Fréedericksz transition in cholesteric liquid crystals without external fields

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The structure of a cholesteric liquid crystal in a plane cell is analyzed for the case in which the surface orientation of the director is rigidly fixed along the normal to the faces. The homogeneous and homeotropic structure is unstable at $qL \geq \pi K_{33}/K_{22}$, where L is the cell thickness, and $h = 2\pi/q$ is the pitch of the cholesteric helix. Measurements of the transition characteristics can yield the pitch h, and this method would be particularly important at $h \gtrsim 1\ \mu$m.

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The helical structure is known to be stable in large volumes for the mesophase of a cholesteric liquid crystal. Over distances $|\Delta\mathbf{r}| \lesssim h = 2\pi/q$ smaller than or of the order of the pitch of the helix h, on the other hand, a cholesteric liquid crystal is essentially indistinguishable from a nematic liquid crystal. For nematic crystals the confining surfaces can be treated in such a manner that the orientation of the director at the surface is rigidly fixed along the normal to the surface. The corresponding homogeneous orientation over the volume of a plane cell holding a nematic liquid crystal is called "homeotropic."

Clearly, if the cell thickness satisfies $L \ll h$, a homeotropic orientation is also possible for cholesteric liquid crystals, but if $L \gg h$ such a configuration would be unstable. Do distortions of the homeotropic structure arise even at small values of L/h? In this letter we show that up to the threshold value $(L/h)_{\text{th}} = K_{33}/2K_{22}$ the homogeneous and homeotropic structure remains stable, but slightly above this threshold the steady-state amplitude of the distortion is proportional to $(q/q_{\text{th}}-1)^{1/2}$.

For this analysis we will assume that the director $\mathbf{n}(\mathbf{r})$ is rigidly fixed at the $z=0$ and $z=L$ planes. For the problem which is homogeneous in the (x,y) plane we can adopt

$$\mathbf{n}(z) = \mathbf{e}_x n_x(z) + \mathbf{e}_y n_y(z) + \mathbf{e}_z n_z(z) \equiv \vec{\nu}(z) + \mathbf{e}_z n_z(z), \tag{1}$$

$$n_x(0) = n_y(0) = n_x(L) = n_y(L) = 0,$$

where $n_x^2 + n_y^2 + n_z^2 = 1$. The free energy F (in ergs per cubic centimeter) of the cholesteric liquid crystal is

$$F = \frac{1}{2}\left[K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n} + q)^2 + K_{33}[\mathbf{n}\operatorname{rot}\mathbf{n}]^2\right]. \tag{2}$$

The unperturbed state corresponds to $\mathbf{n}(z) = \mathbf{e}_z = \text{const}$. Assuming the perturbations

$n_x(\mathbf{r}, t)$ and $n_y(\mathbf{r}, t)$ to be small, and varying (2) and the dissipative function $R = 0.5\eta\dot{\mathbf{n}}^2$ (ergs per cubic centimeter), we find the following equation, which holds within terms of order $\vec{\nu}^3$ inclusively:

$$-\eta\frac{\partial\vec{\nu}(z,t)}{\partial t} + K_{33}\frac{\partial^2\vec{\nu}}{\partial z^2} - 2K_{22}q\left[\mathbf{e}_z\frac{\partial\vec{\nu}}{\partial z}\right] + K_{22}q\vec{\nu}^2\left[\mathbf{e}_z\frac{\partial\vec{\nu}}{\partial z}\right] = 0. \tag{3}$$

Here η (in poises) is the relaxation constant.

It is not difficult to see that $\vec{\nu}(z,t) = 0$ is an exact solution of the problem. To determine the stability of this solution, we should retain only those terms in (3) which are linear in $\vec{\nu}$. The general solution of the linearized version of Eq. (3) is

$$\nu_i(z,t) = \sum_\alpha c_\alpha \exp(\Gamma_\alpha t) D_i^\alpha(z), \tag{4}$$

where $D_i^{(\alpha)}$ are the eigenfunctions and Γ_α the eigenvalues of the Hermitian operator

$$\eta^{-1}\left[K_{33}\frac{\partial^2}{\partial z^2}\delta_{ik} + 2K_{22}qe_{ikz}\frac{\partial}{\partial z}\right]D_k(z) = \Gamma D_i(z), \tag{5}$$

with the boundary conditions $\mathbf{D}(0) = \mathbf{D}(L) = 0$. These eigenfunctions and eigenvalues are given by

$$\begin{aligned} \mathbf{D}_m^{(\pm)} &= (\mathbf{e}_x \pm i\mathbf{e}_y)\exp\left(\pm i\frac{K_{22}}{K_{33}}qz\right)\sin\frac{m\pi z}{L}, \\ \Gamma_m &= \eta^{-1}\left[-K_{33}\left(\frac{m\pi}{L}\right)^2 + q^2\frac{K_{22}^2}{K_{33}}\right]. \end{aligned} \tag{6}$$

