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CONVECTIVE INSTABILITIES IN HOMEOTROPIC NEMATIC LIQUID CRYSTALS: EXPERIMENTAL RESULTS IN THICK LAYERS

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Experimental results are presented for Rayleigh–Bénard convection in thick horizontal layers of Homeotropic Nematic Liquid Crystal heated from below under external magnetic fields. New results are shown about the subcritical bifurcation in the oscillatory and steady convective regimes. Also, a more accurate estimation of the codimension-two point is made. The frequencies of the oscillatory solutions and other nonlinear effects are measured.

1. Introduction

In the last twenty years, many works were developed in the study of the hydrodynamics of the Nematic Liquid Crystals. The anisotropic properties and its sensibility to applied external forces (as magnetic or electric fields) have allowed the observation of many interesting dynamical phenomena [de Gennes & Prost, 1993]. In the case of thermal convective instabilities for horizontal Nematic samples, the instabilities are drastically different from those obtained in homogeneous isotropic fluid layers [Dubois-Violette *et al.*, 1978; Guyon *et al.*, 1979; Salan & Guyon, 1983]. Two geometries can be distinguished depending on the molecular orientation (characterized by a unitary vector in the direction the averaged molecular orientation, named the Director): The one named Planar, when the Director is parallel to the horizontal boundaries, and the Homeotropic one when the Director is perpendicular to them. Surely the most interesting effect is the existence of a linear instability when a horizontal Homeotropic layer is heated from above [Pieranski *et al.*, 1973]. The existence of this instability rests on the coupling between the flow induced by the buoyancy forces and the distortion of the initial Director orientation. This coupling, added to the anisotropic thermal conductivity of these materials yields a heat focusing-defocusing effect when spontaneous Director fluctuations are generated. Though the density gradients are stable, the heat focusing mechanism enhances the initial Director fluctuations and then convection will occur above a relatively low threshold. [Pieranski *et al.*, 1973].

When the Nematic layer is heated from below, the unfavorable couple between the convective flow field and the Director, due to the heat focusing mechanism, give rise to an oscillatory instability rather than an increase of the threshold (with respect to the isotropic case) [Lekkerkerker, 1977]. This oscillatory effect comes from the existence of very different time scales for the relaxation of the Director (stabilizing mechanism), large compared with the thermal relaxation time (related with the destabilizing buoyancy force). The application of an external magnetic field parallel to the Director diminishes the characteristic time of the relaxation of the Director fluctuations, due to the contribution of the anisotropic part of the magnetic forces to the elastic one. Following a simple linear model (such as that presented in [Guyon *et al.*, 1979]) we can obtain that the characteristic time for the relaxation of the Director, $T_d = \gamma d^2/\pi^2 K$ (where γ is the rotational viscosity, K should be an averaged value of the Frank elastic constants, and d is the thickness of the cell. See for instance, [de Gennes & Prost, 1993]) is modified into $T_d = \gamma/(\chi_a H^2 + q^2 K)$, where $\chi_a = \chi_\parallel - \chi_\perp$ is the anisotropic part of the magnetic susceptibility and the signs $\parallel$ and $\perp$ refer to the parallel and normal components of the magnetic susceptibility, and $q = \pi/d$. The first experiments and theoretical linear model including the effect of external magnetic fields were made by Guyon *et al.* [1979]. In this work and in the later ones [Velarde & Zuniga, 1979; Dubois-Violette & Gabay, 1982; Feng, 1992; Feng *et al.*, 1992; Alhers, 1996], experiments and linear or nonlinear models are developed about this problem. Among the results associated with the oscillatory instability, they observe the growing of the time-periodic perturbations near the linear threshold, obtaining an inverted bifurcation known as Hopf bifurcation [Manneville, 1993]. This problem is analog to the thermal convection in horizontal films of binary-fluid mixtures heated from below, where depending on the sign of Soret number (non-dimensional number relating linearly the thermal gradients and flux of matter), it is possible to also obtain a Hopf bifurcation (See for instance, [Velarde & Antoranz, 1981; Cross & Hohenberg, 1993]).

