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The Observation of Hydrodynamic Instability in a Cholesteric Liquid Crystal Layer Having a Free Surface†

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It was for the first time that a hydrodynamic instability has been experimentally observed in a layer of a cholesteric liquid crystal (CLC) having a free surface and being locally heated from below. Steady structures of CLC motion have been obtained in the form of rotating (in the place) spirals consisting of distinct rolls with sharp boundaries; motion in every such roll was note also. It is supposed that the convective instability observed is due to Marangoni effect.

1 INTRODUCTION

When using thermosensitive cholesteric liquid crystals for studying plastic deformation of NaCl single crystals which is accompanied by local heat release in slip bands, we have observed, besides colour changes of CLC, a characteristic convective motion of liquid crystal in the regions of local heat release which could not be explained by only the effect of free convection, since spreading of CLC occured, therefore, resulting in formation of a groove in the CLC layer over the slip band. In the present work, the results of microscopic investigation of the phenomena in CLC are given.

[†]Paper presented at the Eighth International Liquid Crystal Conference, Kyoto, Japan, June 30-July 4, 1980.

2 EXPERIMENT

For investigation, NaCl specimens were used in the form of square prisms $(3 \times 3 \times 12 \text{ mm}^3)$ in size) which were cleaved out from NaCl single crystals on (100) cleavage planes. A scratch which served as a concentrator of stress was brought on one of the lateral prism faces in order to make the appearance of slip bands in a fixed site. The surface of the prism on which slip bands were expected to appear was covered by a layer of CLC (30–50 microns thick), the other three lateral faces were blackened to obtain better conditions for observing colour changes of CLC. Deformation was made by compression at a constant temperature with help of a microdynamometer placed in a thermostat. The free (horizontal) surface of the CLC layer was investigated in polarized and unpolarized reflected light as well as in polarized transmitted light.

CLC mixtures on the basis of cholesteryl alcanoates were used as CLC of high temperature sensitivity. Straining of NaCl specimens was conducted at the temperature at which the surface layer of CLC on NaCl was red-coloured.

To model the effect of streaming in a layer, a CLC mixture of low temperature sensitivity was also used containing 26% of cholesteryl chloride and 74% of cholesteryl oleate, as well as a nematic liquid crystal (NLC), viz. MBBA, a smectic liquid crystal (SLC) containing 59% of MBBA and 41% of pentyl cyanobiphenyl, and two isotropic liquids (vaseline and vacuum grease) of similar (to LC) viscosity.

As a source of local heating in modeling experiments, a light beam was generated with the help of a Nu-2 type microscope (with objectives \times 25 or \times 50). The intensity of the beam was varied by the objective aperture. The modeling experiments were carried out at room temperature.

3 EXPERIMENTAL RESULTS AND THEIR DISCUSSION

The original CLC layer was red-coloured and had a planar texture consisting of shining confocal domains of finest size. In a fraction of a second after the deformation onset, a long, narrow green-coloured band appears on the red-coloured background beginning at the stress concentrator (Figure 1). The band is of some tens microns in width and to several millimeters in length, and is situated at 45° to the [100] direction (which may be considered coinciding with the edge of the specimen on which the scratch is visible in Figure 1). The green colour of the band can change to blue or violet depending on the intensity of the heat release which is determined by the deformation rate. All the colour changes were very rapid; it was as if the band "flashed out". Just a little later, red-coloured parts of the CLC layer round the band also became

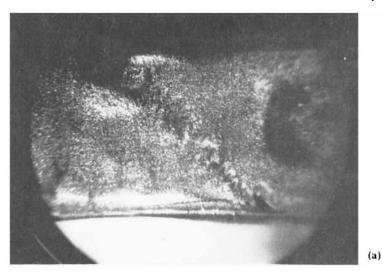


FIGURE 1 The structure of convective motion in CLC layer over a slip band during deformation of a NaCl single crystal. Unpolarized light, x20. a) the onset of CLC movement causes a distortion of the initial texture of the CLC layer; b) a spiral structure of CLC motion; c) a residual picture of CLC motion observed in 1-3 min after the deformation had stopped; d) re-generation of a spiral structure after the deformation recommencing.



green. The location of the band corresponds to slip bands generated during deformation of the NaCl crystal and consisting of edge dislocations, which is confirmed by selective etching of the specimen after deformation as well as by photoelasticity method.² When, during deformation, two slip bands occur

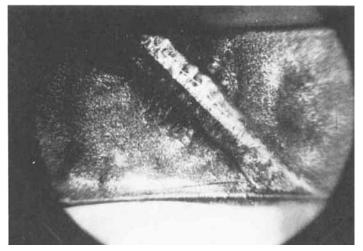


FIGURE 1(c)



(d)

consisting of edge dislocations, two green-coloured bands appear in CLC layer at the concentrator of stress, directed at 45° towards [100] and at 90° towards each other (Figure 2).

