# Instability of the axis of a cholesteric liquid crystal in a magnetic field

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The stability of the simplest stationary structures of a sufficiently large cholesteric liquid crystal with positive magnetic susceptibility anisotropy  $\Delta \chi = \chi_{\parallel} - \chi_{\perp}$  in a homogeneous magnetic field is considered. It is shown that a state in which the cholesteric axis is parallel to the magnetic field is stable provided that the angle  $\theta$  of inclination of the director to the axis is less than some critical value  $\theta_c$  (i.e., in the case of a sufficiently strong "conic deformation"); in the opposite case it is unstable. It is also found that the most favorable direction of the cholesteric axis in weak fields, namely the one perpendicular to the field, can become unstable before the cholesteric crystal changes into a nematic.

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#### **1. INTRODUCTION**

Meyer<sup>1</sup> and de Gennes<sup>2</sup> proposed a theory that describes the behavior of a cholesteric liquid crystal in a magnetic field and the field-induced transition from the cholesteric to the nematic phase. The calculations were made for large enough samples of a substance with positive anisotropic magnetic susceptibility<sup>1)</sup>  $\Delta \chi = \chi_{\parallel} - \chi_{\perp}$ , placed in a uniform magnetic field **H** parallel or perpendicular to the cholesteric axis **h**. In the case  $\mathbf{h} || \mathbf{H}$  it followed from the calculations of Ref. 1 that the angle  $\theta$  between the director **n** and the cholesteric axis should decrease with increasing field and vanish at a certain field  $H = H_{\parallel}$ ; for substance with longitudinal bending modulus  $K_3$  exceeding the torsion modulus  $K_2$ , the decrease of  $\theta$  should be jumplike from  $\pi/2$  to zero, and in the opposite case it should be continuous, with formation of the so-called conical deformation. At  $K_2 > K_3$  the pitch of the helix should decrease in inverse proportion to the field intensity. An experimental investigation of the optical activity<sup>3</sup> and of the "blue shift" of the band of selective light reflection of the cholesteric, both corresponding to this decrease, revealed strong deviations from the prediction of the theory. The director deflection served in Ref. 4 was attributed in Ref. 5 to the polycrystalline structure of the sample. It will be shown in this article that even for an ideal crystal one can expect agreement between the theory and the theory of conical deformation only in the angle interval  $0 < \theta < \theta_c$ . The angle  $\theta_c$  depends on the elastic moduli, but is always contained between 45° and arcsin  $(2/3)^{1/2} \approx 55^\circ$ . At  $\theta > \theta c$ the conical deformation is unstable to small perturbations. The regime of nonlinear elimination of this instability turns out to be hard: the conical structure becomes strongly distorted directly after the passage through the instability threshold, and hysteresis should therefore be observed. In particular, the value of  $\theta_c$  obtained below can be approached only by decreasing the magnetic field. It is not excluded that conical deformation in pure form can be obtained simply only by this method.

Besides the stability of a stationary structure with  $\mathbf{h} || \mathbf{H}$  we shall discuss the case  $\mathbf{h} \perp \mathbf{H}$ . In this case it was predicted in Refs. 1 and 2 that the cholesteric helix will be distorted by the magnetic field and its pitch will become infinite in a cer-

tain field  $H = H_1$ . The subsequent experiments (see, e.g., Refs. 9 and 10) have confirmed these predictions. However, as will be shown below, deviations from the theory<sup>2</sup> should be observed for a number of substances. These are due to the instability of the cholesteric axis. A sufficient (but not necessary) condition for such an instability is, for example the inequality  $K_2 > (\pi^2/4)K_3$ . This inequality was satisfied in the experiments,<sup>3</sup> and although their agreement with Ref. 2 was taken to be good, one can note on the plots of the helix pitch against the field intensity in Ref. 3, a difference between the experimental and theoretical curves. More accurate measurements will probably permit a reliable determination of this difference and of the instability threshold.

We note that the cholesteric-axis instabilities discussed here, in contrast to those investigated theoretically<sup>10-12</sup> and experimentally<sup>13-15</sup> before, are due not to competition between the orientational actions of the field and of the sample boundaries, but to the fact that the character of the orientational action of the field on the axis depends on the field itself. This difference manifests itself most distinctly in sufficiently large liquid-crystal samples whose dimensions exceed noticeably the pitch of an ideal cholesteric helix. In such samples, surface forces can compete with magnetic forces only if the field is very weak and has practically no effect on the helix. The effects of interest to us, however, which are connected with a noticeable distortion of the helix, take place in a much stronger field and can therefore be considered here without taking the surface forces into account.