In this work we present experimental results about different convective regimes when a horizontal Nematic layer is heated from below. First, we reproduce previous results of the variation of oscillatory thresholds with the intensity of the applied magnetic fields using a very thick Nematic layer. A comparison is made with recent theoretical results considering nonlinear effects [Feng, 1992; Feng *et al.*, 1992]. Measurements of the hysteresis associated to the Hopf bifurcation are presented showing two different singular points, depending on the strength of the magnetic field. The first one corresponds to the transition between oscillatory and stationary convective regimes (codimension-two point). We present measurements of this transition, adjusting well with the theoretical models cited before. The second one corresponds to an inverted-directed bifurcation transition point (tricritical point), where an inverted bifurcation exists associated with stationary instabilities going to a direct bifurcation for the same kind of instability.

2. Experimental Setup

The experimental setup consists in a layer of Homeotropic Nematic MBBA sandwiched between two circular glass plates (see Fig. 1). The thermal conductivity of the plates is six times larger

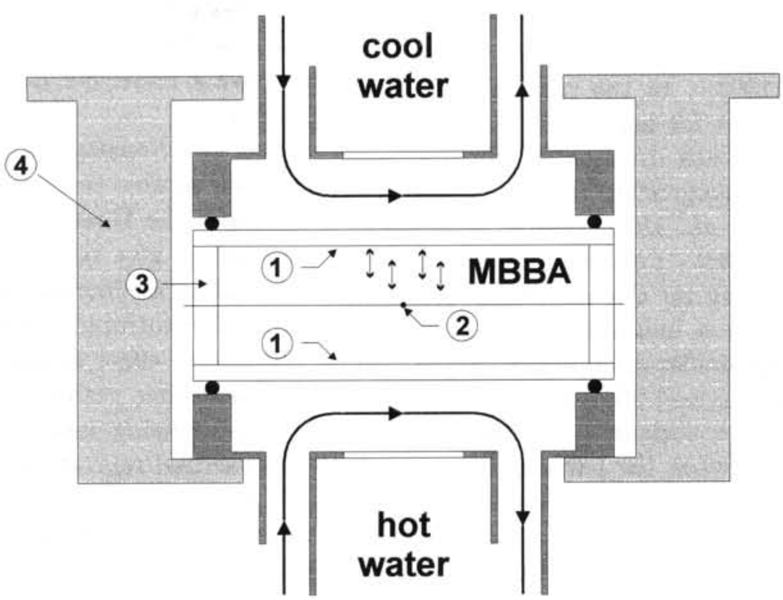

Fig. 1. Experimental cell. A thick layer of homeotropic MBBA is sandwiched between two transparent glasses (1). A set of thermocouples (2) is placed across the cell. The thickness of the cell is defined by a Teflon spacer (3). The cell is placed in a Copper container (4), which does not disturb the applied magnetic field.

than that of the Nematic film. The thickness of the Nematic film is $d = 8$ mm and the diameter of the cell is $D = 52$ mm, so the aspect ratio is $r = D/d = 6.5$. The temperature gradient is created by two independent water circulations over the outer faces of the cell, connected to thermally regulated baths (the oscillations due to the baths are about 0.01°C).

The Homeotropic alignment of the Nematic is obtained by precoating the inner face of the plates with a thin lecithin film. As the cell thickness is large, a magnetic field perpendicular to the plates is also used to improve the homogeneity of the alignment. The magnetic field is applied using a system of Helmholtz coils (range: 0–800 G, with a precision better than 5 G). The thickness of the cell is defined by a thick Teflon spacer ring. A set of three parallel 80 μm thick chromel-alumel thermocouples are placed crossing the cell in its mid-plane. The thermocouples are connected in pairs in a differential setup to reduce the average temperature effects.

The convection is easily detected optically. However optical tests are difficult because high interference effects appear even for small distortion orders (for example, a distortion of 1 degree in the homeotropic alignment corresponds to an optical path difference of 0.2 μm). Despite this difficulty, qualitative direct observations about the evolution and the propagation of spatial patterns are possible. Quantitative results were obtained by thermal measurements, which can be compared with the optical observations.

