Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 21 Mar 2007.

To cite this article: G. Derfel (1983): Shear Flow Induced Cholesteric-Nematic Transition, Molecular Crystals and Liquid Crystals, 92:2, 41-47

To link to this article: http://dx.doi.org/10.1080/01406568308084517

Mol. Cryst. Liq. Cryst. Vol. 92 (Letters), pp. 41-47
0140-6566/83/9202-0041$18.50/0

SHEAR FLOW INDUCED CHOLESTERIC-NEMATIC TRANSITION

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(Received for Publication February 11, 1983)

Abstract: It is shown that the shear flow induced cholesteric-nematic transition occurs if the helix axis is normal to the shear plane and the shear rate exceeds a critical value.

A number of theoretical and experimental investigations have been reported on the rheological properties of cholesteric liquid crystals. Theoretical approaches to this problem were made for two geometries. In the first one the helix axis was parallel to the velocity vector, in the second one - parallel to the velocity gradient (planar texture). In the first case the permeation effect introduced by Helfrich[1] was proposed as a reason for enormous viscosity values observed in Poiseuille flow at low shear rates. In the second case the apparent viscosity was found to oscillate with pitch variation[2].

The third possible geometry i. e. where the helix axis is perpendicular to both the velocity and velocity gradient, has not been considered yet.

Such a configuration is possible in the unidirectionally aligned "fingerprint" texture, which can be obtained as a result of homeotropic boundary conditions. In this paper the problem of the flow of cholesteric will not be solved generally. It will be shown only that the shear flow induces the unwinding of cholesteric structure. The calculus is analogous to the well known treatment of cholesteric-nematic transition in external field[3].

Let us consider the homogeneous cholesteric slab aligned homeotropically between two infinite parallel plates with the helix axis along z. One plate is moving relative to the other with constant velocity V towards y. The x coordinate is normal to the plates. For simplicity one can consider sufficiently thick layer of liquid crystal and ignore the orienting influence of the surfaces which distorts the cholesteric twist. One may therefore assume that the shear rate $dv/dx = u$ is constant at any place in the sample. The components of the director are: $n_x = \cos\varphi(z)$, $n_y = \sin\varphi(z)$, $n_z = 0$, where $\varphi(z)$ is the azimuthal angle between $\vec{n}$ and x axis, counting φ positive for a clockwise rotation. There is neither splay nor bend deformation in thick sample approximation, and twist is the only effect taken into account.

The equation of director motion for static case gives the condition for equilibrium of elastic and viscous torques per unit volume:

$$K_{22}\frac{d^2\varphi}{dz^2} = -u\left(\alpha_3\sin^2\varphi - \alpha_2\cos^2\varphi\right) \qquad (1)$$

where K_{22} is twist elastic constant and α_2 and α_3 are viscosity coefficients. Integration of Eq.(1) gives:

$$\frac{d\varphi}{dz} = \pm\sqrt{\frac{u}{K_{22}}}\sqrt{k - (\alpha_3-\alpha_2)\varphi + (\alpha_3+\alpha_2)\sin\varphi\cos\varphi} \quad (2)$$

One of the two possible senses of helix should be determined e. g. by choice of the + sign. The integration constant k is a function of u. Its value must rise to infinity when u decreases to zero because $d\varphi/dz$ should be constant in cholesteric at rest.

From the definition of pitch P and by use of Eq.(2) one can obtain the following formula:

$$P = 2\int dz = 2\int \frac{dz}{d\varphi}d\varphi =$$

$$= 2\sqrt{\frac{K_{22}}{u}}\int \frac{d\varphi}{\sqrt{k-(\alpha_3-\alpha_2)\varphi+(\alpha_3+\alpha_2)\sin\varphi\cos\varphi}} \quad (3)$$

The integration is extended over the period of structure i. e. P/2 or π. It can be shown that for arbitrary k there exists the integration range of width π, such that the integral in Eq.(3) is infinite. This is equivalent to infinite pitch, what denotes the nematic phase. In the well known phenomenon of shear alignment of the nematic the angle between director and velocity gradient is:

$$\varphi_c = \mathrm{arcctg}\sqrt{\frac{\alpha_3}{\alpha_2}} \quad (4)$$

It is therefore reasonable to choose φ_c as the upper limit of integration and $\varphi_c-\pi$ as the lower. (The angle $\varphi_c+\pi$ does not belong to the domain of the integrand.) The critical value of k which gives the infinite pitch is now obtainable. This value, k_c, determines the critical shear rate u_c which causes the transition from cholesteric to nematic aligned at angle φ_c:

$$k_c = k(u_c) = (\alpha_3-\alpha_2)\varphi_c-(\alpha_3+\alpha_2)\sin\varphi_c\cos\varphi_c \qquad (5)$$