#### **BASIC EQUATIONS**

The free energy of a deformed liquid crystal in a magnetic field is of the form

$$F=\int dVW,$$

$$W = \frac{1}{2} \{ K_1(\operatorname{div} \mathbf{n})^2 + K_2(\mathbf{n} \operatorname{rot} \mathbf{n} + q)^2 + K_3[(\mathbf{n} \nabla) \times \mathbf{n}]^2 - \Delta \chi(\mathbf{n} \mathbf{H})^2 \}$$
(1)

To decrease the number of parameters on which F depends, we shall measure the distance in units of  $a = (K_2/\Delta\chi H^2)^{1/2}$ and the energy density in units of  $\Delta\chi H^2$ . The expression for W is then

;

$$W = \frac{1}{2} \{ R_1(\operatorname{div} \mathbf{n})^2 + (\mathbf{n} \operatorname{rot} \mathbf{n} + qa)^2 + R_3[(\mathbf{n} \nabla) \times \mathbf{n}]^2 - (\mathbf{n} \mathbf{H}/H)^2 \},\$$

$$R_1 = K_1/K_2, \quad R_3 = K_3/K_2.$$
 (2)

Let  $\theta$  and  $\varphi$  be the polar and azimuthal angles of the direction of **n** in a coordinate frame with unit vectors  $\mathbf{e}_x$ ,  $\mathbf{e}_y$ , and  $\mathbf{e}_z$ :

 $\mathbf{n} = \mathbf{e}_x \sin \theta \cos \varphi + \mathbf{e}_y \sin \theta \sin \varphi + \mathbf{e}_z \cos \theta.$ 

Introducing the unit vectors

$$\mathbf{e}_{\varphi} = [\mathbf{e}_{z} \times \mathbf{n}] / \sin \theta = -\mathbf{e}_{z} \sin \varphi + \mathbf{e}_{y} \cos \varphi,$$

$$\mathbf{e}_{\theta} = [\mathbf{e}_{\varphi} \times \mathbf{n}] = \mathbf{e}_{x} \cos \theta \cos \varphi + \mathbf{e}_{y} \cos \theta \sin \varphi - \mathbf{e}_{z} \sin \theta$$

we can represent the free-energy density in the form

$$W = \frac{1}{2} \{ R_1 (\mathbf{e}_{\theta} \nabla \theta + \sin \theta \mathbf{e}_{\varphi} \nabla \phi)^2 + (\sin \theta \mathbf{e}_{\theta} \nabla \phi)^2 \}$$

$$-\mathbf{e}_{\varphi}\nabla\theta + qa)^{2} + R_{3}[(\mathbf{n}\nabla\theta)^{2} + \sin^{2}\theta(\mathbf{n}\nabla\varphi)^{2}] - (\mathbf{n}\mathbf{H}/H)^{2}\}.$$
 (3)

The form of (3) allows us to take into account explicitly the director unit length, thus facilitating the determination of the stationary states: they are given by the externals of the functional F:

 $\delta F/\delta \theta = 0, \quad \delta F/\delta \phi = 0.$ 

The necessary and sufficient condition for the stability of the stationary state  $\mathbf{n}^0 = \mathbf{n}(\varphi^0, \theta^0)$  to small perturbation is that the second variation of the functional F on this state be positive-definite.

### 3. CHOLESTERIC AXIS PARALLEL TO MAGNETIC FIELD

A stationary state with  $\mathbf{h}$  ||H was found in Ref. 1. Directing the z axis along the field, we can represent this state in the form

$$\theta^{\circ} = \text{const}, \quad \varphi^{\circ} = \dot{\varphi}z, \quad \dot{\varphi} = qa[\sin^2\theta^{\circ} + R_s\cos^2\theta^{\circ}]^{-1}.$$
 (4)

If  $R_3 < 1$ , the constant  $\theta^0$ , which minimizes the free energy on the class of distributions (4), is specified by the relations

The critical field  $H_{\parallel}$  at which the cholesteric is transformed into a nematic is determined from the condition

$$q^{2}a_{\parallel}^{2} = R_{s} \quad (q^{2}K_{2}^{2} = K_{s}\Delta\chi H_{\parallel}^{2}).$$
 (6)

If  $R_3 > 1$ , then  $\theta^0 = 0$  at  $H > R_3 H_{\parallel}$ ,  $\theta^0 = \pi/2$  at  $H < H_{\parallel}$ , and in the field interval  $H_{\parallel} < H < R_3 H_{\parallel}$  the free energy has on the class of distributions (4) two minima,  $\theta^0 = 0$  and  $\theta^0 = \pi/2$ , with the lower energy corresponding to a state with  $\theta^0 = 0$  if  $H > R_3^{1/2} H_{\parallel}$  and to the state with  $\theta^0 = \pi/2$  in the opposite case.