Almost instantly following the colour change, the CLC begins to move on all the areas of the band which leads to a disturbance of the original CLC texture (Figure 1(a), the initial stage) and to the appearance of characteristic motion structures illustrated in Figure 1 and 2. Since, when photographing,

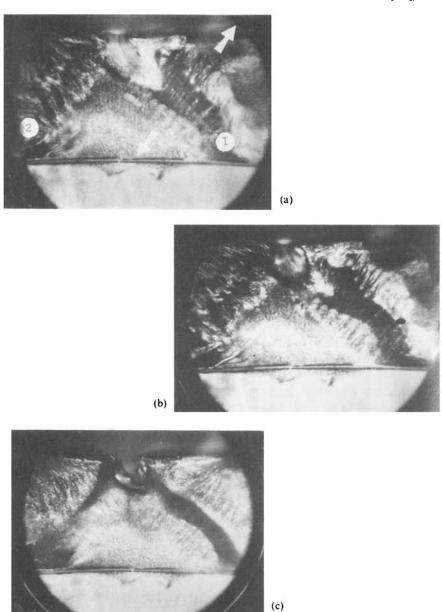


FIGURE 2 Streaming of CLC away from the slip bands during deformation of NaCl single crystal. Unpolarized light. x20. a) a fine structure of the CLC flow over the slip band; b) thinning of the CLC layer over the slip band (dark region) and beginning formation of rolls (on both sides of dark bands 1 and 2); c) complete streaming of CLC away from the slip bands.

deforming of the NaCl specimen was for a while stopped (although the load not taken off), the microphotographs presented do not reflect clearly enough the true picture of CLC motion which is observed at a moment of the deformation of the NaCl crystal. In fact, the photographs show only a residual picture of CLC motion which relaxes in time (Figure 1(c)), but begins again when deformation returns (Figure 1(d)). As a microscopic investigation shows, a characteristic feature of CLC motion here is that the CLC streams away from the slip band on either side of the band in a perpendicular direction (Figure 2 (a) and (b); the directions of streaming are indicated by arrows) and, as the streaming continues, the CLC layer thickness over the band decreases (Figure 2 (a) and (b); the band appears on photographs more and more dark). A complete streaming of CLC away and stripping of the band can occur (Figure 2 (c)) resulting in formation of grooves with thickenings on their sides, which leads to a change of the original surface topography of the CLC layer. The grooves may be clearly visible on the background of coloured CLC layer even to the naked eye and are retained unchanged for a long time after stopping the deformation. By this way one can visualize and preserve just extremely small regions of local surface heating (surface thermophotography) which is used presently in practice to study thermal processes during plastic deformation of crystal solids.

Another remarkable feature of the process of CLC flowing over a slip band during deformation is that the flow is not continuous, but breaks (more or less regularly along the band) into narrow tongues with distinct boundaries (Figure 2 (a)). In some cases, a counter-flow of similar structure develops; an interaction of the opposite streams results in formation of stationary motion structures which appear as spirals rotating in place and consisting of "rolls" (coils) with distinct boundaries (Figure 1 (b) and (d); Figure 2 (b)); motion of CLC in each roll can be clearly seen.

This process of the formation of spirals takes place as a rule on both sides from the slip band (Figure 2). Sometimes, however, perhaps due to a disturbance of the thermal conditions, such spirals occur on only one side of the band (Figure 1). Figure 3 shows a fine structure of rolls in the spirals. Small shining confocals are regularly oriented (Figure 3) and form threads and ribbons which are directed perpendicular to the slip band (a dark region in Figure 3). At the tip of a slip band, movement of CLC in concentric circles occured sometimes (Figure 4).

