there will be a critical strain at which thermally generated dislocation loops will be predicted to grow unstably. This is discussed at length in Sec. VI. This particular value of the critical current is rarely achieved in superfluid helium since there are other mechanisms by which unstable vortex growth can occur. Similarly, for the smectic it will be difficult to attain this value for the critical stress in the presence of other mechanisms for supplying dislocations. This is discussed in some detail in Sec. V.

For example, critical currents are often observed in superfluid helium under conditions where vortices have previously been trapped at surface irregularities. Superfluid currents much smaller than those required to create new vortex lines will be sufficient to cause preexisting lines to grow unstably.[12] Similarly trapped dislocation lines in a smectic will grow unstably at strain levels much below those required to create new dislocations. This is, in fact, one mechanism by which plastic deformation of conventional crystals is believed to occur.[13]

This paper is principally addressed to the elastic properties of smectic samples at stresses below the critical values under the assumption that the smectic is type II.[3] The formalism for dealing with dislocation mo-

ments and their macroscopic effects on the smectic stress-strain relationship is discussed in Sec. II. In Sec. III we develop Green's function solutions for the stress-strain fields surrounding point dislocation moments and then relate the far-field stress-strain patterns of real impurities to equivalent point dislocation moments and monopoles. In Sec. IV we make use of the previous results to make some numerical estimates regarding impurity-stress equilibrium effects. In particular we develop expressions for the diaelastic effects of impurities and also the osmotic smectic stress of impurities. In addition to estimating the critical smectic stress for unstable growth of pinned dislocations referred to above, we discuss the diaelastic effects of trapped dislocations at stresses below the critical.

II. DISLOCATION MOMENTS AND SMECTIC STRESS-STRAIN RELATIONS

Starting from one of the microscopic Maxwell's equation of electromagnetism,

$$\nabla \times \mathbf{B} = 4\pi c^{-1} \mathbf{j} + c^{-1} \frac{\partial \mathbf{E}}{\partial t}, \qquad (2.1)$$

a macroscopic equation is obtained by averaging over some finite volume element.[14] In the static case $\dot{\mathbf{E}} = 0$ and in the absence of macroscopic electric currents,

$$\langle \mathbf{j} \rangle = c\nabla \times \mathbf{M}, \qquad (2.2)$$

where

$$\mathbf{M} = (2c)^{-1} \langle \mathbf{r} \times \mathbf{j} \rangle. \qquad (2.3)$$

Combining Eq. (2.2) and the averaged version of Eq. (2.1) gives $\nabla \times (\mathbf{B} - 4\pi\mathbf{M}) = 0$. When there are macroscopic electric currents, Eq. (2.2) can be replaced by $\langle \mathbf{j} \rangle = \mathbf{J} + c\nabla \times \mathbf{M}$; however, there are formal difficulties in defining $\mathbf{M}$ when $\mathbf{J}$ varies spatially. In the limit that $\mathbf{J}$ varies slowly in space, the averaged form of Eq. (2.1) becomes

$$\nabla \times \mathbf{H} = 4\pi c^{-1} \mathbf{J}, \qquad (2.4)$$

with $\mathbf{H} \equiv \mathbf{B} - 4\pi\mathbf{M}$. The characteristic length that separates the macroscopic description from the microscopic is determined by the practical requirement that the $\mathbf{M}$ obtained from the average in Eq. (2.3) be sufficient to describe the relevant magnetic properties of the material. For example, in the case of a single electric current loop, the average would have to be over a sufficiently large volume that the average magnetic fields due to the loop are not significantly different from the average magnetic fields of a magnetic dipole situated at the center of the loop. In the case of dilute distribution of such loops, there are two different macroscopic regions: one is identical to the case of the individual loop and describes the fields between loops in terms of the fields of "equivalent" magnetic dipoles. For the second macroscopic region one averages over many loops and obtains average bulk magnetizations and fields. As the loops get closer together, the two regions coincide and in the limit of dense packing there is only one macroscopic region.

We can proceed in an identical manner and treat the macroscopic elastic properties of a smectic-A liquid crystal. The one caution worth mentioning is that the

word microscopic here refers to dimensions small com-
compared to the characteristic lengths in the distribu-
tion of dislocation positions, but large compared to the
dislocation core. This can be large compared to the
molecular dimensions and is not the conventional use of
"microscopic". With this in mind the microscopic prop-
erties of smectic-A liquid crystals can be discussed in
terms of a vector $\mathbf{m}(r)$ satisfying the differential
equation

$$\nabla \times \mathbf{m}(\mathbf{r}) = \mathbf{l}(\mathbf{r}), \qquad (2.5)$$

where $\mathbf{l}(\mathbf{r})$ is the density of dislocation lines (i.e., l
lines/cm^2).[1] Away from any dislocation line, $\mathbf{m}(r)$ can
be written as the gradient of a scalar variable describ-
ing the position of the smectic layers $[\mathbf{m}(r) = - a^{-1}\nabla u(r)]$;
however, this is not generally true since $\oint \mathbf{m}(\mathbf{r}) \cdot d\mathbf{r}$
will not be zero if the path encloses a net number of dis-
location lines. As in the magnetic problem, one aver-
ages and obtains

$$\langle \mathbf{l} \rangle = \mathbf{L} + \nabla \times \mathbf{P}. \qquad (2.6)$$

Following the usual practice in magnetic problems, we
will emphasize examples in which the average density
of dislocation lines $\mathbf{L} = 0$. For example, a smectic sam-
ple that is homeotropically aligned between glass slides
that are parallel on average can be regarded as having
$\mathbf{L} = 0$ everywhere and the average dislocation moment

$$\mathbf{P} = \tfrac{1}{2}\langle \mathbf{r} \times \mathbf{l} \rangle. \qquad (2.7)$$

Combining Eqs. (2.5) and (2.7) gives $\nabla \times (\mathbf{m} - \mathbf{p}) = 0$ and
allows one to infer

$$\mathbf{m} = - a^{-1}\nabla u + \mathbf{p}. \qquad (2.8)$$

In analogy to the current loop examples discussed
above, one can consider a distribution of small disloca-
tion loops. For a dilute concentration of such loops the
scalar u actually describes layer displacements in the
space between the loops. For concentrated distributions
of loops, u describes an average displacement.

In order to illustrate the significance of Eq. (2.8),
consider the example of a small edge dislocation of unit
strength curved to form a circular loop of radius R
centered at the origin (see Fig. 1). Except for a sign
change, the strain m_3 for this loop was previously calcu-
lated[1] and we quote the result for the spatial Fourier
transform

$$m_3(\mathbf{q}) = 2\pi\lambda^2 R q_\perp^3 J_1(q_\perp R)(q_3^2 + \lambda^2 q_\perp^4)^{-1},$$

where J_1 is the ordinary Bessel function of order one
and $q_\perp^2 = q_1^2 + q_2^2$. Averaging over dimensions large com-
pared to R is equivalent to considering only those $q_\perp$
$\ll R^{-1}$. In this region the dislocation moment density can
be written $P_3(\mathbf{r}) = \pi R^2 \delta(\mathbf{r})$ and its Fourier transform $P_3(\mathbf{q})$
$= \pi R^2$. Also $J_1(q_\perp R) \approx \tfrac{1}{2}q_\perp R$ and

$$m_3(\mathbf{q}) = P_3(\mathbf{q})\lambda^2 q_\perp^4 (q_3^2 + \lambda^2 q_\perp^4)^{-1}. \qquad (2.9)$$