We put in (3)

 $\theta = \theta^{0} + \theta', \quad \varphi = \varphi^{0} + \varphi'$ 

and confine ourselves to terms quadratic in  $\theta'$  and  $\varphi'$ . As a result we obtain an expression for the second variation  $F_2$  of the functional F on the stationary state  $\theta^0$ ,  $\varphi^0$ . With the aid of this expression we readily find that the state with  $\theta^0 = 0$  is stable at  $H > H_{\parallel}$  and unstable at  $H < H_{\parallel}$ , while the state with  $\theta^0 = 0$  is stable at  $H > H_{\parallel}$  and unstable. Since no possibilities other than  $\theta^0 = 0$  and  $\theta^0 = \pi/2$  exist for  $R_3$ , it remains to investigate the stability at  $R_3 < 1$  in the field interval  $R_3 H_{\parallel} < H < H_{\parallel}$ , where

conical deformation exists. To this end it is convenient to improve the choice of units of length and energy by making the changes

$$\mathbf{r} \rightarrow \dot{\phi}^{-1}\mathbf{r}, \quad W \rightarrow \dot{\phi}^2 W.$$

Since  $F_2$  is invariant to shifts and rotations in the x,y plane, it can be assumed without loss of generality that at the instability threshold perturbations are excited of the form

$$\varphi' = (\xi_1 e^{i\varphi x} + c.c.) / \sin \theta^0, \quad \theta' = \xi_2 e^{i\varphi x} + c.c.$$

Taking into account the second relation of (5), we obtain for these perturbations

$$F_{2} = \int dV W_{2} = V \overline{W}_{2}, \quad \overline{W}_{2} = \langle \xi | \hat{L} | \xi \rangle$$

$$= \left\langle R_{i} \left| -\frac{d\xi_{2}}{dz} \sin \theta^{0} + i\rho (\xi_{1} \cos \theta^{0} \cos z - \xi_{1} \sin z) \right|^{2}$$

$$+ \left| -\frac{d\xi_{1}}{dz} \sin \theta^{0} + i\rho (\xi_{1} \cos \theta^{0} \cos z + \xi_{2} \sin z) - \xi_{2} \sin 2\theta^{0} \right|^{2}$$

$$+ R_{3} \left[ \left| \frac{d\xi_{2}}{dz} \cos \theta^{0} + i\rho\xi_{2} \sin \theta^{0} \cos z \right|^{2}$$

$$+ \left| \frac{d\xi_{1}}{dz} \cos \theta^{0} + i\rho\xi_{1} \sin \theta^{0} \cos z \right|^{2}$$

$$+ 2\operatorname{Re} \left( -\xi_{2} \cdot \frac{d\xi_{1}}{dz} \sin 2\theta^{0} + i\rho\xi_{2} \cdot \xi_{1} \cos 2\theta^{0} \cos z \right)$$

$$\times \sin \theta^{0} - |\xi_{2}|^{2} \sin^{2} 2\theta^{0} \right] \right\rangle.$$
(7)

The angle brackets denote here averaging over z; further,  $\xi$  is a two-component vector and L is a matrix Hermitian operator that depends on four parameters  $(R_1, R_3, \theta^0, \rho)$ :

$$\xi = \begin{pmatrix} \xi_1 \\ \xi_2 \end{pmatrix}, \quad \hat{L} = \begin{pmatrix} \hat{L}_{11} & \hat{L}_{12} \\ \hat{L}_{21} & \hat{L}_{22} \end{pmatrix} = \hat{L}_0 + i\rho \hat{L}' + \rho^2 \hat{L}''.$$

The operators  $\hat{L}_0$ ,  $\hat{L}'$ ,  $\hat{L}''$  do not depend on  $\rho$ .

The necessary and sufficient condition for the stability of conical deformation to small perturbation is that  $\hat{L}$  be positive definite at all values of  $\rho$ . At the stability threshold [which must exist in our case, since conical deformation with  $\theta^{0} \rightarrow + 0 (H \rightarrow H_{\parallel} - 0)$  is stable, and one with  $\theta^{0} \rightarrow (\pi/2) - 0$  $(H \rightarrow R_{3}H_{\parallel} + 0)$  is not],  $\theta^{0}$  is a function of  $R_{1}$  and  $R_{3}$ :

$$\theta^{0} = \theta_{c}(R_{1}, R_{3}).$$
(8)

The critical field  $H_c$  is uniquely connected with  $\theta_c$  by the relations (4) and (5):

$$H_{c} = \frac{q}{\sin^{2}\theta_{c} + R_{s}\cos^{2}\theta_{c}} \left(\frac{K_{s}}{\Delta\chi}\right)^{4/s}.$$
(9)

Certain conclusions concerning the function  $\theta_c(R_1,R_3)$ can be drawn without calculations. Thus, for example, since the first term of (7) is positive and the last term is negative at the instability threshold,  $\theta_c$  is an increasing function of  $R_1$ and a decreasing one of  $R_3$ . The highest and lowest values of  $\theta_c$  on the segment  $0 < R_3 < 1$ ,  $R_1 = \text{const}$ , are reached respectively at  $R_3 \rightarrow 0$  and  $R_3 = 1$ . In the case  $R_3 \rightarrow 0$  the only danger to the stability are perturbations that cause the first two terms of (7) to vanish. From the exact equality of these terms to zero follows the relation

