Heat Convection in Liquid Crystals Heated from Above

P. Pieranski, E. Dubois-Violette, and E. Guyon
Laboratoire de Physique des Solides, Université Paris-Sud, Centre d'Orsay, 91405 Orsay, France*
(Received 29 December 1972)

The anisotropic viscoelastic and heat-conductivity properties of nematic liquid crystals offer the possibility of obtaining thermal convection with very low thresholds in homeotropic films heated from above. We present experiments on *p*-methoxybenzilidene-*p*-*n*-butylaniline (MBBA) and a simplified two-dimensional calculation.

Nematic liquid crystal films (LC's) have unusual heat-convective properties associated with their viscoelastic behavior. We discuss here the occurrence of convection in homeotropic films (director axis $\vec{n}$ perpendicular to the limiting glass plates) due to the anisotropic heat diffusivity $\kappa_a = \kappa_\parallel - \kappa_\perp$ ($\parallel$ and $\perp$ refer to a heat flow parallel and perpendicular to $\vec{n}$).

One of us[1] has discussed theoretically the heat-convection effects in planar films ($\vec{n}$ parallel to the plates). The anisotropic mechanism is similar to the "Carr-Helfrich" mechanism[2,3] associated with the electrical-conductivity anisotropy. If $\kappa_a > 0$, the predicted reduction (by a factor 10^3) of the temperature-difference threshold ΔT_{cr} with respect to an isotropic material of the same average properties has been observed experimentally[4] in *p*-methoxybenzilidene-*p*-*n*-butylaniline (MBBA) ($\kappa_a \sim +4\times10^{-4}$ cgs).[5]

Heat convection in *materials heated from above*, which does not occur in homogeneous ordinary liquids, can take place in LC's. Reference 1 discusses this possibility in planar samples with a negative κ_a. This condition is not usually met in nematics. In the following, we will see that such a convection is found in the *converse* case: homeotropic samples with a positive κ_a anisotropy. We use MBBA films of thicknesses $d = 0.5$ and 1 mm. The homeotropic alignment is obtained by coating the glass plates with a monomolecular layer of detergent.[6] A vertical temperature difference is applied through the horizontal optical cell by circulating water at different temperatures on the outer sides of the plates. If the undistorted LC is illuminated with ordinary light, between crossed polarizers, the field of view is dark. The initial development of the distortion due to the convection is accurately detected by the observation of the first interference colors.

When the LC is heated from below, no distortion is produced up to the largest temperature difference available (30°). With the high-temperature side above, convection cells can be observed (Fig. 1) in thick films above a threshold, after waiting sufficiently long (see discussion below). ΔT_{cr} is 5.6° for $d = 1$ mm and $> 35°$ for $d = 0.5$ mm. This is of the order of magnitude of the thresholds in planar samples ($\Delta T_{cr} = 2.2°$ for 1 mm; 15.5° for 0.5 mm.[4]

The anisotropic mechanism is indicated schematically on Fig. 2 which will be used to explain why convective instabilities can be obtained only when the sample is heated from above. We consider an orientation fluctuation of the director,

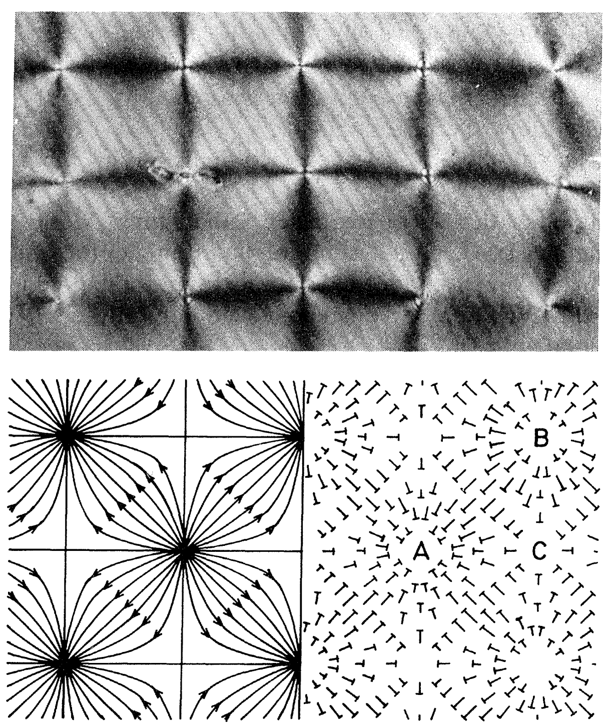

FIG. 1. Upper: between cross polarizers, obtained by heating a 1-mm-thick LC film from above ($\Delta T \sim 15°$). Lower: left side of the corresponding figure, giving the horizontal projection of the flow for $z = \frac{1}{4}d$; right side, giving the projection of the molecular orientation.