This same result is trivially obtained by combining
Eq. (2.8) with the previous relations defining the stress
tensor.[1] Assuming the pressure $P = 0$, we get

$$\sigma_{33} \equiv - \phi_3 = ab m_3,$$

$$\sigma_{13} \equiv - \phi_\perp = - ab\lambda^2 \nabla_\perp (\nabla_\perp \cdot \mathbf{m}_\perp), \qquad (2.10)$$

$$\nabla \cdot \phi = 0. \qquad (2.11)$$

On substitution of $\mathbf{P}_\perp - a^{-1}\nabla_\perp u$ for $\mathbf{m}_\perp$ the 3 derivative of
Eq. (2.11), $\nabla_3(\nabla \cdot \phi) = 0$ can be written

$$- ab(\nabla_3)^2 m_3 + ab\lambda^2 \nabla_\perp^2(\nabla_3\nabla_\perp \cdot \mathbf{P}_\perp - a^{-1}\nabla_\perp^2\nabla_3 u) = 0.$$

However, since $a^{-1}\nabla_3 u = P_3 - m_3$, one obtains

$$\nabla_3^2 m_3 - \lambda^2(\nabla_\perp^2)^2 m_3 = \lambda^2(\nabla_\perp)^2(\nabla_3\nabla_\perp \cdot \mathbf{P}_\perp - \nabla_\perp^2 P_3). \qquad (2.12)$$

With $\mathbf{P}_\perp = 0$ in the present example, Eq. (2.9) is trivially
obtained.

The macroscopic elastic properties of a smectic
densely packed with dislocations will follow from Eqs.
(2.8), (2.10), and (2.11) on averaging over sufficiently
large volume elements, if one knows how the disloca-
tion moment density $\mathbf{P}$ depends on the macroscopic
stress ϕ. Assuming $\mathbf{P}$ is a linear local function of ϕ, to
lowest order in wave vector, one can write

$$P_3 = \chi_{33}\phi_3 + \chi_{3\perp}\nabla_3\nabla_\perp \cdot \phi_\perp,$$
$$\mathbf{P}_\perp = \chi_{\perp 3}\nabla_\perp\nabla_3\phi_3 + \chi_{\perp\perp}\phi_\perp. \qquad (2.13)$$

Since

$$\phi_3 = - abP_3 + b\nabla_3 u,$$
$$\phi_\perp = ab\lambda^2\nabla_\perp\nabla_\perp \cdot \mathbf{P}_\perp - b\lambda^2\nabla_\perp\nabla_\perp^2 u, \qquad (2.14)$$

substitution of (2.13) into (2.14) obtains

$$\phi_3 \approx b(1 + ab\chi_{33})^{-1}\nabla_3 u,$$
$$\phi_\perp \approx - b\lambda^2\nabla_\perp^2\nabla_\perp u, \qquad (2.15)$$

where we have neglected terms of higher order in wave
vector. The interpretation of Eq. (2.15) is that a finite
dislocation moment density will induce a renormalization
of the smectic elastic constant b,

$$b_{\text{eff}} = b(1 + ab\chi_{33})^{-1}. \qquad (2.16)$$

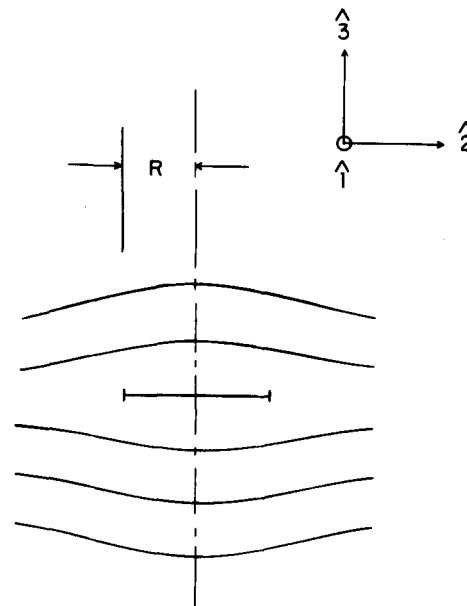If $\chi_{33} > 0$, this is the diaelastic effect alluded to earlier.[1]

FIG. 1. Schematic representation of a circular dislocation loop
of radius R. Note that the sense of l is into the page on the
right and out on the left. This is just opposite to the loop dis-
cussed in detail in Sec. VIII of Ref. 1. The coordinate system
shown here will also be used throught this paper. The 3 axis
is normal to the smectic planes.

Although finite values of χ_{33} could possibly arise from thermally generated dislocation loops, this effect is probably better treated by other approaches. This is discussed in Sec. VI. On the other hand, pinned dislocation lines or loops will give rise to χ_{33} effects. As we will discuss below, certain impurities in a smectic induce stress-strain fields similar to dislocation arrays and the motions of these impurities in response to external stresses will also contribute a renormalization factor to b.

Note also that in the case of a point dipole located at the origin, $\mathbf{P} = (p_3\vec{x}_3 + \mathbf{p}_\perp)\delta(r)$, Eq. (2.11) reduces to

$$\nabla_3^2 u - \lambda^2(\nabla_\perp^2)^2 u = a[p_3\nabla_3\delta(r) - \lambda^2(\mathbf{p}_\perp)\cdot(\nabla_\perp)\nabla_\perp^2\delta(r)], \quad (2.17)$$

for which the solution can be given in terms of a Green's function to be defined below [Eq. (3.2)]:

$$u(r) = p_3 G(r) - \lambda x_3|x_3|^{-1}(\mathbf{p}_\perp)\cdot(\nabla_\perp)G(r). \quad (2.18)$$

Although it is explicitly shown below, it is also apparent from Eq. (2.17) that $G(r)$ is an odd function of x_3 and even in $(\mathbf{r}_\perp)$ while the second term is even in x_3 and odd in $(\mathbf{r}_\perp)$.

III. GREEN'S FUNCTION CONSIDERATIONS

Given any arbitrary distribution of magnetic dipole moments it is always possible to find an equivalent distribution of electric currents such that the macroscopic magnetic fields of the two are identical. In the spirit of the analogy between smectic strains and magnetic fields we ask whether or not the macroscopic strain fields of an arbitrary distribution of localized impurities can be expressed in terms of an equivalent dislocation distribution. We will demonstrate below that this is not generally possible, since strain fields have source terms analogous to magnetic monopoles. Nevertheless in examples where the smectic is macroscopically uniaxial (or planar), impurities analogous to both positive and negative magnetic monopoles exist in equal numbers and the smectic can once again be treated analogously to a magnetic material.

To develop these ideas mathematically, consider a Green's function $G(r - r')$ defined by

$$\nabla_{33}G - \lambda^2(\nabla_\perp^2)^2 G = a\nabla_3\delta(\mathbf{r} - \mathbf{r}'), \quad (3.1)$$

with boundary conditions that $G \to 0$ as $\mathbf{r} \to \infty$. It is straightforward to solve Eq. (3.1) and obtain $G(\mathbf{r} - \mathbf{r}')$. For $\mathbf{r}' = 0$ this gives

$$G = a(8\pi\lambda x_3)^{-1}\exp[-r_\perp^2(4\lambda|x_3|)^{-1}]. \quad (3.2)$$

Assume an infinite sample with a finite impurity located near the origin $\mathbf{r} = 0$. Using conventional Green's function methods, the strain at any point $\mathbf{r}'$ can be expressed as a surface integral over the impurity:

$$a\nabla_3'u(r') = \int dA_i[-\delta_{i3}\nabla_3 u(r) + \lambda^2(\nabla_\perp)_i\nabla_\perp^2 u(r)]G(r - r')$$