$$\sin \theta^{\circ} \frac{d}{dz} \xi_{1} \xi_{2} = i \rho \left( |\xi_{1}|^{2} + |\xi_{2}|^{2} \right) \sin z - |\xi_{2}|^{2} \sin 2\theta^{\circ},$$

with the aid of which it is easy to verify that such an equality is possible only at

$$\xi \sim \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \rho = 0.$$

An approximate equality is possible as  $\rho \rightarrow 0$ . For the most dangerous perturbations

$$\xi = \begin{pmatrix} \sin \theta^{0} - i\rho \cos \theta^{0} \sin z \\ i\rho \cos z \end{pmatrix}$$
(10)

so that the first two terms in (7) are proportional to  $\rho^4$ . Substituting (10) in (7) and leaving out terms of order  $\rho^4$  we obtain

$$\overline{W}_2 = R_3 \rho^2 (1 - \frac{3}{2} \sin^2 \theta^0),$$

from which follows

$$\max_{R_3} \sin^2 \theta_c = \sin^2 \theta_c |_{R_3 \to 0} = \frac{2}{3}.$$
 (11)

In the opposite limiting case  $(R_3 = 1)$  we assume first that  $R_1$  is also equal to unity. In the single-constant approximation  $(R_1 = R_3 = 1)$  the operator  $\hat{L}$  simplifies to the limit:

$$\hat{L}_{11} = \hat{L}_{22} = -d^2/dz^2 + \rho^2, \quad \hat{L}_{21} = \hat{L}_{12}^+ = -2i\rho\sin\theta^0\cos z.$$
 (12)

The substitution  $\xi_2 \rightarrow i \xi_2$  makes the operator (12) real, so that its eigenfunctions can be chosen in the form

$$\xi = \left( \begin{array}{c} \eta_1 \\ i \eta_2 \end{array} \right)$$

with real  $\eta_1$  and  $\eta_2$ . The equations for the functions  $\eta_1$  and  $\eta_2$ do not change when the subscript 1 and 2 are interchanged, so that we can put, without loss of generality,  $\eta_1 = \eta_2$  or  $\eta_1 = -\eta_2$ . Actually it suffices to consider one of these cases, since each reduces to the other by the replacement  $z \rightarrow z + \pi$ . Putting

$$\xi = \left( \begin{array}{c} \eta \\ i\eta \end{array} \right),$$

we obtain for  $\eta$  the well-investigated Mathieu equation:

$$(-d^2/dz^2+\rho^2-2\rho\sin\theta^0\cos z)\eta=\lambda\eta$$

With the aid of tables or plots (see, e.g., Ref. 16) it is easy to ascertain that at the instability threshold there are excited long-wave  $(\rho \rightarrow 0)$  perturbations, periodic in z, for which

$$\eta = 1 + 2\rho \sin \theta^{\circ} \cos z + O(\rho^2), \quad \lambda = \rho^2 \cos 2\theta^{\circ} + O(\rho^4).$$

Consequently,

$$\theta_c|_{R_1=R_3=1}=\pi/4.$$

Since  $\theta_c$  is an increasing function of  $R_1$ , we have

$$\theta_c|_{R_3=1, R_1 \ge 1} \ge \theta_c|_{R_1=R_3=1} = \pi/4.$$
(13)

(14)

On the other hand, putting in (7)  $R_3 = 1, \rho \rightarrow 0$ , and

$$\boldsymbol{\xi} = \begin{pmatrix} \sin \theta^{\alpha} \\ i\rho \cos z \end{pmatrix},$$

we obtain  $W_2 = \frac{1}{2}\rho^2 \cos 2\theta^0$ , whence

 $\theta_c|_{R_{s=1}} \leq \pi/4.$ 

The inequalities (13) and (14) are compatible only at

$$\Theta_{c}|_{R_{3}=1, R_{1}\geq1}=\pi/4.$$
(15)

In all the particular cases considered above, long-wave perturbations  $(\rho \rightarrow 0)$  were excited at the instability threshold. If this property is preserved in the general case, the instability threshold can be obtaned analytically. Indeed, at  $\rho = 0$  the operator  $\hat{L}$  is positive-definite, as is clear without calculations, since the solution (4), (5) minimizes the free energy on the class of director distributions that depend only on z. The smallest eigenvalue that  $\hat{L}$  has at  $\rho = 0$  is zero and corresponds to the eigenmode<sup>21</sup>  $\xi^0 = {1 \choose 0}$ . As  $\rho \rightarrow 0$  the smallest eigenvalue  $\lambda (\rho, \theta^0, R_1, R_3)$  of the operator  $\hat{L}$  corresponds to a mode close to  $\xi^0$  and can be obtained by perturbation theory:

$$\lambda = \lambda_1 \rho^2 + \lambda_2 \rho^4 + \dots \tag{16}$$

The coefficient  $\lambda_1$  vanishes on the surface  $\theta^0 = \theta_c(R_1, R_3)$ , defined by the equation

$$\sin^{2} \theta_{c} = [2R_{1}(1-2R_{3})-R_{3}(1+R_{3}) + \{[2R_{1}(1-2R_{3})-R_{3}(1+R_{3})]^{2} + 4R_{3}(1-R_{3})(1+R_{1})(3R_{1}+R_{3})\}^{\frac{1}{2}}][2(1-R_{3})(R_{3}+3R_{1})]^{-1}.$$
(17)