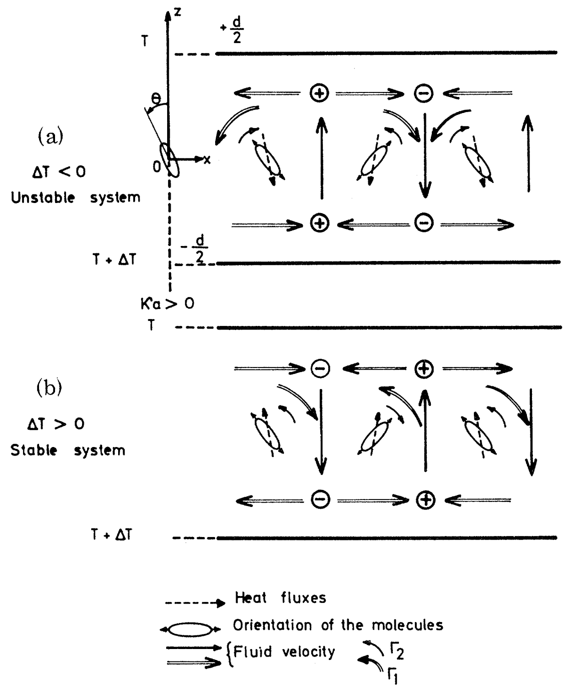

FIG. 2. Schematic of the anisotropic mechanism. Because of the thermal conductivity anisotropy $\kappa_a > 0$, the heat fluxes are deviated along the molecules. Under the influence of the buoyancy forces, warmer (+) and cooler (−) regions move up and down. Γ_1 is the viscous torque due to the "transverse" velocity gradient. Γ_2 is viscous torque due to the "longitudinal" velocity gradient. (a) Total torque destabilizing, and (b) stabilizing.

characterized by a small angle θ with Oz. The heat-flux lines are not parallel to the applied thermal gradient but are deviated along the molecules if the anisotropic heat conductivity is positive. The temperature becomes modulated along Ox. Under the influence of the buoyancy forces, warmer and cooler regions move up and down. This motion corresponds to a shear flow along Ox and Oz. The dissipative torque Γ_{vis} exerted on the director is

$$\Gamma_{\mathrm{vis}} = \gamma_1 \theta + \frac{\gamma_1 - \gamma_2}{2}\frac{\partial V_x}{\partial z} - \frac{\gamma_1 + \gamma_2}{2}\frac{\partial V_z}{\partial x}. \qquad (1)$$

It is well known that in nematics the torques $\Gamma_1 = \frac{1}{2}(\gamma_1 - \gamma_2)\partial V_x/\partial z$ and $\Gamma_2 = \frac{1}{2}(\gamma_1 + \gamma_2)\partial V_x/\partial z$ exerted on the director by the normal and parallel components of the fluid velocity have opposite signs, and that $\Gamma_1 \gg -\Gamma_2$ [$-\gamma_2 \gtrsim \gamma_1 > 0$, which implies $\gamma_1 - \gamma_2 \gtrsim 2\gamma_1 \gg -(\gamma_1 + \gamma_2) > 0$].

