ALIGNMENT-INVERSION WALLS IN NEMATIC LIQUID CRYSTALS IN THE PRESENCE OF A MAGNETIC FIELD

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It is shown theoretically that nematic liquid crystals in magnetic fields may display alignment inversion walls partially similar to Bloch and Néel walls in ferromagnetics.

We wish to present theoretical considerations indicating that in a magnetic field nematic liquid crystals may display sharp, planar texture irregularities. The structures would loosely resemble the well-known Bloch and Néel walls of ferromagnetic materials. Their essential feature is the gradual reversal of the alignment within a thin boundary layer between uniformly oriented regions. Some of the observations recently made by Williams[1] on nematic p-azoxyanisole can probably be explained in terms of these structures which we will call alignment inversion walls.

Nematic liquid crystals are uniaxial materials. Their preferred axis may vary from place to place, being in general a continuous function of position. The axis coincides with the long axes of the molecules (or rather with the average orientation of these axes). No nematic materials are known, at least to date, which possess a spontaneous magnetic or electric polarization. The preferred axis is, therefore, nonpolar and can be reversed without changing the physical situation. This contrasts with ferromagnetics where a reversal of the orientation changes the sign of the polarization.

The orientation pattern of nematic liquid crystals is in its state of lowest mechanical energy when it is uniform. However, uniformity is not usually observed. It is often prevented by an irregular boundary alignment imposed, for instance, by the container walls. In thick samples (>0.1-1 mm in p-azoxyanisole[2]) it seems to be destroyed by convection due to minute temperature differences.[3] A more or less uniform alignment can usually be achieved by applying a magnetic field of a several hundred oersteds or above.[4] The anisotropy of the molecular diamagnetic susceptibility of nematic materials is generally such that the preferred axis tends to align parallel to the field.

Let us consider a nematic substance in a homogeneous magnetic field. We assume that for some reason the alignment is reversed by 180° in a plane between two regions aligned parallel to the field, the reversal taking place in a gradual fashion in accordance with continuum theory. The orientation pattern in the wall will be such that the energy associated with the reversal assumes a minimum. The energy is composed of a magnetic part due to the misalignment in the field and of a mechanical part arising from the distortion of the orientation pattern. Because of their energy, alignment inversion walls will not in general be stable structures. However, in certain cases they may not only form but be stable or metastable, as is to be discussed below. The orientation pattern of the wall may also be viewed as a local equilibrium between field-induced and distortional torques per unit volume. The magnetic and, therefore, the mechanical torques must everywhere be perpendicular to the applied field.

In order to develop a theory of alignment inversion walls we first introduce a right-handed Cartesian coordinate system x, y, z with the z axis parallel to the magnetic field. The local alignment may be described by the two angles φ and θ. $\sin\theta\sin\varphi$, $\sin\theta\cos\varphi$, and $\cos\theta$ are the projections of the preferred axis, represented by a unit vector, on the x, y, and z axes, respectively. We use Frank's[5] formula for the distortional Gibbs free energy g per unit volume:

$$g=\tfrac{1}{2}k_{11}(s_1+s_2)^2+\tfrac{1}{2}k_{22}(t_1+t_2)^2+\tfrac{1}{2}k_{33}(b_1^{\,2}+b_2^{\,2})-(k_{22}+k_{24})(s_1s_2+t_1t_2). \qquad (1)$$

The splays (s), twists (t), and bends (b) denote the distortions, while k_{11}, k_{22}, k_{33}, and k_{24} are the elastic moduli. We substitute ξ, η, ζ for Frank's local right-handed coordinates x, y, z. The ζ direction coincides with the preferred axis which has an artificial polarity depending on the sign of $\cos\theta$. The ξ and η axes are chosen normal and parallel, respectively, to a plane containing the z and ζ directions. Using φ and θ (so that Frank's ∂L_x and ∂L_y become $\sin\theta\partial\varphi$ and $\partial\theta$) one has

$$s_1=\sin\theta(\partial\varphi/\partial\xi),\quad s_2=\partial\theta/\partial\eta,\quad t_1=-\partial\theta/\partial\xi,$$
$$t_2=\sin\theta(\partial\varphi/\partial\eta),\quad b_1=\sin\theta(\partial\varphi/\partial\zeta),\quad b_2=\partial\theta/\partial\zeta.$$