In accord with the statements made above,  $\sin^2\theta_c$  increases with increasing  $R_1$ , decreases with increasing  $R_3$ , and can take on values from the interval (1/2; 3/2), the minimum being reached on the straight line  $R_3 = 1$  and the maximum as  $R_3 \rightarrow 0$ . The coefficient  $\lambda_2$  turns out to be positive on the surface  $\theta^0 = \theta_c(R_1, R_3)$ . From this and from (16) it follows that perturbations with  $\rho \ll 0$  are the most unstable of the perturbations with  $\rho \ll 1$  (the only one for which the expansion (16) is valid). Perturbations with  $\rho \gg 1$  can easily be shown to be stable at all values of  $R_1$ ,  $R_3$ , and  $\theta^0$ . All this confirms the assumption that the instability is long-wave.

At the instability threshold are excited the perturbations

$$\begin{pmatrix} \varphi' \sin \theta^{\circ} \\ \theta' \end{pmatrix} \sim \begin{pmatrix} 1 \\ 0 \end{pmatrix} \cos \rho x + \begin{pmatrix} b_{1} \sin z \\ b_{2} \cos z \end{pmatrix} \rho \sin \rho x + \begin{pmatrix} c_{1} \cos 2z \\ c_{2} \sin 2z \end{pmatrix} \rho^{2} \cos \rho x + \dots$$
(18)

The coefficients  $b_i$ ,  $c_i$ , etc. are explicitly calculable functions of  $R_1$ ,  $R_2$ , and  $\theta^0 \approx \theta_c$ . Their estimated value is unity. Equation (18) is applicable only in the linear approximation in the perturbation amplitude. To ascertain how strongly the state of the cholesteric is changed on going through the instability threshold it is necessary to take into account the nonlinearity. The general form of perturbations of finite but small amplitudes, which appear near the threshold, is

$$\begin{pmatrix} \varphi' \sin \theta^{\circ} \\ \theta' \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \frac{f(\varepsilon x)}{\varepsilon} - \begin{pmatrix} b_{1} \sin \zeta \\ b_{2} \cos \zeta \end{pmatrix} f'(\varepsilon x) - \begin{pmatrix} c_{1} \cos 2\zeta \\ c_{2} \sin 2\zeta \end{pmatrix} \varepsilon f''(\varepsilon x) + \begin{pmatrix} d_{1} \sin 2\zeta \\ d_{2} + d_{2} \cos 2\zeta \end{pmatrix} [f'(\varepsilon x)]^{2} + \dots$$
(19)

Here  $\varepsilon \lt 1$  is a small parameter connected with the long-wave character of the instability,  $\zeta = z + f(\varepsilon x)/\varepsilon$ , and the prime

denotes differentiation of the trial function f with respect to its argument. The expansion in (19) is in powers of f and  $\varepsilon$ , and the ratio  $f/\varepsilon$  may also not be small. After substituting the expansion (19) in the exact expression for the free energy, the latter takes the form

$$F = \operatorname{const+}^{1}/_{2} \int dV \{\lambda_{1}(f')^{2} + \varepsilon^{2} \lambda_{2}(f'')^{2} + A(f')^{4} + \dots \}.$$

Calculation shows that the coefficient  $A(R_1, R_3)$  is negative. Consequently the nonlinearity exerts a destabilizing action on the conical deformation, and the state of the cholesteric is strongly changed on going through the instability threshold. Since the field  $H_c$  is strong enough to "crush" the helix, the equilibrium state of the cholesteric after passing through the threshold (i.e., at  $H < H_c$ ) can turn out to be quite complicated-the concept of cholesteric axis may have in this state no meaning even locally. It is not excluded, however, that directly after the destruction of the conical deformation the cholesteric arrives, after going through a sequence of nonequilibrium and complicated states at a state with a cholesteric axis perpendicular to the field. We note that such a state certainly sets in starting with a certain field  $H'_{c}$  (see the beginning of the next section). The question we leave open is whether  $H'_c$  coincides with  $H_c$ , and if not, through what equilibrium states does the cholesteric go through when the field is decreased from  $H_c$  to  $H'_c$ .