It follows that the "anisotropic mechanism" favors the convective process when the sample is heated from above [Fig. 2(a)] since the total dissipative torque $\Gamma_1 + \Gamma_2$ is destabilizing in that case.[7] On the contrary, when the sample is heated from below [Fig. 2(b)], the anisotropic mechanism induces a stabilizing dissipative torque: The convective instability is strongly repressed.[8]

A one-dimensional model, as used in Ref. 1, with all quantities depending only on x, is not applicable: The only torque that would be considered is Γ_2, which is not the dominant one. We must look for a two-dimensional solution with fluctuation modes defined as

$$\begin{aligned} \theta &= \theta_0 \cos(q_x x)\exp(iq_z z)e^{st},\\ \vec{v} &= v_0\left[i\frac{q_z}{q_x}\cos q_x x,\, 0,\, \sin q_x x\right]\exp(iq_z z)e^{st},\\ \delta T &= \delta T \sin(q_x x)\exp(iq_z z)e^{st}; \end{aligned} \qquad (2)$$

δT and $\vec{v}$ are the temperature fluctuation and velocity induced by the orientation fluctuation. The threshold is obtained by solving all the conservation laws and the appropriate boundary conditions for θ, $\vec{v}$, and δT. A detailed determination of the threshold will be given in a separate article. The calculated threshold can be written as

$$\begin{aligned} \Delta T(q_x) &= -\frac{q_x^4 d}{\rho g \alpha}\bar{\kappa}_\perp \bar{\eta}\left(-1 + \frac{\kappa_a}{\bar{K}_1}\frac{\langle \gamma_1 - \gamma_2\rangle}{2}\right)^{-1}\\ &= -\Delta T_I\left(-1 + \frac{\kappa_a}{\bar{K}_1}\frac{\langle \gamma_1 - \gamma_2\rangle}{2}\right)^{-1}, \end{aligned} \qquad (3)$$

where the thermal diffusivity $\bar{\kappa}_\perp$, the elastic constant $\bar{K}_1$, the viscous coefficients $\langle \gamma_1 - \gamma_2\rangle$ and $\bar{\eta}$ are mean values depending on the possible modes (characterized by $p_i = q_z/q_x$) which satisfy the boundary conditions. The form of Eq. (3) shows that convective instabilities can appear when the sample is heated from above ($\Delta T < 0$) if $\kappa_a > 0$. ΔT_I has the form of a temperature threshold for an isotropic liquid of comparable average properties, heated from below. The absolute value ΔT_{cr} is reduced by a factor $(\kappa_a/\bar{K}_1)\frac{1}{2}\langle \gamma_1 - \gamma_2\rangle \sim 10^2$ to 10^3 as compared with the isotropic case. A tentative estimate of the threshold is obtained by setting $q_{x,\mathrm{cr}} = \pi/d$. This value is in good agreement with the spatial period measured experimentally. Using typical values of the viscosities[9] and of κ and K in MBBA, we get $\Delta T_{\mathrm{cr}} \sim 0.5°$, $1°$ for $p = \frac{1}{2}$, 1, and $d = 1$ mm, in reasonable agreement with the experimental result, taking into account the crudeness of this model.

A vertical applied magnetic field is found experimentally to increase the threshold. Some ex-

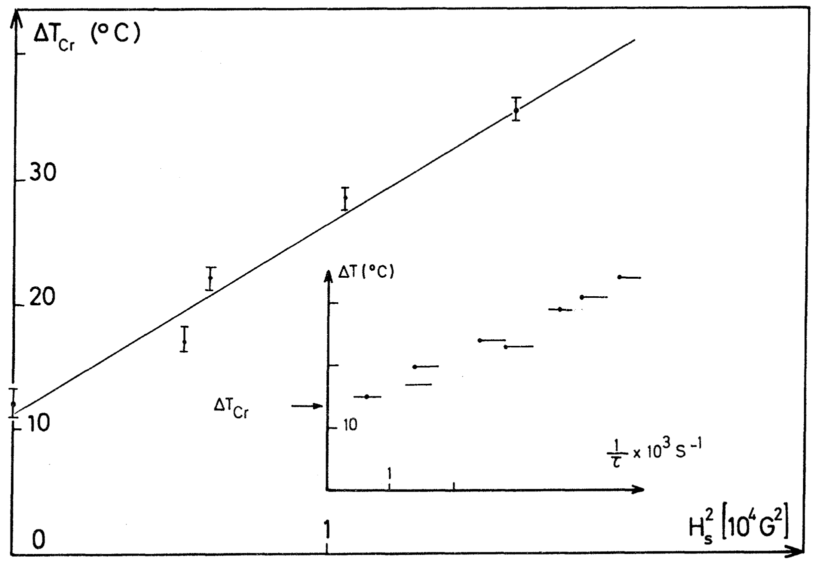

FIG. 3. Temperature-difference threshold ΔT_{cr} in a stabilizing magnetic field H_s. Inset, determination of ΔT_{cr} (here for $H_s = 0$) from the divergence of the rise time τ of the convection.