$$+ \int dA_i[-\lambda^2\nabla_\perp^2 u(r)](\nabla_\perp)_i G(r - r')$$

$$+ \int dA_i[\lambda^2(\nabla_\perp)_i u(r)]\nabla_\perp^2 G(r - r')$$

$$+ \int dA_i[-\lambda^2 u(r)](\nabla_\perp)_i \nabla_\perp^2 G(r - r')$$

$$+ \int dA_i(\delta_{i3}u)\nabla_3 G(r - r'), \quad (3.3)$$

with $d\mathbf{A}(r)$ pointing out of the impurity and the convention

that $(\nabla_\perp)_i$ is zero if $i = 3$ and $\nabla_\perp^2 = \nabla_1^2 + \nabla_2^2$. A moment expansion is obtained from the expansion

$$G(r - r') = -G(r') + r_i\nabla_i'G(r') - 2^{-1}r_ir_j\nabla_i'\nabla_j'G(r') + \cdots \quad (3.4)$$

on grouping together terms with the same asymptotic behavior as $\mathbf{r}'$ approaches infinity. Since $G(r)$ goes to zero algebraically as x_3^{-1} along parabolic curves specified by constant values of $r_\perp^2|x_3|^{-1}$, it is convenient to consider the asymptotic behavior along this curve. The perpendicular derivatives, for example,

$$(\nabla_\perp')_i G(r') = -(r_\perp')_i(2\lambda|x_3'|)^{-1}G(r'),$$

fall off as $|x_3'|^{-3/2}$ along the curve. Grouping together terms of similar asymptotic behavior

$$G(r - r') = -G(r') + (r_\perp)_i(\nabla_\perp')_i G(r')$$

$$+ [+x_3\nabla_3 G(r') - 2^{-1}(r_\perp)_i(r_\perp)_j(\nabla_\perp')_i(\nabla_\perp')_j G(r')]$$

$$+ \text{terms of order } |x_3|^{-5/2}\exp[-r_\perp^2(4\lambda|x_3|)^{-1}]. \quad (3.5)$$

With this expansion for G substituted into Eq. (3.3), the coefficient of the leading term is

$$I^0 = \int dA_i[+\delta_{i3}\nabla_3 u(r) - \lambda^2(\nabla_\perp)_i(\nabla_\perp)^2 u(r)]. \quad (3.6)$$

Taking the stress tensor to have components[1]

$$\sigma_{33} = -b\nabla_3 u = -\phi_3,$$

$$(\sigma_{\perp 3})_i = \lambda^2 b\nabla_\perp^2(\nabla_\perp)_i u = -\phi_i, \quad (3.7)$$

$$(\sigma_{\perp\perp})_{ij} = -\lambda^2 b(\nabla_\perp)_i(\nabla_\perp)_j\nabla_3 u,$$

gives $I^0 = b^{-1}\int dA_i\sigma_{i3}$, which is just the 3 component of the net force on the impurity and can be taken to vanish. The coefficient of the term proportional to $(\nabla_\perp)_i G$,

$$(I^1)_i = \int dA_j\{[-\delta_{j3}\nabla_3 u + \lambda^2(\nabla_\perp)_j\nabla^2 u](r_\perp)_i - \delta_{ij}^\perp\lambda^2\nabla_\perp^2 u\}, \quad (3.8)$$

vanishes since it is just the torque on the impurity. To demonstrate this, take the net torque exerted on the outer sample surface to be zero and use the divergence theorem to write

$$(I^1)_i = \int dA_j[b^{-1}\sigma_{j3}(r_\perp)_i] + \int dV\lambda^2(\nabla_\perp)_i\nabla_\perp^2 u, \quad (3.9)$$

where $\sigma_{j3} = \sigma_{33}\delta_{j3} + (\sigma_{\perp 3})_j$. The sign in front of the second term is positive because the vector $d\mathbf{A}$ points out of the impurity and into the sample. However, one can also write

$$\lambda^2(\nabla_\perp)_i\nabla_\perp^2 u = b^{-1}\nabla_3[x_3(\sigma_{\perp 3})_i] + b^{-1}(\nabla_\perp)_j[x_3(\sigma_{\perp\perp})_{ij}], \quad (3.10)$$

so that on using the divergence theorem and the symmetry of the stress tensor

$$(I^1)_i = b^{-1}\int dA_j[\sigma_{j3}(r_\perp)_i - \sigma_{ji}x_3], \quad (3.11)$$

where $\sigma_{ji} = (\sigma_{\perp 3})_i\delta_{j3} + (\sigma_{\perp\perp})_{ji}$. Requiring the perpendicular components of the torque to vanish demands $(I^1)_i = 0$.

The first nonvanishing terms in the moment expansion of Eq. (3.3) are of the order of $|x_3|^{-2}\exp[-r_\perp^2(4\lambda|x_3|)^{-1}]$. These are the terms proportional to $\nabla_3'G(r')$ and $(\nabla_\perp')_i(\nabla_\perp')_m G(r')$:

$$a\nabla_3'u(r') = \int dA_i\{\delta_{i3}[u(r) - x_3\nabla_3 u(r)] + \lambda^2 x_3(\nabla_\perp)_i\nabla_\perp^2 u(r)\}\nabla_3'G(r')$$

$$- 2^{-1}\int dA_i\{(r_\perp)_i(r_\perp)_m[-\delta_{i3}\nabla_3 u(r) + \lambda^2(\nabla_\perp)_i\nabla_\perp^2 u(r)]$$
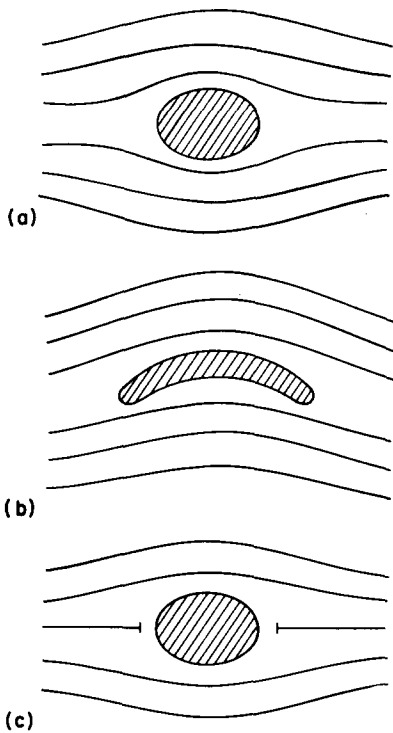
FIG. 2. Schematic illustration of impurities with nonzero values of (a) p_3 and (b) Q_{ij}. (c) An impurity for which p_3 is modified by a pinned dislocation.

$$-\lambda^2[\delta_{li}(r_\perp)_m + \delta_{lm}(r_\perp)_i]\nabla_\perp^2 u(r)$$

$$+2\lambda^2(\nabla_\perp)_i u(r)\delta_{lm}\}(\nabla_\perp')_i(\nabla_\perp')_m G(r')$$

$$+\text{terms of order } |x_3|^{-5/2}\exp[-r_\perp^2(4\lambda|x_3|)^{-1}].$$

$$(3.12)$$

The first term in Eq. (3.12) corresponds to the first term in the solution to the point dipole problem given by Eq. (2.18) if we define

$$p_3 = a^{-1}\int dA_i[\delta_{i3}(u-x_3\nabla_3 u) + \lambda^2 x_3(\nabla_\perp)_i\nabla_\perp^2 u]. \qquad (3.13)$$

The divergence of the integrand vanishes so that the moment p_3 defined this way is independent of the choice of surface for the integration.