## 4. CHOLESTERIC AXIS PERPENDICULAR TO MAGNETIC FIELD

In a sufficiently weak magnetic field that causes practically no distortion of the shape of the cholesteric helix,<sup>3)</sup> the stationary state with  $h\perp H$  turns out to be energetically most favored, since the liquid crystal averaged over the helix turn has negative anisotropy of the magnetic susceptibility relative to the cholesteric axis (see, e.g., Ref. 17). In a stronger magnetic field the stability of the stationary state obtained in Ref. 2, with a cholesteric axis perpendicular to the field, is not obvious. Directing the x axis along H and the z axis along h, this state can be represented in the form

$$\theta^{\circ} = \pi/2, \quad \varphi^{\circ} = \pi/2 + \operatorname{am}(z/\varkappa, \varkappa).$$
 (20)

Here am is the Jacobi elliptic amplitude and  $\varkappa$  is the root of the equation

$$(2/\pi)E(\varkappa) = \varkappa qa, \tag{21}$$

where E is a complete elliptic integral of the second kind. The function  $\varphi^0$  minimizes the free energy on a class of distributions, that depend only on z, of the director with  $\theta = \pi/2$ . The minimum value of the average energy density is

$$\overline{W} = \frac{1}{2} (a^2 a^2 - \frac{1}{\kappa^2})$$

Equation (21) has a root at  $H < H_{\perp}$ , where  $H_{\perp}$  is determined from the condition  $\varkappa \rightarrow 1 - 0$ , i.e.,

$$\frac{\pi}{2} q a_{\perp} = 1 \quad \left( \Delta \chi H_{\perp}^2 = \frac{\pi^2}{4} K_2 q^2 \right).$$
 (22)

As  $H \rightarrow H_{\perp} - 0$  the pitch of the helix becomes infinite. In fields exceeding the critical value, the nematic state is energetically favored. It corresponds to an average energy density  $\overline{W} = \frac{1}{2}(q^2a^2 - 1)$ . The same average energy density cor-

responds to the deformation (20) with x = 1, which can be called a "twist soliton":

$$\Theta^{\circ} = \pi/2, \quad \varphi^{\circ} = 2 \operatorname{arct} g e^{z}.$$
 (23)

However, the energy per unit area in the (x,y) plane is larger for a twist soliton than for a nematic state, by an amount  $\pi qa(H/H_{\perp} - 1)$ .

We examine now the stability of the stationary state (20) at arbitrary magnetic field intensity. To this end it is convenient to improve the choice of the units of length and energy by making the substitutions

$$\mathbf{r} \rightarrow \varkappa \mathbf{r}, \quad W \rightarrow \varkappa^{-2} W.$$

By virtue of the invariance of the second variation  $F_2 = \int W_2 dV$  of the free energy on the state (20) relative to shifts in the (x, y) plane one can assume, without loss of generality, that at the instability threshold (if it exists<sup>4</sup>) are excited perturbations of the form

$$\varphi' = \varphi - \varphi^{0} = \xi_{1} e^{i\rho \mathbf{r}} + \text{c.c.}, \quad \theta' = \theta - \theta^{0} = i\xi_{2} e^{i\rho \mathbf{r}} + \text{c.c.},$$

$$\varphi = \varphi(\mathbf{e}_{\mathbf{r}} \cos \alpha + \mathbf{e}_{\mathbf{r}} \sin \alpha), \quad \alpha = \text{const}, \quad \rho = \text{const}.$$
(24)

For these perturbations

$$\begin{split} \overline{W}_{2} &= \langle R_{1} | d\xi_{2} / dz + \rho \xi_{1} \sin (\varphi^{0} - \alpha) |^{2} + | d\xi_{1} / dz + \rho \xi_{2} \sin(\varphi^{0} - \alpha) |^{2} \\ &+ 2 (g - \ln z) \left[ |\xi_{2}|^{2} \ln z - \rho (\xi_{1} \xi_{2}^{*} + \xi_{1}^{*} \xi_{2}) \cos (\varphi^{0} - \alpha) \right] \\ &+ R_{3} \left[ |\rho \xi_{1} \cos (\varphi^{0} - \alpha) - \xi_{2} \ln z |^{2} + \rho^{2} |\xi_{2}|^{2} \cos^{2} (\varphi^{0} - \alpha) \right] \\ &+ \kappa^{2} (|\xi_{1}|^{2} \cos 2\varphi^{0} + |\xi_{2}|^{2} \cos^{2} \varphi^{0}) \rangle \equiv \langle \xi | L | \xi \rangle. \end{split}$$

$$(25)$$

The angle brackets denote here averaging over z, and

$$dn z = (1 - \varkappa^2 \operatorname{sn}^2 z)^{\frac{1}{2}}, \quad \operatorname{sn} z = \sin \operatorname{am} z = -\cos \varphi^0$$

are Jacobi elliptic functions;  $g = \chi q a$  is an auxiliary parameter uniquely connected with the magnetic field intensity;  $\xi$ is a two-component vector and  $\hat{L}$  is a Hermitian matrix operator that depends on the five parameters:  $R_1, R_3, g, \rho, \alpha, \varkappa$  is a function of g), and

$$\boldsymbol{\xi} = \begin{pmatrix} \boldsymbol{\xi}_{1} \\ \boldsymbol{\xi}_{2} \end{pmatrix}, \quad \hat{\boldsymbol{L}} = \begin{pmatrix} \hat{\boldsymbol{L}}_{1} + \rho^{2} \boldsymbol{\mathcal{L}}_{1}, & -\rho \hat{l}^{+} \\ -\rho \hat{l}, & \hat{\boldsymbol{L}}_{2} + \rho^{2} \boldsymbol{\mathcal{L}}_{2} \end{pmatrix}$$