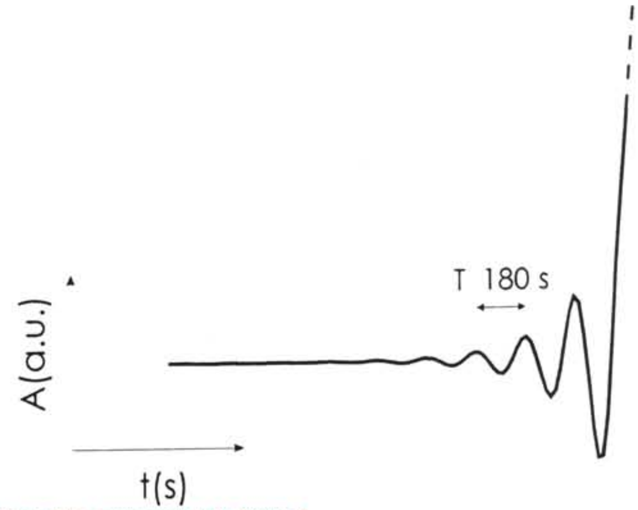

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Fig. 2. Applied magnetic field in dimensionless units versus the Rayleigh number. The critical Rayleigh number for $h = 0$ is calculated, making use of the linear regression method, and obtaining a value of $R_c(h = 0) = 2100$. The codimension-2 point corresponds to $h_c = 55.5$ and $R_c = 4100$.

3. Experimental Results

For every applied magnetic field, the measurements are made increasing the temperature difference between the plates in 0.1°C every 30 minutes by heating the lower plate, while the temperature of the upper one is kept constant and close to the room temperature. Following previous experimental [Guyon *et al.*, 1979] and theoretical results [Velarde & Zuniga, 1979; Dubois-Violette & Gabay, 1982], we can expect that for moderately strong applied magnetic fields (lower than 500 G in our experiments) a subcritical bifurcation exists near the linear threshold (ΔT_c). Then, it is necessary to increase the applied temperature gradient very slowly in order to approach ΔT_c as closely as possible. The onset of the convection is determined in two different ways: First, by direct observation because very small Director distortions produce strong light scattering of a beam crossing the cell, as it was said before (see Fig. 1). Secondly, recording local thermal fluctuations δT with the thermocouples placed into the cell, as it was cited in the previous paragraph. The fluctuations δT measured in the thermocouples remain constant in the conductive regime (for $\Delta T < \Delta T_c$). When the threshold is reached, the temporal register of δT changes to periodic oscillations, whose amplitude grow quickly, until a fully developed convective regime is reached (see for instance Fig. 2). This convective regime is characterized by very strong distortions on the Director orientation, showing a strong optical contrast between adjacent rolls.

In Fig. 3 we show the measured linear threshold ΔT_c for different magnetic fields using nondimensional variables, the Rayleigh number $R = \frac{\alpha \rho g d^3 \Delta T}{\nu \kappa}$ (where $\alpha = (-1/\rho)(\partial\rho/\partial T) > 0$ and ρ, ν, κ correspond to density, kinematics viscosity and thermal difusivity respectively. g is the gravity constant and ΔT the applied difference of temperature) as a function of the reduced magnetic field $h = H/H_f$, where H_f is the Freedericksz magnetic field defined as $H_f = q\sqrt{\frac{K}{\chi_a}}$ [de Gennes & Prost, 1993]. In this figure, we can distinguish two different regions for the threshold. A first region where a transitory oscillatory regime is detected, as described above, and a second one, for magnetic fields larger than 520 G where steady convective regimes are reached without oscillating effects. The quasi-linear