The relation between k and u can be found by minimalization of the average free energy density calculated for a layer of thickness P/2 and unit area in the x-y plane:

$$F = \frac{K_{22}}{P}\int_{\varphi_c-\pi}^{\varphi_c}\left(\frac{d\varphi}{dz}+\frac{2\pi}{P_o}\right)^2\frac{dz}{d\varphi}\,d\varphi +$$

$$+\frac{u}{P}\int_{\varphi_c-\pi}^{\varphi_c}\left[(\alpha_3+\alpha_2)\sin\varphi\cos\varphi-(\alpha_3-\alpha_2)\varphi\right]\frac{dz}{d\varphi}\,d\varphi \qquad (6)$$

The second term results from the action of viscous torque. Its form was obtained by integration of this torque over φ. The last expression can be rewrite as:

$$F = \frac{2}{P}\sqrt{K_{22}u}\int_{\varphi_c-\pi}^{\varphi_c}\sqrt{k-(\alpha_3-\alpha_2)\varphi+(\alpha_3+\alpha_2)\sin\varphi\cos\varphi}\,d\varphi -$$

$$-\frac{2\pi^2}{P_o}K_{22}\left(\frac{2}{P}-\frac{1}{P_o}\right)-\frac{uk}{2} \qquad (7)$$

where Eqs.(2) and (3) were applied. Minimalization condition $dF/dk = 0$ gives the relation:

$$\sqrt{\frac{u}{K_{22}}}\int_{\varphi_c-\pi}^{\varphi_c}\sqrt{k-(\alpha_3-\alpha_2)\varphi+(\alpha_3+\alpha_2)\sin\varphi\cos\varphi}\,d\varphi = \frac{2\pi^2}{P_o} \tag{8}$$

The integrals appearing in Eqs.(3) and (8) will be denoted by $G_1(k)$ and $G_2(k)$ respectively:

$$G_1(k) = \int_{\varphi_c-\pi}^{\varphi_c}\frac{d\varphi}{\sqrt{k-(\alpha_3-\alpha_2)\varphi+(\alpha_3+\alpha_2)\sin\varphi\cos\varphi}} \tag{9}$$

$$G_2(k) = \int_{\varphi_c-\pi}^{\varphi_c}\sqrt{k-(\alpha_3-\alpha_2)\varphi+(\alpha_3+\alpha_2)\sin\varphi\cos\varphi}\,d\varphi \tag{10}$$

For $k=k_c$ G_1 is infinite, whereas G_2 reaches finite value. Setting $k=k_c$ in Eq.(8) one can calculate the critical threshold shear rate u_c:

$$u_c = K_{22}\left(\frac{2\pi^2}{P_o G_2(k_c)}\right)^2 \tag{11}$$

From Eqs.(8) and (11) it is also possible to find the relation between k and u:

$$\frac{u}{u_c} = \frac{G_2^2(k_c)}{G_2^2(k)} \tag{12}$$

This relation can be used to find pitch as function of shear rate:

$$P = 2\sqrt{\frac{K_{22}}{u}}\,G_1(k(u)) \tag{13}$$

As the functions G_1 and G_2 have no analytical representation, only the numerical calculation of relations between considered quantities and their critical values is possible. Everywhere below the values of MBBA parameters are used: $\alpha_2=-77\cdot10^{-3}$ Ns/m^2, $\alpha_3=-1\cdot10^{-3}Ns/m^2$, $K_{22}=4\cdot10^{-12}N$, and undistorted pitch $P_o=1\cdot10^{-5}m$ is assumed. One obtains: $\varphi_c=83.5^o$, $k_c=0.11953Ns/m^2$, $u_c=15.6s^{-1}$. Figure 1 presents a plot of the computed shear rate dependence of pitch. Numerical solution of Eq.(2) yields the $\varphi(z)$ function. The dashed line in Figure 2 presents the result for $u=0.71u_c$ obtained by use of the boundary condition φ_c at z=0. In the real not entirely unwinded helical structure there is realized the $\varphi(z)$ dependence defined by periodically