Deriving the distortional torque per unit volume, m_d, from (1) we note that $m_{d,\zeta}$ must always vanish. The two other components are[6]

$$m_{d,\xi} = -k_{11}\partial(s_1+s_2)/\partial\eta + k_{22}\partial(t_1+t_2)/\partial\xi - k_{33}\partial b_2/\partial\zeta,$$

$$m_{d,\eta} = k_{11}\partial(s_1+s_2)/\partial\xi + k_{22}\partial(t_1+t_2)/\partial\eta + k_{33}\partial b_1/\partial\zeta.$$

The last term in (1) is without effect.[6,7] The torque per unit volume due to the magneitc field H has only one component:

$$m_{H,\xi} = \Delta\chi\cos\theta\sin\theta H^2,$$

where H is the field strength and $\Delta\chi$ (>0) the difference between the magnetic susceptibilities parallel and perpendicular to the preferred axis. (The secondary field due to the diamagnetic susceptibility can be neglected.)

The differential equations governing the spatial dependence of φ and θ are

$$m_{H,\xi} + m_{d,\xi} = 0, \quad m_{d,\eta} = 0. \tag{2}$$

Instead of dealing with the general case $\varphi(x,y,z)$, $\theta(x,y,z)$, let us consider plane walls parallel and perpendicular to the magnetic field and guess the solutions. In the first case we choose, without loss of generality, the plane subtended by the y and z axes. The orientation of the preferred axis may then be expected to depend on x only. The boundary conditions are

$$\theta(-\infty) = 0, \quad \theta(+\infty) = \pm\pi,$$
$$d\theta(-\infty)/dx = d\theta(+\infty)/dx = 0, \tag{3}$$

θ being a function of x. A simple solution is obtained by setting $\varphi = 0$ so that t_1 is the only nonvanishing distortion and (2) reduces to

$$\Delta\chi\cos\theta\sin\theta H^2 - k_{22}(d^2\theta/dx^2) = 0. \tag{4}$$

Double integration of this equation yields the following pair of solutions:

$$x = \pm(k_{22}/\Delta\chi H^2)^{1/2}\int_{\pm\frac{1}{2}\pi}^{\theta} d\theta[1-\sin^2(\theta+\tfrac{1}{2}\pi)]^{-1/2}$$
$$= \pm(k_{22}/\Delta\chi H^2)^{1/2}\ln\tan\tfrac{1}{2}\theta \tag{5}$$

if the boundary conditions (3) are taken into account. Rather than plot this function[8] we write down the wall width which may be defined as the half-value width $L_{1/2}$ over which θ changes from $\pm45°$ to $\pm135°$. One finds

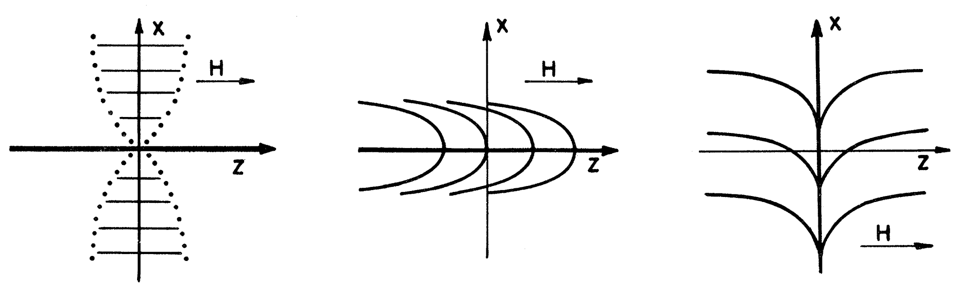
$$L_{1/2} = 1.76(k_{22}/\Delta\chi)^{1/2}H^{-1}.$$

A schematic diagram of the orientation pattern is shown in Fig. 1(a). The ferromagnetic analog is the Bloch wall.