Second, observe that the value of p_3 depends on both the impurity itself and also the externally applied strain surrounding the impurity. For example, p_3 depends on the difference between u and $x_3\nabla_3 u$ rather than just u alone. Stated another way, this implies that the dislocation moment of an impurity is somewhat polarizable by external stresses. As a practical matter, this is probably not too important for small strains, but it is fundamental. The second term in Eq. (3.12) does not correspond to the strain field of any dislocation distribution. Note that $(\nabla_\perp')_i(\nabla_\perp')_j G(r')$ is even in $r_\perp$ but odd in x_3, so that on integration it gives a term in $u(r)$ that is even in both $r_\perp$ and x_3. For example, defining

$$Q_{lm} = -(2a)^{-1}\int dA_i\{(r_\perp)_i(r_\perp)_m[-\delta_{i3}\nabla_3 u(r) + \lambda^2(\nabla_\perp)_i\nabla^2 u(r)]$$

$$-\lambda^2[\delta_{li}(r_\perp)_m + \delta_{lm}(r_\perp)_i]\nabla_\perp^2 u(r) + 2\lambda^2(\nabla_\perp)_i u(r)\delta_{lm}\}$$

$$(3.14)$$

a solution of the same form as Eq. (3.12) would be ob-

tained from the differential equation

$$\nabla_3^2 u - \lambda^2(\nabla^2)u = a[p_3\nabla_3 + Q_{lm}(\nabla_\perp)_i(\nabla_\perp)_m]\delta(r). \qquad (3.15)$$

This differs from Eq. (2.18) in two respects: First, the expansion equation (3.12) was only carried as far as terms of order $|x_3|^{-2}\exp[-r^2(4\lambda|x_3|)^{-1}]$ and the $p_\perp$ term in Eq. (2.18) comes from the next term in the expansion. More important is the fact that the Q_{lm} term cannot be represented in terms of an equivalent dislocation distribution. In the spirit of the analogy between smectic strains and magnetic fields, one can think of this term as arising from impurities analogous to magnetic monopoles. For example, the source terms analogous to perpendicular components of dislocation dipoles [see Eq. (2.18)] $-a\lambda^2(p_\perp)_i(\nabla_\perp)_i\nabla_\perp^2\delta(r)$ has one more derivative than the Q_{lm} term in Eq. (3.15). This is the conventional difference between dipole and monopole source terms.

Figures 2(a) and 2(b) illustrates schematically the type of impurities that would have nonvanishing values of either p_3 or Q_{xx}, respectively. In both cases we assume boundary conditions on the impurity that constrain the smectic layers to follow the surfaces. In Fig. 2(a) there are obviously large local strains at the sides of the impurity and it is possible that a dislocation loop will become pinned around the impurity as illustrated in Fig. 2(c). This does not alter the principal physical point that an impurity like the one shown in 2(a) will have some net nonvanishing value of p_3 even if the bare value is partially shielded by dislocation effects. The impurity illustrated in Fig. 2(b) should couple most strongly to splay-type distortions and in the absence of macroscopic splay any local smectic region should contain equal numbers of impurities oriented like the one shown and those of opposite sign, e.g., turned upside down. This also is suggestive of monopole effects since in both electricity and magnetism the absence of macroscopic fields E (or H) implies equal numbers of charges (or monopoles) of opposite sign.

Note also that ap_3 has the dimensions of volume. The smectic distortions produced by an impurity like the one illustrated in Fig. 2(a) are obviously related to the amount of smectic material that has to be displaced to deform the layers as shown. Figure 2(c), however, emphasizes that in real cases ap_3 is not just the volume of the impurity, but rather it represents an effective volume that is most likely smaller than the true value. Different boundary conditions between smectic and impurity can also produce negative values for p_3.

In the two examples previously treated in this manuscript, that is in the absence of any impurities and also in the case of point dislocation dipoles with $p_3 \neq 0$, equations like (3.15) followed from the condition $\nabla_i\sigma_{i3} = 0$. In the present case this should also follow and we make the identification

$$\sigma_{33} = -\phi_3 = -b\nabla_3 u + bap_3\delta(r),$$

$$(\sigma_\perp)_{i3} = -(\phi_\perp)_i = b\lambda^2(\nabla_\perp)_i\nabla_\perp^2 u + baQ_{ji}(\nabla_\perp)_j\delta(r),$$

$$(\sigma_{\perp\perp})_{ij} = -b\lambda^2(\nabla_\perp)_i(\nabla_\perp)_j\nabla_3 u - baQ_{ij}\nabla_3\delta(r). \qquad (3.16)$$

The first terms are the "external" stresses on the impurity and include effects such as boundary conditions and images due to the boundaries.

The interaction energy between externally applied stresses and impurities with moments p_3 and Q_{ij} can be obtained by considering a virtual displacement of the system under conditions that no external work is done. Then the interaction energy δE^{INT} is just the negative of the change δE in the elastic energy. Taking the elastic energy to be

$$E = 2^{-1}b \int dV[(\nabla_3 u)^2 + \lambda^2(\nabla_\perp^2 u)^2], \qquad (3.17)$$

the interaction energy $\delta E^{INT} = -\delta E$,

$$\delta E^{INT} = -b \int dV[\nabla_3 u \nabla_3 \delta u + \lambda^2(\nabla_\perp^2 u)\nabla^2 \delta u]. \qquad (3.18)$$

Integration by parts give a surface term δW_s describing the work done on the smectic by the boundaries that is zero and a volume term

$$\delta E^{INT} = -b \int dV[-\nabla_3^2 u + \lambda^2(\nabla_\perp^2)^2 u]\delta u. \qquad (3.19)$$

From Eq. (3.15) this can be expressed as

$$\delta E^{INT} = -ab[p_3 \nabla_3 \delta u - Q_{ij}(\nabla_\perp)_i(\nabla_\perp)_j \delta u]. \qquad (3.20)$$

The term $-abp_3\nabla_3\delta u = -ap_3\phi_3$ is analogous to the energy of a magnetic dipole in an extenal field, $-\mathbf{u} \cdot \mathbf{B}$. An interesting feature of the Q_{ij} term is the result that although a uniform value of $(\nabla_\perp)_i(\nabla_\perp)_j u$ (corresponding to a uniform splay) induces no change in the stress tensors given by Eqs. (3.16), it does change the energy of impurities with nonvanishing values of Q_{ij}. The effects of uniform splaylike distortions on Q_{ij} are identical to the effects that a uniform magnetostatic potential would have on a magnetic monopole. Equation (3.20) consitutes a potential energy that is a nonlocal function of the stress. The force, however, is a local function of the stress. This is best illustrated by considering the special case of an impurity with $Q_{ij} = Q\delta_{ij}^\perp$. In the presence of inhomogeneities in δu, there will be forces on the impurity that can be calculated by taking the gradient of the interaction term in Eq. (3.20). Writing

$$\phi_3 = +b\nabla_3 u = -\sigma_{33}^{ext} \quad \text{and} \quad (\phi_\perp)_i = -\lambda^2 b(\nabla_\perp)_i\nabla^2 u = -(\sigma_\perp)_{i3}^{ext}$$

gives

$$F_k = +ap_3\nabla_k\phi_3 + a\lambda^{-2}Q(\phi_\perp)_k - a\delta_{k3}Q\nabla_\perp^2\phi_3. \qquad (3.21)$$

The force proportional to $Q\phi_{\perp k}$ is exactly what one expects for a monopole in the presence of a constant field. The p_3 term is analogous to the force on a magnetic dipole in a magnetic field gradient and the last term is a higher-order correction. There are, however, two special points to make regarding the Q_{ij} terms.