The necessary and sufficient condition for the stability of the stationary (20) to small perturbation is that  $\hat{L}$  be positivedefinite at all values of  $\rho$  and  $\alpha$ . The stability threshold is specified by a certain function

$$g=g_{*}(R_{1},R_{3}). \tag{26}$$

Knowing  $g_s$ , we can use the formula

$$H_{s} = \left(\frac{K_{2}}{\Delta \chi}\right)^{\frac{1}{2}} \frac{q_{\varkappa}(g_{s})}{g_{s}}$$
(27)

to calculate the critical field, which is larger than smaller g. Inasmuch as with increase of the parameters  $R_1$  and  $R_3$  the functional  $F_2$  increases and the stability region broadens,  $g_s$  is a decreasing function of  $R_1$  and  $R_3$ . The stability condition is most rigid at  $R_1 < 1, R_3 < 1$  and least stringent at  $R_1 > 1, R_3 > 1$ .

Let  $R_1 < 1$  and  $R_3 < 1$ . Then at  $\varkappa^2 < R_1, R_3$  the necessary stability condition is violated: the operator  $\hat{L}_2$  is not possitive-definite. On the contrary, at  $\varkappa^2 < R_1, R_3$  the state (20) is stable, since the reasoning presented at the beginning of the section is valid. The instability threshold lies in the region  $x^2 \ll 1$ , where

$$g \approx 1 - \frac{\kappa^2}{4}$$
,  $\operatorname{dn} z \approx g - \frac{\kappa^2}{4} \cos 2\varphi^0$ .

The perturbations excited near the threshold cause vanishing of the second term in (25). Therefore the stability criterion contains in fact only the ratios  $R_1/R_3$ ,  $\varkappa^2/R_3$  and the critical value of  $\varkappa^2$  is given by a function of the form

$$R_{s}f(R_{i}/R_{s}) = \varkappa_{s}^{2} \approx 4(1-g_{s}). \qquad (28)$$

In the case  $R_1 > 1, R_3 > 1$  the only perturbations dangerous to the stability are

$$\xi = c \left( \frac{\mathrm{dn} z}{\rho \cos(\varphi^0 - \alpha)} \right), \quad \rho \to 0,$$

which cause the vanishing of the first and fourth terms in (25). However, even these perturbations turn out to be stable at all values of g. We note that considerable progress in investigation of the stability of perturbations with  $\rho \rightarrow 0$  is possible at arbitrary  $R_1$  and  $R_3$  through the use of the neutrally stable mode  $\xi \propto {\binom{dn}{0}}$  which exists at  $\rho = 0$ . Now, however, in contrast to the case  $\mathbf{h} || \mathbf{H}$ , such an investigation does not cover the problem exhaustively, since perturbations with  $\rho \rightarrow 0$  are generally speaking not the most dangerous.

Thus, depending on the relation between the elastic moduli of the liquid crystal, the stationary state (2) can either become unstable in fields weaker than  $H_1$ , or remain stable in arbitrary fields in which the initial equation for the free energy is valid. If the values of the parameters  $R_1$  and  $R_2$  (the only ones on which the ratio  $H_s/H_1$  depends) are such that  $H_s > H_1$ , the predictions of Ref. 2 should hold all the way to the transformation of the cholesteric into a nematic. If  $H_s < H_1$ , the instability of the cholesteric axis in the field interval  $H_s < H < H_1$  should cause deviations from the theory of Ref. 2. It appears that such deviations have apparently not been reliably recorded as yet, although for the substance used in Ref. 3, with  $R_3 \approx 1/4$ , the condition  $H_s < H_1$  was certainly satisfied. The latter is easiest to verify by noting that in the case

$$H_{\parallel} > H_{\perp} \qquad (R_{s} < 4/\pi^{2})$$
 (29)

the state (20) cannot remain stable as  $H \rightarrow H_{\perp} = 0$ . Indeed, as  $H \rightarrow H_{\perp} = 0$  it goes over into a twist soliton, for which  $\mathbf{n} || \mathbf{H}$  in an overwhelming fraction of the volume. On the other hand the state with  $\mathbf{n} || \mathbf{H}$ , as shown in Sec. 3, is unstable at  $H < H_{\parallel}$ . The instability criterion (29) (and even a stronger criterion) can be obtained formally by substituting in (25) the trial function

$$\xi^{\infty} \left( \begin{array}{c} \cos \phi^{\circ} \\ \mathbf{1} \end{array} \right) \, .$$

#### **5. CONCLUSION**

The calculations performed above show the following: a) De Gennes' predictions,<sup>2</sup> confirmed by many experiments, are still not universal—they should be satisfied for a number of substances at not all values of the field intensity.

b) The Meyer theory of conic deformation, so far not confirmed by even one experiment, has nevertheless a region of validity.