Another pair of solutions of (2) can be obtained by setting $\varphi = \frac{1}{2}\pi$. This eliminates both twists, one splay, and one bend and again ensures $m_{d,\eta} = 0$. The balance of torques becomes

$$\Delta\chi\cos\theta\sin\theta H^2 - k_{11}\cos\theta\frac{d}{dx}\left(\cos\theta\frac{d\theta}{dx}\right) - k_{33}\sin\theta\frac{d}{dx}\left(\sin\theta\frac{d\theta}{dx}\right) = 0. \tag{6}$$

The differential equation has the same simple



(a) (b) (c)

FIG. 1. Schematic diagrams of some alignment inversion walls. (a) Twist wall subtended by y and z axes. Projection of preferred axis (or long molecular axis) on x,y plane is shown. (b) Splay-bend wall parallel to field, subtended by y and z axes. Orientation lines (they are tangential to preferred axis) are shown. (c) Splay-bend wall vertical to field. Orientation lines are shown.

form as (4) if we assume $k_{11}=k_{33}=\tilde{k}$. Then the half-value width is

$$L_{1/2}=1.76(\tilde{k}/\Delta\chi)^{1/2}H^{-1}.$$

The orientation pattern is sketched in Fig. 1(b). It is comparable to that of the Néel wall in ferromagnetics.

The energy per cm² of the two alignment inversion walls is

$$E=\tfrac{1}{2}\int_{-\infty}^{+\infty}k(d\theta/dx)^2dx+\tfrac{1}{2}\int_{-\infty}^{+\infty}H^2\Delta\chi\sin^2\theta dx,$$

k being either k_{22} or $\tilde{k}$, respectively. The first integral of (4),

$$(d\theta/dx)^2=(\Delta\chi H^2/k)\sin^2\theta,$$

shows that the local distortional and magnetic contributions are equal for the considered walls. Accordingly, one has

$$E=\int_0^{\pi}k\,|d\theta/dx|\,d\theta=2(k\Delta\chi)^{1/2}H.$$

The only substance for which the three elastic moduli k_{11}, k_{22}, and k_{33} have been measured is p-azoxyanisole (see below). As $k_{22}<k_{11},k_{33}$ one would expect the Bloch-type wall to be more stable than the Néel-type structure. The following calculation shows, moreover, that pure twist walls parallel to the field represent the solutions of lowest possible energy per cm², including walls not parallel to the field. We use the relation $s_1s_2+t_1t_2=0$, valid if no surface torques are in operation,[6,7] to eliminate t_2. A few simple transformations convert (2) into

$$g=\tfrac{1}{2}k_{22}(s_2^2+t_1^2+b_2^2)+\tfrac{1}{2}(k_{11}-k_{22})(s_1+s_2)^2+\tfrac{1}{2}k_{22}[s_1^2+(s_1^2s_2^2/t_1^2)]+\tfrac{1}{2}(k_{33}-k_{22})b_2^2+\tfrac{1}{2}k_{33}b_1^2.$$

The first term may be given the form

$$\tfrac{1}{2}k_{22}\left[\left(\frac{\partial\theta}{\partial\xi}\right)^2+\left(\frac{\partial\theta}{\partial\eta}\right)^2+\left(\frac{\partial\theta}{\partial\zeta}\right)^2\right]=\tfrac{1}{2}k_{22}(\mathrm{grad}\,\theta)^2.$$

For any alignment inversion wall θ has to change by $\pm\frac{1}{2}\pi$ as one goes from one side to the other. For any path through the wall, as described by the travelled length l, one has

$$g\geq\tfrac{1}{2}k_{22}(\mathrm{grad}\,\theta)^2\geq\tfrac{1}{2}k_{22}(\partial\theta/\partial l)^2,$$

provided $k_{22}<k_{11},k_{33}$. Because of this inequality and since Eq. (5) is the solution of minimum energy for a function θ of only one variable, it is impossible to find a path on which the wall energy is less than $2(\Delta\chi k_{22})^{1/2}H$. This result applies, of course, to the general case where both φ and θ are functions of all three variables x, y, and z, comprising structures that are not solutions of (2).

An alignment inversion wall perpendicular to the magnetic field is sketched in Fig. 1(c). It is characterized by the differential equation

$$\Delta\chi\sin\theta\cos\theta H^2-k_{11}\sin\theta\frac{d}{dz}\left(\sin\theta\frac{d\theta}{dz}\right)-k_{33}\cos\theta\frac{d}{dz}\left(\cos\theta\frac{d\theta}{dz}\right)=0. \quad (7)$$

φ can be chosen arbitrarily but must be constant. A solution of (2) involving twist apparently does not exist. This situation seems to be exceptional. It is readily seen that for plane walls of all other orientations there is just one pair of solutions containing only splay and bend (with $\varphi=\frac{1}{2}\pi$). There is at least one other pair involving twist and, for walls not parallel to the field, the other distortions. The splay-bend solutions are unstable if $k_{22}<k_{11},k_{33}$, as may be seen by varying φ.