First, Q_{ij} itself is not conserved since an impurity oriented like the one shown in Fig. 2(b) can presumably flip over and thus reverse the sign of Q. Second, there is no reason to assume that all impurities must have the symmetric form assumed above (i.e., $Q_{ij} = Q\delta_{ij}^\perp$), and in the general case Eq. (3.20) implies that some orientations of the impurity will have lower energy than others for a given inhomogeneity δu. It is reasonable, however, to expect that in the presence of an inhomogeneity in δu the impurity will relax so that its orientation is in statistical equilibrium with the local configuration. This orientational relaxation will occur in some "microscopic" time characterized by the impurity dimensions, etc., and the local properties of the smectic. Our main con-

cern here, however, is not with phenomena occurring on microscopic times, but the long-wavelength long-time elastic behavior and for these purposes we can replace Q_{ij} by a statistical average that is nonzero only because $(\nabla_\perp)_i(\nabla_\perp)_j u$ is nonzero. That is, the statistical average of Q_{ij} will be proportional to $(\nabla_\perp)_i(\nabla_\perp)_j\delta u$ and by partial integrations the average value of the Q_{ij} term in δE is proportional to $(\nabla_\perp^2 u)^2$.

Since this is not different from the result that would be obtained if we started by assuming $Q_{ij} = Q\delta_{ij}^\perp$, we proceed by neglecting the part of Q_{ij} lacking cylindrical symmetry. Note also that if the expectation value of Q_{ij} is proportional to $(\nabla_\perp)_i(\nabla_\perp)_j\delta u$, the expectation value of $(\nabla_\perp)_i Q_{ij}$ is proportional to σ_{j3} or $(\phi_\perp)_j$. In the discussion of dislocation moments p_3, the assumption was made that a macroscopic average over a sufficiently large volume would result in the moment density $P_3 = \chi_{33}\phi_3$. The implication here is that a similar macroscopic average of the Q_{ij} would result in a "monopole" density satisfying

$$(\nabla_\perp)_i Q_{ij} = \chi^*(\phi_\perp)_j. \qquad (3.22)$$

Macroscopic averages of Eq. (3.16) give

$$\sigma_{33} = -\phi_3 - baP_3,$$
$$(\sigma_\perp)_{i3} = -(\phi_\perp)_i = b\lambda^2(\nabla_\perp)_i\nabla^2 u + ba(\nabla_\perp)_j Q_{ij}, \qquad (3.23)$$
$$(\sigma_{\perp\perp})_{ij} = -b\lambda^2(\nabla_\perp)_i(\nabla_\perp)_j\nabla_3 u - ba\nabla_3 Q_{ij},$$

where $Q_{ij} = Q\delta_{ij}^\perp$. With the assumptions made just above,

$$\sigma_{33} = -\phi_3 = -b(1 + ba\chi_{33})^{-1}\nabla_3 u,$$
$$(\sigma_\perp)_{i3} = -(\phi_\perp)_i = -b\lambda^2(1 + ba\chi^*)^{-1}(\nabla_\perp)_i\nabla_\perp^2 u. \qquad (3.24)$$

In the same way that dislocation moments p_3 can be responsible for diaelastic effects on the σ_{33} stress, so can impurities with moments p_3. In addition, impurities with "monopole"-like boundary conditions have diaelastic effects on the nematiclike elasticity. Presumably these latter effects are also present in nematics.

IV. IMPURITY-STRESS EQUILIBRIUM EFFECTS

Consider a smectic-A sample homeotropically aligned between glass slides in thermal equilibrium with a stress-free smectic region as shown in Fig. 3. Assume that the stress-free region acts as a reservoir of impurities that diffuse into the region between the glass slides. Take the impurity concentration in the stress-free region to have a constant value c_0 [number/cm^3] and assume that the impurities have dislocation moments p_3. In terms of the interaction energy derived from Eq. (3.20), thermal equilibrium between the two regions requires

$$kT\ln[c] - ap_3\phi_3 = kT\ln[c_0]. \qquad (4.1)$$

From Eqs. (3.23) with $P_3 = p_3 c$,

$$\phi_3 = -abp_3 c + b(\nabla_3 u), \qquad (4.2)$$

so that

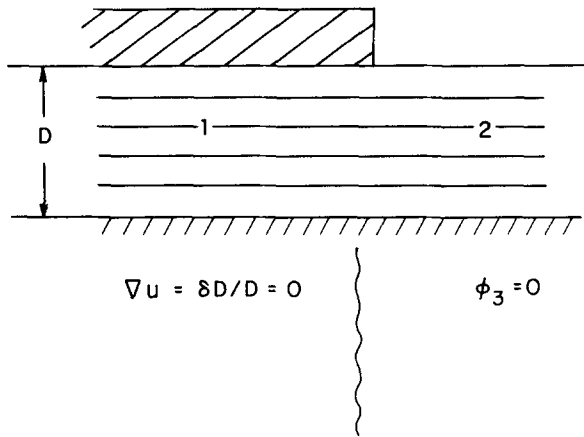$$\phi_3 + abp_3 c_0 \exp[ap_3\phi_3(kT)^{-1}] = b\nabla_3 u, \qquad (4.3)$$

FIG. 3. Schematic illustration of a smectic sample with non-uniform boundary conditions. The spacing D is originally chosen so that the entire sample is stress free.

and with $a p_3 \phi_3 \ll kT$,

$$\phi_3 = \frac{b\nabla_3 u - a b p_3 c_0}{1 + a^2 p_3^2 b c_0 (kT)^{-1}}. \tag{4.4}$$

Consider now the geometry in Fig. 3 where initially there are no impurities and both regions are stress free. If impurities are then dissolved in the region outside the rigid boundary [region (2)], they will tend to diffuse into region (1). Assuming the spacing between the glass plates are held fixed (i.e., $\nabla_3 u = 0$), the increasing impurity concentration will develop a stress gradient tending to oppose further diffusion. The equilibrium stress will be given by Eq. (4.4) and $\nabla_3 u = 0$. Taking $a p_3 \sim 10^{-21}$ cm³, $b \sim 10^8$ dyn cm⁻² would obtain an "osmotic smectic stress" of the order of 10^6 dyn/cm² (1 atm) for a reservoir concentration of 10^{19} impurity molecules/cm³. If p_3 is positive so that the impurity behaves as extra smectic layers, the induced stress is negative, implying a compressional smectic stress. In practice this value would not be achieved if the osmotic stress exceeds the critical stress for unstable dislocation growth. As a rough rule of thumb, one might expect that the critical stress is of the same order as the stress required to change the over-all sample thickness by the thickness of half of one smectic layer. Taking the sample thickness to be D implies a critical stress $\sim ab(2D)^{-1}$. Using the values quoted above, the sample would have to be thinner than 50 smectic layers to attain the full value. Note also the "renormalization factor" represented by the denominator of Eq. (4.4). Referring to Eq. (3.23) an externally applied stress ϕ_3 induces a change in impurity concentration that has the effect of making the effective value of b smaller than for the pure sample. For the numerical values assumed above, concentrations $c_0 \sim 10^{20}$ cm⁻³ are required to make $b_{eff} \sim \frac{1}{2}b$.