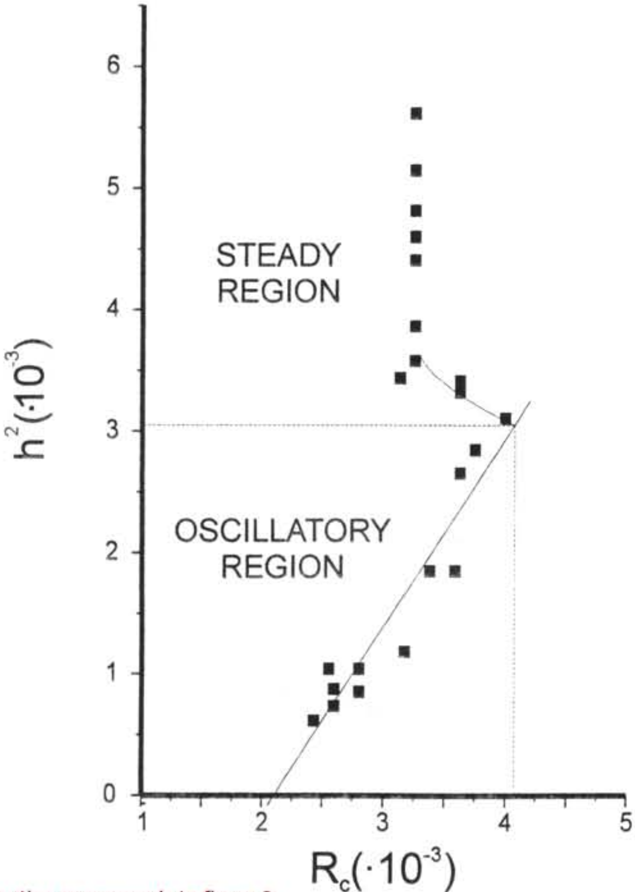

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Fig. 3. Representation of the thermal fluctuations behaviors close to the threshold. After one or more oscillations, the amplitude fluctuation increases up to a finite value.

variation of the critical Rayleigh number $R_c = R(\Delta T_c)$ with h^2 in the oscillatory region agrees with previous works, both experimental and theoretical [Guyon *et al.*, 1979; Feng, 1992; Feng *et al.*, 1992]. The transition between the oscillatory and the stationary threshold is obtained for $H_c \approx 520$ G. That is, h_c (8 mm) ≈ 54 using the non-dimensional variable $h_c = H_c/H_f$. A linear fit of the experimental results in the oscillatory region allows us to estimate $\Delta T_c(H = 0) \approx 2.7°$C.

On the other hand, in the region of steady modes, the ΔT_c for different magnetic fields larger than $H = 580$ G is nearly constant. It is interesting to remark that when H is very large, the critical thermal gradient is greater than $\Delta T_c(H = 0)$. The linear models [Guyon *et al.*, 1979; Velarde & Zuniga, 1979] predict that in the limit of very large H, $\Delta T_c(\infty) = \Delta T_c(0)$. Weakly nonlinear models [Dubois-Violette & Gabay, 1982; Feng, 1992; Feng *et al.*, 1992] only present results for finite magnetic fields.

In the oscillatory region only two or three oscillations are observed before the strongly nonlinear convective regime is reached. So, it has been possible to measure the period T of the oscillations only for magnetic fields between 300 and 400 G, obtaining $T \sim 180$ s (Previous experiments of Guyon *et al.* [1979] report transitory oscillations with periods of 120 s for $d = 5$ mm and $H = 400$ G). By direct visualization it seems that the transitory convective regime appears as traveling waves but more systematic experiments must be done in order to assure this. When the large amplitude convection is got, the register of δT show non-regular oscillations with periods greater than 10^3 s (for constant applied thermal gradients and field).

To characterize the existence of subcritical instabilities, a second kind of experiment is done. The procedure is to begin in the fully developed convective regime and then to diminish the thermal gradient in the same way as the experiment described above (0.2°C/hour). Observing the register of δT

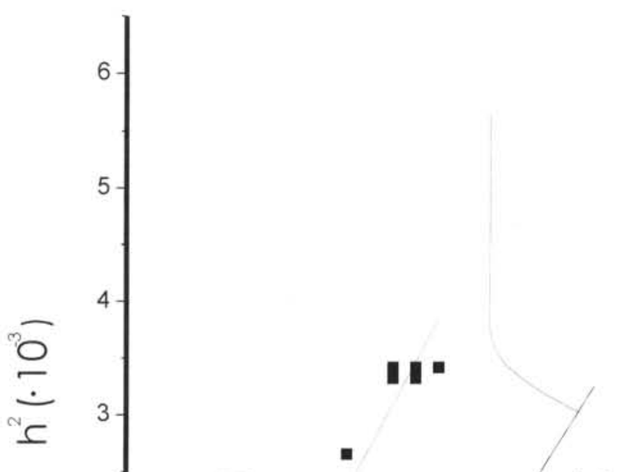

Fig. 4. Dependence of the measured thresholds for the disappearance of the convection versus the applied magnetic field in dimensionless units. The dotted line corresponds to the convection onset.

it is possible to detect where the convection disappears (the direct visual observations are difficult due to the large number of defects that remain after the disappearance of the convection). The variation of the measured thresholds $R_s(\Delta T_s)$ for this convection disappearance is shown in Fig. 4. For low magnetic fields this variation is quasi-linear and diverges from the thresholds of the convection onset ΔT_c defined before.