In p-azoxyanisole which is nematic between 118 and 135°C one has[9] $k_{11}=0.7\times10^{-6}$ dyn, $k_{22}=0.43\times10^{-6}$ dyn, $k_{33}=1.7\times10^{-6}$ dyn, and $\Delta\chi=1.3\times10^{-7}$ cgs units, taking the values for 120°C. At 500 Oe the width of a twist wall, i.e., Bloch-type wall, is

$$L_{1/2}=0.64\times10^{-2}\text{ cm}.$$

The width of walls involving splay and bend but no twist lies between the values for $\tilde{k}=k_{11}$ and $\tilde{k}=k_{33}$ [cf. Eqs. (6) and (7)]:

$$0.81\times10^{-2}\text{ cm}<L_{1/2}<1.3\times10^{-2}\text{ cm}.$$

We may conclude that at 500 Oe all types of alignment inversion walls in p-azoxyanisole should be about 1×10^{-2} cm thick.

In the experiments of Williams,[1] 0.25- to 1.65-mm thick layers of p-azoxyanisole were spread on a glass substrate. The material was illuminated from the bottom and viewed through a microscope from the top, the magnetic field being parallel to the substrate. At 500 Oe and above Williams observed sharp lines which he interpreted as a kind of grain boundaries. An explanation in terms of alignment inversion walls appears plausible for the following reasons. First, Williams' pictures show that most of the walls, if they were not simply lines, must have stood upright on the substrate. Erectness may be expected because it minimized the area and thus the energy of the wall. Second, the lines were not wider than 1×10^{-2} cm, the width calculated for alignment inversion walls at 500 Oe. Third, no well-defined boundaries were seen below 500 Oe. This seems to indicate that the effects of convection currents prevailed over the aligning force of weak fields. It appears reasonable that the critical wall width is comparable with the distance over which the orientation is usually uniform if no field is applied (see Naggiar's result above).

An alignment inversion wall can be stable or metastable if there are boundary restraints preventing it from moving out of the sample or, if it is cylinderlike, from contracting until it vanishes. A wall may also be stabilized if the sample shape does not allow for a decrease in area by migration. Mainly the first factor was presumably responsible for Williams's results. It is interesting to note that similar considerations are used to explain the stability of disinclinations, the frequently observed linear orientation irregularities characteristic of the nematic mesophase. The formation of alignment inversion walls upon applying the magnetic field requires a strongly nonuniform orientation pattern in the fieldless state. The nonuniformity may be produced by the aforementioned convection currents or by the effect of boundaries. Diffuse inversions performed in this way would become sharp walls when the field is applied.

We wish to point out that we do not attempt to explain the optical activity observed by Williams.[1] However, additional experiments[10] seem to lend further support to our view that he saw alignment inversion walls.

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[10]R. Williams, private communication.

PHASE INCOHERENCE IN THE dc SUPERCONDUCTING TRANSFORMER*

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A model is presented which explains the observed dependence of the coupling between the primary and secondary vortices on the driving current, or primary vortex velocity, in the dc superconducting transformer. The basic idea is that viscous drag effects lead to a slippage between the vortices in the primary film and those in the secondary film for sufficiently large vortex velocities.

The existence of the dc superconducting transformer[1-4] convincingly supports the idea of current-induced vortex motion. In this experiment, two superconducting films sandwich a very thin dielectric layer. One of the metal films is characteristically about twice as thick as the other. The dielectric layer (SiO) electrically insulates the two films. Passing a current through the thicker film (called the primary) results in a force being exerted upon the Abrikosov microstructure[5] established in the film by means of an applied perpendicular magnetic field. As soon as this force is large enough to depin the vortices, they are said to move through the film and a voltage appears across the primary. Since the thinner film (called the secondary) is positioned so close to the primary, the motion of the primary vortices exerts a drag upon the microstructure