V. PINNED DISLOCATIONS

Irregularities at the surfaces containing the smectic and impurities within the smectic can serve to pin dislocations within the material. Figure 4 illustrates a hypothetical edge dislocation line of length L trapped by points A and A'. For simplicity we assume that in the absence of a macroscopic stress the segment is straight An applied stress ϕ_3 will produce a force per unit length

on the dislocation ($\mathbf{F} = a\mathbf{l} \times \boldsymbol{\phi}$) that will cause the line to bow. Assuming that the dislocation has a line tension γ, the deformed configuration shown in Fig. 4 has an energy

$$E = \gamma R(2\theta - 2\sin\theta) - a\phi_3 R^2 2^{-1}(2\theta - \sin2\theta), \tag{5.1}$$

with $2R\sin\theta = L$. Near $\theta \approx 0$ Eq. (5.1 can be expanded in a power series to obtain

$$E \approx \frac{1}{6}\gamma L\theta^2 - \frac{1}{6}a\phi_3 L^2\theta \tag{5.2}$$

which has a minimum at

$$\theta \approx a\phi_3 L(2\gamma)^{-1}. \tag{5.3}$$

Bowing of the dislocation line induces a change in the dislocation moment

$$\delta p_3 \approx aL^3\phi_3(12\gamma)^{-1}, \tag{5.4}$$

such that if there are n_0 such pinned lines per cm³ ($P_3 = n_0\delta p_3$), Eq. (2.14) gives

$$\phi_3 = -n_0 a^2 L^3 b\phi_3(12\gamma)^{-1} + b\nabla_3 u \tag{5.5}$$

and [Eq. (2.16)]

$$b_{eff} = b[1 + n_0 a^2 L^3 b(12\gamma)^{-1}]^{-1}. \tag{5.6}$$

If we take the order of magnitude of $\gamma \sim ba^2$, the denominator of Eq. (5.6) is $1 + \frac{1}{12}n_0 L^3$. Assuming dislocations have a core radius r_c the fractional volume occupied by dislocations $\delta v/v$ is of the order $r_c^2 L n_0$ such that $n_0 L^3 \sim (\delta v/v)(L/r_c)^2$. Since $L/r_c \gg 1$, a reasonable density of edge dislocations can result in significant reduction of b_{eff}.

The above analysis is only applicable to low stresses. For larger values the dislocation will become unstable resulting in plastic deformation in the same manner that these effects occur in crystals.[13] Glaberson and Donnelly[12] discussed the analogous effect of unstable growth of pinned vortex lines in superfluid helium as the result of the Magnus force. By numerical methods one can show from Eq. (5.1) that for

$$\phi_3^c \gtrsim 2ba/L, \tag{5.7}$$

the bowed dislocation becomes absolutely unstable for any finite value of R. Nabarro discussed the numerous possible mechanisms that are available for plastic flow under this condition.[13] Since real smectic samples will usually have a distribution of pinned dislocations of
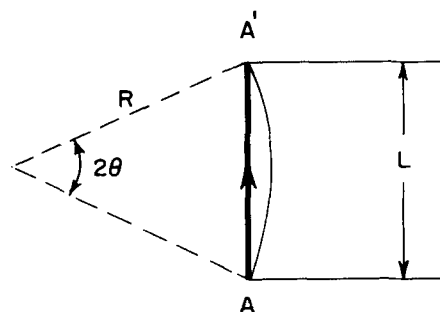

FIG. 4. Schematic illustration of a pinned edge dislocation running from A to A'. The heavy line indicates the straight equilibrium configuration in the absence of an external stress and the lighter solid line illustrates the deformed configuration in the presence of a stress $\phi_3 > 0$. The $\hat{3}$ direction is out of the page.

varying lengths and configurations, it is difficult to make make a specific prediction for the actual critical stress. As a practical matter, one might expect that the longest "free dislocation length" L might be of the same order as the sample thickness. This would predict a critical strain not too different from values suggested in the experiments of Clark.[15]

VI. THERMALLY GENERATED DISLOCATION LOOPS

Earlier in this paper we emphasized that it is not always profitable to speak of dislocations in a smectic system for the same reasons that it is not always profitable to speak of fluxoids in a superconductor. In the latter case there are type-I and type-II superconductors and discussions of fluxoids are restricted to the latter. de Gennes[3] originally made this point with regard to smectics. Confusion arises if one loses sight of the fact that both the smectic order parameter and the defects in in the order (i.e., dislocations) are macroscopic phenomena in the sense that they involve correlations in the behavior of many individual molecules. These quantities are defined as macroscopic averages in which a considerable number of microscopic fluctuations are assumed to average out. In this sense there is some minimum number of molecules (or in terms of a correlation length ξ a minimum volume $\sim \xi^3$) that enters into the definition of the order parameter and unless the number of molecules involved in the average exceeds this, the order parameter will be poorly defined. The macroscopic symmetry of the smectic phase tells us that it is always possible to identify a volume large enough to allow a meaningful definition of the order parameter.

On the other hand, in order to define a dislocation one first defines the macroscopic smectic order parameter over an extended region of the sample and then finds that topological considerations demand a defect somewhere. Thus the dislocation can only be defined if it is surrounded by an extended region of sample free of macroscopic defects. For example, it is not self-consistent to assume a model in which the distance between dislocations is less than the correlation length ξ needed in the macroscopic averages by which the order parameter is defined. More to the point of the present discussion is that confusion will arise if one attempts discussion of dislocation loops with dimensions smaller than ξ. In regard to thermally generated dislocation loops, the length ξ should probably be defined self-consistently to be large enough that the equilibrium number of loops with dimensions comparable or larger than ξ is vanishingly small. In summary, we are arguing that there is a continuous distribution of "microscopic" thermal fluctuations that must be averaged over in order to define the smectic order parameter and that it makes no sense to define an averaging volume smaller than the largest thermal defect to appear in significant numbers. Macroscopic effects of all these "microscopic" fluctuations are summarized by the equilibrium value and the thermal fluctuations in the order parameter. In this regard dislocation theories of melting should be regarded most cautiously since the best to be expected from them is a prediction of the onset of a pretransition region in which ξ begins to grow. The width of the pretransition

region and the actual character of the phase transition are beyond any such theory.

All of these remarks are also applicable to thermally generated vortex loops in superfluid helium. In the presence of macroscopic current the superfluid becomes unstable in the sense that microscopic thermal fluctuations can grow into macroscopic vortex loops. Langer and Fisher,[9] Vinen,[10] and Iordanskii[11] all discussed this in detail and their results can be directly applied to the smectic. Consider the paper by Langer and Fisher.[9] They argue that in the presence of a macroscopic superfluid velocity v_s there is a critical radius $R_c(v_s)$ for vortex rings. Rings with $R < R_c$ will tend to contract and sink into the thermal distribution of microscopic fluctuations, while rings with $R > R_c$ will tend to grow without bound (except for pinning effects). Since vortex rings with radii of the order of R_c are virtually nonexistent in thermal equilibrium, Langer and Fisher calculate the probability of their appearance in terms of a nucleation and growth process. The macroscopic Landau-Ginzburg equations can be used to obtain an expression $E(R)$ for the energy of a vortex ring of radius $R \gg \xi$. For sufficiently small currents v_s [e.g., $R_c(v_s) \gg \xi$], Langer and Fisher obtain the following expression for the nucleation rate per unit volume of unstable vortex rings:

$$f \approx f_0 \exp[-E(R_c)/kT], \qquad (6.1)$$

where f_0 is a characteristic rate for microscopic processes. They estimate f_0 crudely by dividing a frequency of the order of (sound velocity)(molecular dimension)$^{-1}$ by the molecular volume. Then they assume some value for the smallest detectable nucleation rate f_L and calculate the value of $R_c(v_s)$ such that $E(R_c) = kT \ln(f_0/f_L)$. The velocity v_s thus obtained defines a critical current for homogeneous nucleation of dissipative processes in superfluid helium.

The one practical difference between the problem of critical smectic stress and critical superfluid current is the boundary conditions. Typically critical smectic stress might be experimentally studied in thin smectic samples homeotropically aligned between glass slides. Critical stresses must then exceed values corresponding to strains of the order of $a(2D)^{-1}$, where a is the smectic layer spacing and D is the sample thickness.[1] Langer and Fisher considered the superfluid problem analogous to $D \to \infty$.