perimental results for $d = 1$ mm are presented in Fig. 3 and agree with a form

$$\Delta T_{cr}(H) = \Delta T_{cr}(H=0)[1 + (H/H_0)^2], \qquad (4)$$

with a value $H_0 = 85$ G. This result is exactly equivalent to the form obtained for the effect of a horizontal stabilizing field for convection in the planar geometry,[4] and the theoretical analysis follows that of Ref. 1. The field $H_0 = (\pi/d)(K/\chi_a)^{1/2}$ has the form of a Freedericksz critical field. The variation (4) is easily understood as due to the additional stabilizing effect of the magnetic torque and of the elastic one. Because of the boundary conditions, the elastic constant $\bar{K} = K_1 + K_3 p_i^2$ is a mixture of a splay term along Ox (K_1) and of a bend term along Oz (K_3).

A horizontal magnetic field H lifts the angular degeneracy of the distortion obtained in the homeotropic geometry (Fig. 1) and tends to favor the formation of rolls with axis perpendicular to H. In the limit $H \to 0$, the threshold for the formation of rolls is of the same order as the one for the square pattern shown in the photograph. However, rolls are found to degenerate into square cells if the cause of uniaxial dissymmetry is suppressed.

We have performed a dynamic study of the convection. For a given temperature difference, a vertical field H, large enough to "erase" the convection structure ($H \gg H_0$), is applied for a standard time $\delta t_e = 5$ min; then the field is removed ($t = 0$). The reappearance of the distortion is observed by measuring the light intensity diffracted by the structure, using a directional photocell at an angle with the incident light beam. Far enough above the threshold, the intensity follows an exponential law $I(t) = e^{t/\tau}$. The value I_0 depends on the sample history before $t = 0$ and, in particular, on the value of δt_e. The time constant τ is a function of ΔT as shown in the inset of Fig. 3. It diverges at ΔT_{cr}. This provides a new determination of this threshold. However, near ΔT_{cr} where the times involved are very large (several hours), we have not been able to characterize the evolution from a single exponential law.

The square-cell structure can be thought to be a linearly independent superposition of crossed convection rolls. This structure can be deduced from the optical study: The lower part of Fig. 1 gives the horizontal projection of the flow in a plane $z = \frac{1}{4}d$. The projection of $\vec{n}$ in a horizontal plane (shown in the lower right-hand side of this figure) shows a network of $+1$ (points A, B) and -1 (point C) nonsingular disinclinations. The lattice points extend as vertical disinclination lines connecting the two limit plates.

When ΔT is much larger than ΔT_{cr}, the square pattern is replaced by a complex one with hexagonal cells.[10] When the upper temperature is sufficiently large, a nematic-isotropic interface is created in the sample; the nature of the convection is quite different from the case discussed above because of the nearly planar boundary condition at the interface. This subject will be discussed in a further report.

P. K. Currie has obtained independently similar theoretical results. A detailed numerical calculation together with a comparison with Currie's description will be given elsewhere.

*Laboratoire associé au Centre National de la Recherche Scientifique.

[1]E. Dubois-Violette, C. R. Acad. Sci. 21, 275, 923 (1971).

[2]E. F. Carr, Mol. Cryst. Liquid Cryst. 7, 253 (1969).

[3]W. Helfrich, J. Chem. Phys. 51, 4092 (1969).

[4]E. Dubois-Violette, E. Guyon, and P. Pieranski, in Proceedings of the International Conference on Liquid Crystals, Kent, Ohio, 1972 (Mol. Cryst. Liquid Cryst., to be published).

[5]P. Pieranski, F. Brochard, and E. Guyon, J. Phys. (Paris) 33, 681 (1972).

[6]I. Haller, J. Chem. Phys. 57, 1400 (1972).

[7]A more complete discussion of the "stabilizing" or "destabilizing" character of this dissipative torque can be found in Ref. 3 and in F. M. Leslie, Quart. J. Mech. Appl. Math. 19, 357 (1966); O. Parodi, J. Phys. (Paris) 31, 581 (1970).