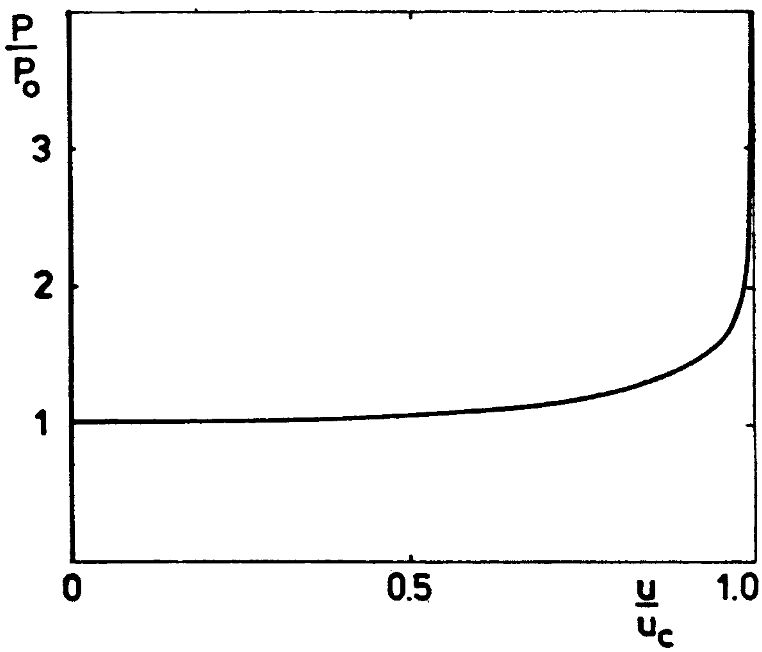

FIGURE 1 Ratio of the pitch at finite shear rate to the pitch at rest, P/P_o, plotted as a function of reduced shear rate u/u_c.

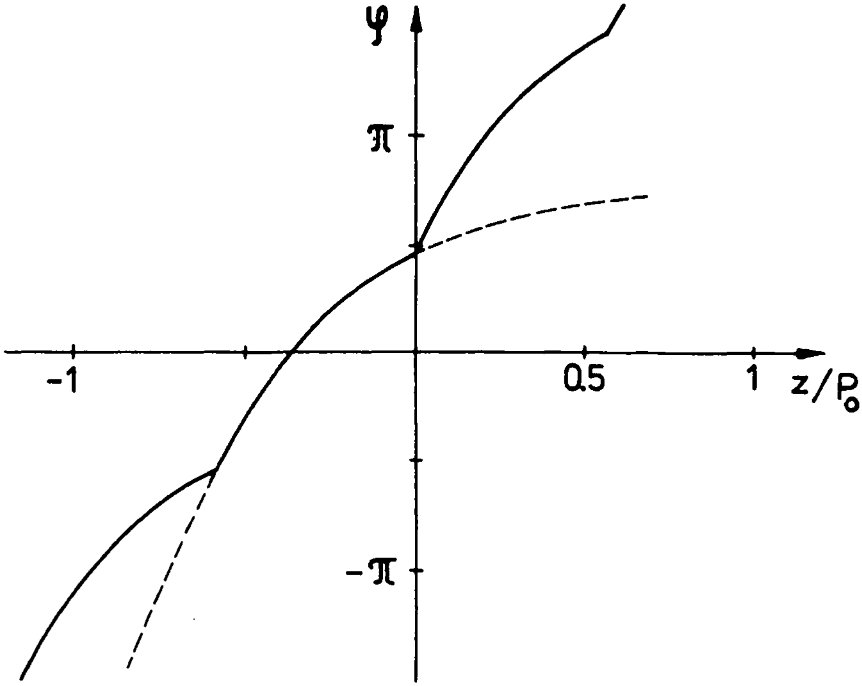

FIGURE 2 The azimuthal angle φ as a function of z for $u=0.71u_c$; $P=1.16P_o$.

repeated section of this curve extending from $\varphi_c-\pi$ to φ_c, since it corresponds to the z range of length P/2. This dependence is shown by continuous line in Figure 2. The pitch is lengthened to $1.16P_o$.

Experimental verification of the effect predicted here is under consideration now.

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