The general picture of the behavior of cholesteric liquid crystals in a magnetic field turns out to depend substantially on the direction of the field variation (hysteresis). To classify the possible types of behavior of cholesterics with increasing field (from a sufficiently small value) it is useful to note that the condition  $H_s = H_1(g_s = 2/\pi)$  can define a certain curve  $R_3 = \Phi(R_1)$ . It is obvious from the results of Sec. 4 that this curve exists and lies entirely in the region  $R_3 > 4/\pi^2$ . If  $R_3 > \Phi(R_1)$ , then  $H_s > H_1$  and the predictions of Ref. 2 should be satisfied all the way to the transformation of a cholesteric into a nematic. We note that in this case there can be observed in the nematic state, at  $H < H_s$ , metastable states—twist solitons. If  $R_3 < \Phi(R_1)$ , then  $H_s < H_1$  and deviations from the de Gennes predictions<sup>2</sup> should be observed in the field interval  $H_s < H < H_{\perp}$ . The question of the character of the deviations remains open. Some information might be obtained by investigating the regime of the nonlinear instability that sets in at  $H = H_s$ . In the de Gennes case, however, the formal difficulties are so great that we were unable to solve even the simpler problem of finding  $H_s$ . With decreasing magnetic field (from a sufficiently large value to which the nematic state corresponds), substances with  $R_3 < 1$  and  $R_3 > 1$  behave differently. If  $R_3 < 1$ , conical deformation appears in the region  $H < H_{\parallel}$ ; it remains stable all the way to the field  $H_c$  determined by Eqs. (9) and (17). On going through  $H_c$  the state of the cholesteric changes jumpwise in view of the rigidity of the regime of the nonlinear elimination of the instability. Finally, at a certain field  $H'_{c}$  the cholesteric goes over into a state with an axis perpendicular to the field, after which the theory of Ref. 2 becomes applicable. If  $R_3 > 1$  then, as is known, no conical deformation occurs, but if we put  $H_c = H_{\parallel}$  everything said above concerning the case  $R_3 < 1$  can formally be retained.

<sup>1)</sup> The distribution of the director in substances with  $\Delta \chi < 0$  at the most suitable cholesteric axis orientation along the magnetic field is obviously indendent of the field intensity.

<sup>3)</sup>But is still sufficient to permit neglect of surface forces.

- <sup>1</sup>R. B. Meyer, Appl. Phys. Lett. 12, 281 (1068).
- <sup>2</sup>P. G. De Gennes, Sol. St. Comm. 6, 163 (1968).
- <sup>3</sup>H. Baessler, T. M. Larange, and M. M. Labes, J. Chem. Phys. **51**, 3213 (1969).
- <sup>4</sup>C. J. Gerritsma and P. Van Zanten, Mol. Cryst. 15, 257 (1971).
- <sup>5</sup>C. J. Gerritsma and P. Van Zanten, Phys. Lett. 42A, 329 (1972).
- <sup>6</sup>R. B. Meyer, Appl. Phys. Lett. 14, 208 (1969).
- <sup>7</sup>G. Durand, L. Leger, F. Rondeles, and M. Veyssie, Phys. Rev. Lett. 22, 227 (1969).
- <sup>8</sup>F. J. Kahn, Phys. Rev. Lett. 24, 209 (1970).
- <sup>9</sup>S. V. Belyaev, L. M. Blinov, and V. A. Kizel', Pis'ma Zh. Eksp. Teor. Fiz. **29**, 344 (1979) [JETP Lett. **29**, 310 (1979)].
- <sup>10</sup>W. Helfrich, Appl. Phys. Lett. **17**, 531 (1970).
- <sup>11</sup>J. P. Harrault, J. Chem. Phys. **59**, 2068 (1973).
- <sup>12</sup>J. M. Delrieu, J. Chem. Phys. **60**, 1081 (1974).
- <sup>13</sup>C. J. Gerritsma and P. van Zanten, Phys. Lett. 37A, 47 (1971).
- <sup>14</sup>T. J. Scheffer, Phys. Rev. Lett. 28, 593 (1972).
- <sup>15</sup>F. Rondelez and J. P. Hulin, Sol. St. Comm. 10, 1009 (1972).
- <sup>16</sup>M. Abramowitz and I. A. Stegun, eds. Handbook of Math. Functions, Dover, 1964, Chap. 20.
- <sup>17</sup>L. M. Blinov, Elektro- i magnitooptika zhidkikh kristallov (Electro- and Magnetooptics of Liquid Crystals), Nauka, 1978, Chap. VI.

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<sup>&</sup>lt;sup>2)</sup> The neutral stability of this mode is due to the invariance of the free energy to shifts of the director distribution over z.

<sup>&</sup>lt;sup>4)</sup> To dispense with similar stipulations, we assume hereafter that the threshold field  $H_s$  is infinite in those cases when the state (20) is stable at all H.