Since the original operator is real, the eigenfunctions $\mathbf{D}_m^{(+)}$ and $\mathbf{D}_m^{(-)}$, which are complex conjugates of each other, have identical values of Γ_m. It is not difficult to see that with $q = 0$ (i.e., for a nematic liquid crystal) we have $\Gamma_m < 0$ for all m and that the homeotropic orientation is stable. At $q = q_0 = \pi K_{33}/K_{22}L$ the eigenvalue Γ_1 crosses zero and changes sign. It is this event which corresponds to a Fréedericksz transition without an external field.

When the threshold value of the parameter L/h is exceeded, a distorted structure arises. When L/h is instead reduced through the threshold value, there may be either a hysteresis or a singled-valued behavior of the distortions, depending on the sign of the quantity $K_{11} - 3(K_{33} - K_{22})$.

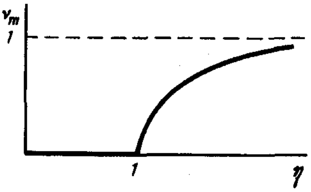

FIG. 1. Schematic plot of the equilibrium amplitude of the perturbation, ν_m, vs the parameter $\eta = qLK_{22}/\pi K_{33}$ under the condition $K_{11} - 3(K_{33} - K_{22}) > 0$.

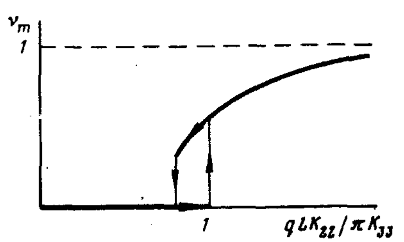

FIG. 2. The same as in Fig. 1, but for $K_{11} - 3(K_{33} - K_{22}) < 0$. In this case there should be a hysteresis.

Just slightly above the threshold the amplitude is stabilized by the terms nonlinear in $\vec{\nu}$ which we discarded in writing Eq. (3). The first nonvanishing corrections to Eq. (3) are of order ν^3. At $K_{11} - 3(K_{33} - K_{22}) > 0$ it is in fact the terms $\sim\nu^3$, which stabilize the amplitude of the perturbation near the threshold; writing $q = q_0\eta$, where $0 < \eta - 1 \ll 1$, we then find the following expressions from the nonlinear equations:

$$\mathbf{n}(z) = \mathbf{e}_z\sqrt{1 - \vec{\nu}^2} + \vec{\nu}(z),$$

$$\vec{\nu}(z) = \sqrt{\eta - 1}\sqrt{\frac{K_{11} - 3(K_{33} - K_{22})}{4K_{33}}}\sin\frac{\pi z}{L} \left\{\mathbf{e}_x\left(\sin\frac{\pi z}{L} - \beta\right) - \frac{q}{|q|}\mathbf{e}_y\cos\left(\frac{\pi z}{L} - \beta\right)\right\}, \tag{7}$$

where β is an arbitrary parameter. The state in (7) corresponds to a loosely twisted helix with an amplitude proportional to the square root of the extent to which the threshold is exceeded. Figure 1 is a schematic plot of $\nu_m = |\vec{\nu}(z = L/2)|$ vs the parameter qL for this case. This would be the situation, in particular, for a single-constant cholesteric liquid crystal.

In the case $K_{11} - 3(K_{33} - K_{22}) < 0$ we must solve the nonlinear equations even more accurately in order to determine the stationary state, since the terms $\sim\nu^3$ no longer stabilize the perturbations but, on the contrary, cause them to grow. We will omit the corresponding calculations here, and we will simply point out that in this case the Fréedericksz transition without an external field should exhibit a hysteresis, as shown schematically in Fig. 2.

The threshold for the Fréedericksz transition of a cholesteric liquid crystal without an external field could be detected by simple polarized-optics methods. The transition itself could be observed by varying the temperature, the hydrostatic pressure, the concentration of a chiral impurity, the cell thickness, etc. This transition could be used to measure the parameter q of the crystal at (the extremely interesting) small values of q, where optical interference methods cannot be used.

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