Kleman[16] was the first to apply the techniques well known in theories of crystalline dislocations to the problem of the self-energy $E(R)$ for a smectic dislocation loop. Translated into the present notation he quotes the following general result for an edge dislocation loop with unit Burgers vector and zero external stress

$$E(R) = a2^{-1} \int_L \int \phi_3 \, dA, \qquad (6.2)$$

where the integral is over the area of the loop. Although Eq. (6.2) does not explicitly include the core energy of the dislocation, this can be implicitly included by a suitable choice of the core radius. In the case of an infinite smectic sample, Kleman obtains the following expression for the self-energy of a circular loop of radius R:

$$E^0(R) = a^2 b \lambda \pi R^2 2^{-1} \int_0^{q_c} J_1(qR)^2 q \, dq, \tag{6.3}$$

where q_c is an arbitrarily chosen cutoff. Presumably this upper cutoff in the q integral corresponds to a lower cutoff for R at the core radius, i.e., $q_c \sim 2\pi \xi^{-1}$ and $R \gtrsim \xi$. The integral can be evaluated to obtain

$$E^0(R) \approx 2\pi\gamma R, \tag{6.4}$$

where the line tension γ is given by

$$\gamma = a^2 b \lambda (2\xi)^{-1}. \tag{6.5}$$

If $\lambda \sim 2\xi$, this is consistent with earlier estimates of the magnitude of γ.[1] It is also clear that ξ can be chosen to include the core energy.

The self-energy for the same dislocation loop in the median plane of a sample contained between parallel plane boundaries with "rigid" boundary conditions[1] (i.e., smectic layers parallel to the boundaries), a distance D apart, and zero external stress (i.e., $\phi_3 \to 0$ far from the loop) can be estimated from Eq. (6.2) using $a\phi_3$ obtained by substitution of Eq. (8.7) of Ref. 1 into the image solution given by Eq. (6.1a) of that same reference. The more general result for an arbitrarily located loop is essentially the same as for this simpler example as long as the loop is not too near the boundaries. On separating out a "line tension" term equal to $E^0(R)$ [Eq. (6.4) above], the self-energy is

$$E(R) = E^0(R) + a^2 b \lambda \pi R^2 \int_0^{q_c} [q^{-1} J_1(qR)^2][2q^2 \exp(-\lambda D q^2)$$

$$\times [1 + \exp(-\lambda D q^2)][1 - \exp(-2\lambda D q^2)]^{-1}\} \, dq. \tag{6.6}$$

If $R \ll (\lambda D)^{1/2}$, $J_1(qR)^2$ can be taken as $(\tfrac{1}{2}qR)^2$ with the result that

$$E(R) - E^0(R) \approx 2\pi\gamma\xi (R^2/\lambda D)^2 \times \text{(factor of order unity)}. \tag{6.7}$$

Under these assumptions, the right-hand side makes a negligible correction to the self-energy calculated for the infinite sample.

On the other hand, if $R \gg (\lambda D)^{1/2}$, the term in curly brackets in Eq. (6.6) can be replaced by a constant value $2(\lambda D)^{-1}$ and for $Rq_c \gg 1$, the remaining integral is approximately $\int_0^\infty x^{-1} J_1(x)^2 \, dx = 4^{-1}$. The final result for $R \gg (\lambda D)^{1/2}$ is

$$E(R) = E^0(R) + 2^{-1} a(ba/D)\pi R^2. \tag{6.8}$$

Note that ba/D is just the absolute value of the stress inside the loop when the region outside the loop is stress free [See Eq. (8.18) of Ref. 1] and πR^2 is the absolute value of the dislocation moment p_3 for this loop. So long as $R \ll (\lambda D)^{1/2}$, the main contribution to $E(R)$ is the "line tension" of the loop. When $R \gg (\lambda D)^{1/2}$, the boundary conditions make it increasingly difficult for the loop to grow. If, however, one applies an external stress ϕ_3^{ext}, the term $-ap_3\phi_3$ [See Eq. (3.20)] must be added to Eq. (6.8) to obtain

$$E(R) = 2\pi\gamma R + 2^{-1} a(ba/D)\pi R^2 - ap_3\phi_3. \tag{6.9}$$

Consider the example of a dislocation loop with $p_3 > 0$ (i.e., $p_3 = +\pi R^2$). Then

$$E(R) = 2\pi\gamma R + a\pi R^2 [ba(2D)^{-1} - \phi_3], \tag{6.10}$$

and so long as $\phi_3 < ba(2D)^{-1}$ or the strain $\nabla_3 u < a/2D$, the minimum value of $E(R)$ is obtained for vanishingly small

loops. For larger values of ϕ_3 there is a critical radius

$$R_c = \gamma a^{-1} [\phi_3 - ba(2D)^{-1}]^{-1}, \tag{6.11}$$

with energy

$$E(R_c) = \pi\gamma^2 a^{-1} [\phi_3 - ba(2D)^{-1}]^{-1}. \tag{6.12}$$

Loops with $R > R_c$ will grow unstably while for $R < R_c$ they will shrink. For the opposite type of loop, i.e., $p_3 < 0$, similar results are obtained except with opposite signs

$$R_c = \gamma a^{-1} [|\phi_3| - ba(2D)^{-1}]^{-1}, \tag{6.13}$$

$$E(R_c) = \pi\gamma^2 a^{-1} [|\phi_3| - ba(2D)^{-1}]^{-1}. \tag{6.14}$$

Equation (6.12) or (6.14) can be substituted directly into the Langer-Fisher result for the nucleation rate [Eq. (6.1)] and one obtains a critical stress ϕ_3^c,

$$|\phi^c| - ba(2D)^{-1} = \pi\gamma^2 [akT \ln(f_0/f_L)]^{-1} \tag{6.15}$$

or a critical strain $\delta D^c/D$ given by

$$|\delta D^c| - 2^{-1} a = \pi D \gamma^2 [abkT \ln(f_0/f_L)]^{-1}. \tag{6.16}$$

The radius R_c corresponding to this value of the critical stress as obtained from Eqs. (6.5), (6.13), and (6.15) is

$$R_c = kT(\pi a^2 b)^{-1} (2\xi/\lambda) \ln(f_0/f_L). \tag{6.17}$$

Since the present analysis is applicable only so long as $R_c \gg \xi$, we require

$$kT \ln(f_0/f_L) \gg \tfrac{1}{2}\pi a^2 b \lambda. \tag{6.18}$$

Following a procedure similar to Langer and Fisher's $f_0 \sim (10^5 \text{ cm sec}^{-1}/2 \times 10^{-7} \text{ cm}) (3 \times 10^{-22} \text{ cm}^3)^{-1} \sim 10^{33}$ sec^{-1} cm^{-3}, and we choose $f_L \sim 10^{-1}$ sec^{-1} cm^{-3}. If $2\pi\lambda \approx a \approx 2 \times 10^{-7}$ cm, we obtain $b \ll 8 \times 10^8$ dyn cm^{-2}. Assuming b less than this, substitution of the expression (6.5) for γ gives

$$|\phi_3^c - ba(2D)^{-1}| \approx b(\lambda/\xi)^2 \{\pi b a^3 [4kT \ln(f_0/f_L)]^{-1}\}. \tag{6.19}$$

Taking $\lambda/\xi \sim \tfrac{1}{2}$, b will have to be of the order of 10^7 dyn cm^{-2} if the critical *strain* is to be less than 10^{-2}. Although this is still a relatively large strain, it does illustrate the conditions under which homogeneous nucleation might be observable.