[8]Of course, for samples with $\kappa_a = 0$, the usual Bénard process, for isotropic liquids heated from below, takes place. But as soon as $\kappa_a < 10^{-6}$ cm^2 sec^{-1} the anisotropic mechanism is the dominant one.

[9]We use the notations and values of the viscosity coefficients given by Ch. Gähwiller, Phys. Lett. 36A, 311 (1971).

[10]In isotropic liquids, the formation of hexagons is observed when the viscosity changes with temperature are important. A general discussion of heat-convection effects in isotropic liquids is given in S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability* (Oxford Univ. Press, Oxford, England, 1961).

Nonlinear Behavior of Stimulated Brillouin and Raman Scattering in Laser-Irradiated Plasmas*

D. W. Forslund, J. M. Kindel, and E. L. Lindman
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544

(Received 4 December 1972)

Both analytic theory and inhomogeneous plasma simulations are presented to show the nonlinear development of the Brillouin and Raman backscatter instabilities. Nonlinear fluid behavior predicts a backscatter energy on the order of the incident laser energy, but particle trapping and heating effects associated with the excited electrostatic wave can significantly reduce this.

When laser light above certain power levels irradiates a plasma, collective processes occur which can either enhance[1] or retard the light absorption.[2-5] In the latter case the incident energy is reflected by the plasma through the decay of the incident light wave into a backward-traveling light wave and either a low-frequency ion wave (stimulated Brillouin scattering) or an electron plasma wave (stimulated Raman scattering). The energy in the light wave is thus prevented from arriving at the critical surface (that place in the plasma were the plasma frequency equals the light-wave frequency) where enhanced absorption can take place. Since energy absorption[1] is crucial in laser fusion and ionospheric radar modification applications,[6] a knowledge of the penetration depth allowed by these instabilities and the incident energy reflected is required.

These backscatter instabilities[2] are examples of the backward wave oscillator problem. The temporal linear theory (real k, complex ω) and the spatial linear theory (complex k and ω) have appeared in the literature.[7]

Nonlinear problem.—As in the linear theory the fluid equations for ions and electrons are combined with Maxwell's equations using the vector potential formalism,[3b] but the reaction of the backscattered and electrostatic waves on the pump wave is no longer neglected. As discussed in Ref. 2, there are three linear waves excited by the circularly polarized pump wave, two electromagnetic and one electrostatic. In both the Brillouin and Raman problems, one of the electromagnetic waves, the forward scattered wave, is not in resonance and can be neglected. Insofar as electrostatic steepening terms and higher-order mode coupling terms are negligible, the following set of equations describe both the Brillou-

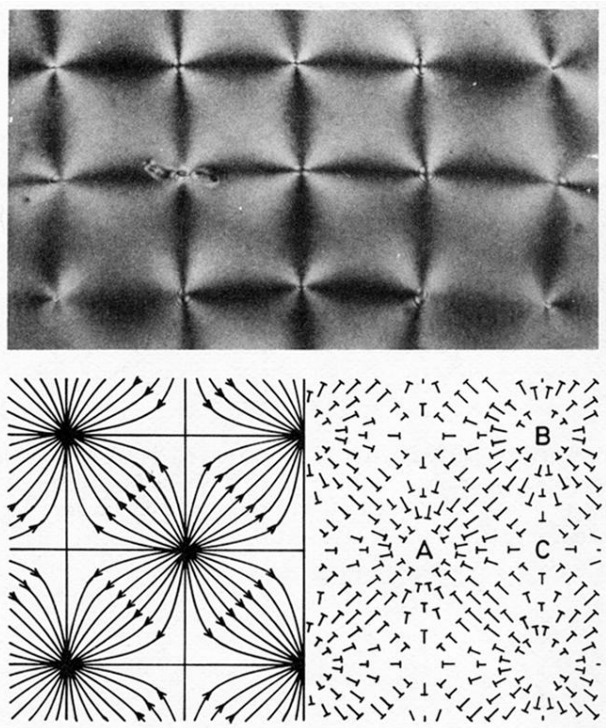

FIG. 1. Upper: between cross polarizers, obtained by heating a 1-mm-thick LC film from above ($\Delta T \sim 15°$). Lower: left side of the corresponding figure, giving the horizontal projection of the flow for $z = \frac{1}{4}d$; right side, giving the projection of the molecular orientation.