A second important result is that the subcritical thermal gradients ΔT_s still exist for the steady regimes region. Although in the present state of the experiments it is difficult to accurately set the point where the bifurcation changes from subcritical to supercritical (tricritical point), we can assure that for $H \leq 540$ G the bifurcation is still subcritical, while for $H \geq 580$ G and $\Delta T_c \approx 3.5°$C it is already supercritical. The observed spatial patterns in the region of oscillatory convection are rolls parallel to the wire of the thermocouples. The typical width of the rolls is the same as the thickness of the cell. For large enough times, the pattern did not change.

4. Discussion and Conclusions

As it was said before, the use of a very thick layer allows us to apply large enough magnetic fields to observe convective phenomena in the regions of oscillatory and stationary modes. In previous experiments [Guyon *et al.*, 1979; Alhers, 1996] several important regions could not been reached for two main reasons: The applied magnetic fields were not sufficiently intense, or the thermal thresholds imply non-Boussinesq effects. As it was said before, the experimental results for the variation of the linear thresholds with H, both in oscillatory and steady regions, adjust at least qualitatively well with the theoretical predictions, and with the previous experiments. The maximum of the measured thresholds (see Fig. 4) corresponds to the oscillatory-steady transition point of convective modes, $\Delta T_c(H = H_c) \sim 5.1°$C, in very good agreement with the predicted one: $\Delta T_c(H = H_c) = 5°$C [Feng, 1992]. Using non-dimensional parameters in [Feng *et al.*, 1992] it is easily obtained that $h_{ct} = H_{ct}/H_F = 51.2$ according to our experiments where $h_{ct} = 54$. A comparison of these results with the experiments for $d = 5$ mm. [Guyon *et al.*, 1979] is difficult because in that one the magnitude of $\Delta T_c(H = H_c)$ could not be measured and only an extrapolation is possible, giving a value of h_{ct} between 43 and 48. It is important to remark that the thermal thresholds in these experiments near the oscillatory-steady transition point correspond to differences of temperature greater than 10°C, so non-Boussinesq effects would be important, and then, the comparison has to take into account the influence of this effects. (In Nematic MBBA, there is a rapid variation of the viscosity coefficients with the temperature, especially near the transition to the isotropic phase, as was the case in the experiments for $d = 5$ mm [Gahwiller, 1973]).

A transitory oscillatory effect is observed for $H < H_{ct}$ in our experiments. Although the experiments do not allow us to study the variation of the frequencies with H, we can do so for $h \sim 35$, which corresponds to the predicted maximum of the frequencies after the recent theoretical model, $h = 32$ [Feng, 1992; Feng *et al.*, 1992].

The weakly nonlinear model cited predicts, in the oscillatory range, the existence of traveling and standing waves before the codimension-two point. Nevertheless, in this situation neither the traveling nor standing waves solutions should be stable. So, only transitory oscillations should be observed before the system reaches a large-amplitude solution that cannot be described by this small-amplitude model. A situation like this corresponds to a subcritical bifurcation. This kind of result was experimentally obtained in our experiments. Also, after our observation it seems that the first convective transitory regime corresponds to traveling waves. However, more systematic experiments would be necessary to assure it.

When the final large-amplitude state is attained, we observe temperature fluctuations δT with periods larger than 10^3 s. On the other hand, Alhers [1996] shows that the first spatial pattern of parallel rolls turns out to be unstable for very long times (with ΔT and H constants) near the threshold, due to a zig-zag instability leading to an spatially and temporally disordered patterns. This kind of result can explain our experimental findings about non-regular thermal fluctuations if it is considered that the evolution of the spatial pattern should produce fluctuations in the local thermocouples. In fact, the obtained times (10^3 s) are in the same order of magnitude as the characteristic times for the evolution showed by Alhers [1996]. Nevertheless, in our case, the existence of the thermocouples surely prevents the possibility of seeing the same kind of spatial pattern evolution.

About the measurements of ΔT_s, we can say that its variation with H in the oscillatory region agrees, at least qualitatively, with those obtained by Guyon *et al.* [1979] and by Alhers [1996]. A second result is that the subcritical behavior persists in a range of the steady regimes region. Only the theoretical models [Feng, 1992; Feng *et al.*, 1992] predict this kind of result. After our experiments we can place the tricritical point between $h_z = 60$ and $h_z = 64$ (to compare with $h_z = 61.3$ predicted by the theoretical models cited before).

We can conclude that the experimental results presented in this work agree well with the theoretical prediction, in particular with the most recent nonlinear model developed by Feng *et al.* [1992].

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