To emphasize the requirement of small b, consider values greater than defined in Eq. (6.18). With $R_c \lesssim \xi$, Eq. (6.15) cannot be used with confidence to estimate ϕ_3^c. A reasonable guess in that case might be the value ϕ_3 that makes dislocation loops of radius $R \sim \xi$ unstable. Substitution of γ as given by Eq. (6.5) into either (6.11) or (6.13) gives

$$|\phi_3^c - ba(2D)^{-1}| \approx ba\lambda(2\xi^2)^{-1}. \tag{6.20}$$

If $2\pi\lambda \approx a$, the critical *strain* exceeds $\pi(\lambda/\xi)^2$. Using de Gennes' criteria for type-II behavior, i.e., $(\lambda/\xi)^2 > 2^{-1}$, we obtain an unreasonably large value for ϕ_3^c. The conclusion is that smectic-A liquid crystals with values of $b \gtrsim 10^9$ dyn cm^{-2} will certainly not exhibit homogeneous nucleation of unstable dislocations. There are of course the possibilities of other types of instabilities, like, for example, the undulation instabilities under dilative ϕ_3 stresses[15,17] or a stress-induced phase transition from

the smectic A to either the nematic or the smectic C.[18] Discussion of these is beyond the scope of the present article. Smetics with $b \lesssim 10^7$ dyn cm^{-2} might show homogeneous nucleation effects if other instabilities do not occur first.

In principle the elastic properties of cholesterics can also be described in terms of the same equations as for smectics. The equivalent compressional elastic constant can be obtained from the Frank elastic theory. If P is the cholesteric pitch and the three Frank constants are taken equal to K, $b^{eff} \approx 32\pi^2 K(3p^2)^{-1}$. Since typical numbers for b^{eff} are of the order of 10^5 for small pitch cholesterics, unstable growth of thermally nucleated dislocation loops might occur. Unfortunately, the pitch of a cholesteric is at least 10^2 larger than the lattice spacing in a smectic and the criterion derived from $R_c \gg \xi$ [See Eq. (6.18)] is more difficult to satisfy. Nevertheless, unstable growth of dislocation loops in cholesterics is commonly observed.

VII. DISCUSSION

The principal objective of this paper has been elucidation of the effects of dislocations and impurities on the macroscopic mechanical properties of smectic-A liquid crystals. The first conclusion, that smectics will behave as linear elastic materials capable of sustaining certain stresses only so long as the stresses are smaller than some critical value, is not particularly surprising. The same is true of crystals and analogous behavior is also characteristic of superfluids. In the most general circumstances we expect the critical smectic stress to be determined by the number and types of dislocations existing within the sample. In this sense the critical stress depends on the thermomechanical history of the sample. One might hope, however, to produce samples that are self-annealing in that they will naturally exclude defects from their volume. For example, in the case of free smectic films of the type studied by Clark and Meyer,[19] dislocations are attracted to the sample surfaces by the boundary conditions[1] and one does not expect that these samples contain pinned dislocations. Thus free smectic films might very well be used to investigate critical stresses that can be attributed to thermal nucleation of dislocation loops.

The mechanical properties of smectics below critical stresses are sensitive to both sample history (i.e., the number and type of pinned dislocations) as well as impurity concentration. Both dislocations and impurities alter the observable elastic constant associated with changes in layer spacing. However, these effects can only be observed on time scales sufficiently slow that the distributions of both dislocations and impurities can adjust to the externally applied stresses. To put this another way, dislocations and impurities give rise to relaxation processes that must be taken into account in analyzing stress-dependent experiments on smectic liquid crystals. Clark[15] has observed some stress relaxation effects in smectics and these could be interpreted in terms of the effects considered here. For example, when smectics are subjected to dilative stresses above some critical value, strong quasielastic light scattering with rather unique angular properties

is observed.[15,17] Supposing that the effect of the stress is to induce nonequilibrium values of either P_3 or $Q_{ij} = Q\delta_{ij}^{\perp}$, the condition $\nabla_i \phi_i = 0$ applied to Eqs. (3.23) would give

$$u(\mathbf{q}) = a(q_3^2 + \lambda^2 q_\perp^4)^{-1}[iq_3\delta P_3(\mathbf{q}) - q_\perp^2\delta Q(\mathbf{q})]. \qquad (7.1)$$

In the hydrodynamic limit $\nabla_\perp u$ describes the local rotation in the uniaxial smectic axis and $q_\perp u(\mathbf{q})$ is simply related to a local inhomogeneity in the off-diagonal components in the optical dielectric constant. Previous interpretations of the above-mentioned light scattering have all been in terms of expressions like either Eq. (4.1) or its spatial Fourier transform. The essential ingredient in all the explanations was always the factor $(q_3^2 + \lambda^2 q_\perp^4)^{-1}$ that is sharply peaked at $q_3 \ll q_\perp$. We see here that either new or changed dislocation loops, perturbed impurity distributions, or even microscopic focal conic defects with moments p_3 or Q_{ij} will give rise to the observed effects. Further, these defects can decay away giving rise to the observed relaxation effects. Some of the mechanical properties of smectics have been studied by Brillouin scattering.[20] However, the elastic constants derived from these experiments are high-frequency results and could differ considerably from low-frequency elastic constants. Experiments to investigate the frequency dependence of smectic elastic properties are clearly called for and these should be done for samples of varying impurity content. As yet, there is no satisfactory explanation for the difficulty in observing the Brillouin signal due to shear sound waves in smectics when the propagation direction is such that shear sound couples to local splay. Relaxation effects associated with δQ_{ij} have the correct symmetry to explain this.

Impurities like those schematically illustrated in Fig. 2(b) posses the symmetry that would also allow them to have electric dipole moments. Thus elastic effects related to moments Q_{ij} can also couple to external electric fields giving rise to flexoelectric effects.[21-23] This phenomenon has not been treated in the present paper. However, it does suggest that nonvanishing values of Q_{ij} should not be exclusively attributed to impurities; rather, inhomogeneous distributions of molecular orientations can also give rise to nonvanishing values of Q_{ij} if individual molecules lack a center of symmetry. Relaxation effects due to nonequilibrium values of δQ_{ij} will be nonhydrodynamic in the sense that relaxation times will be finite even for infinitely long wavelengths. Since δQ_{ij} has the appropriate symmetry to couple directly to splay distortions $(\nabla_\perp^2 u \sim \nabla_\perp \circ \mathbf{m}_\perp)$ even in the absence of electric dipole effects, the relaxation phenomena observed in the ripple instability experiments[15,17] could have a component due to δQ_{ij} relaxation.

The impurity effects discussed here may also have some biological significance in that both the rods and cones of the eye and the chloroplasts of plants have biological membranes that organize themselves in a lamellar structure similar to that of smectics. Both of these systems function through active participation of specific macromolecular ingredients that might be regarded as impurities in the lamellar structure. One cannot discount the possibility that stress-impurity inter-

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actions are essential to function in these systems. For example, in the eye the primary physical process associated with "rod vision" is the photoinduced isomerization of a rhodopsin molecule. The resultant change in either p_3 or Q_{ij} of the isomerized molecule changes the mechanical equilibrium in the rod and this physical fact could be significant. In a broader sense the stress patterns associated with impurities give rise to long-range elastic forces between impurities that could play a role in facilitating chemical interactions in lamellar structures. For example, we can ask whether or not reactions involving three separate chemical ingredients depend exclusively on random diffusion to bring the three ingredients together. If diffusion is not sufficient, there is the possibility that a reaction like $A + B \rightleftharpoons AB$ induces a local stress whose long-range effect is to attract a C molecule facilitating further reaction. This mechanism might be invoked to explain reactions like $A + C \rightleftharpoons AC$ which only function efficiently at low A, C concentrations in the presence of some species B. These last ideas are obviously rather speculative and the only purpose in presenting them here is to encourage their active examination. In our own laboratory we are studying the effects of impurities on the mechanical properties of lyotropic smectic liquid crystals in order to obtain more quantitative data on the magnitudes and rates of some of the phenomena discussed here.[24]

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