Thus, it can be seen from the experiments on local heating from below a CLC layer having a free upper surface that a hydrodynamic instability is generated in such a CLC layer. Since heating is confined to a region compared in width with that of the slip band, two temperature gradients develop as a matter of fact in conditions of such narrowly localized heating: a vertical one (normal to the CLC layer) and a horizontal one (parallel to the free surface

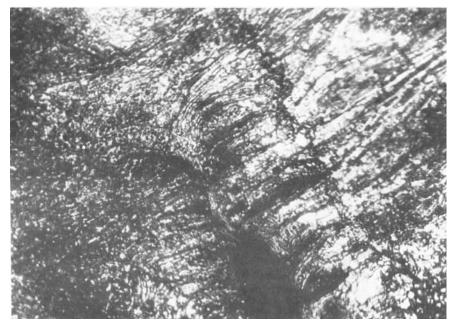


FIGURE 3 The texture of spiral coils. Polarized light.

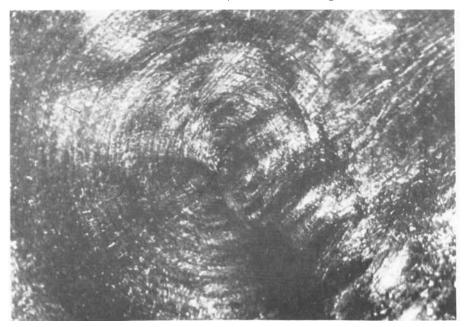


FIGURE 4 A rotational structure of CLC motion at the tip of a slip band. Polarized light.

of the CLC layer). It is known that a vertical temperature gradient in a horizontal liquid layer in 1–g environment may cause the so-called Bénard convective instability.³ A temperature gradient directed along the free surface of a layer results in a gradient of surface tension which generates a surface flow of the substance from hot to cold regions. This is the so-called Marangoni convection.^{4,5}

Theory shows that in isotropic liquids and in NLC these two effects can superpose and act in line with each other; in such cases the instability occurs if the condition $R/R_c + M/M_c = 1$ is met, i.e. if the Rayleigh and Marangoni numbers are less than critical ones. In thin layers of isotropic liquid or NLC, the Marangoni instability will occur at a less ∇T than the Bénard instability. This is due to a different dependence of critical numbers R_c and M_c on layer thickness d ($R \sim d^4$, while $M \sim d^2$). The calculations show that, in the NLC layer of 1 mm thick, Bénard convection should occur at $\Delta T = 3$ °C, but only 0.2°C is needed for Marangoni convection to begin.

In recent years, a great attention was attracted to the Marangoni convection in connection with the development of materials science experiments in space. In particular, a number of works concerned with the research of Marangoni convection in floating zones has appeared.^{5,7,8} Minimization of natural convection in these experiments was achieved by use of a cylindrical floating zone small in size (the zone length less than 4.8 mm and zone diameter less than 6 mm). In such geometry, Marangoni convection flows take the form of toroidal vortices.^{5,8} When locally heating a floating zone of silicone oil by a ring heater, a motion picture was obtained in the form of two toroidal vortices situated symmetrically on both sides of heater (see Figure 2 in [8]).

The Marangoni convection in liquid crystals was observed also in experiments on locally heating from above, with the help of a laser beam, a free layer of MBBA from 60 to 200 microns thick. The effect manifested itself in radially streaming the MBBA away from the "hot" point. The streaming caused a disturbance of molecular arrangement, which was revealed by the appearance of a specific optical picture. If the local heating was powerful enough, the flow penetrated deep into the layer and led to thinning the layer up to complete streaming the substance away and stripping of the most hot areas (formation of craters in the layer).

Cholesteric liquid crystals proved to show a more complex behavior in comparison with isotropic liquids or NLC. The Bénard or Marangoni convection has not yet been observed in CLC.¹⁰ The analysis of the theoretical research concerned with the Bénard mechanism in CLC¹¹ permits one to conclude that a spiral character of molecular structure in CLCs makes them much more stable with regard to the onset of Bénard convection and requires a greater temperature gradient, in comparison with NLC. A theory of Marangoni convection in CLC is unavailable.

The experimental data we have obtained are the first observations of convective instability in cholesteric liquid crystals induced by temperature gradients. A classical Bénard effect concerns with a horizontal layer heated uniformly on all the (infinite) area from below (or, in the case of NLC, both from below and, in certain conditions, 12 from above). In our experiments, heating was conducted from below within a narrow, long ("infinite") band, and steady convective motion of CLC in such geometry may be considered "a linear Bénard effect", but since the layer has a free surface and in the initial stage, the movement occurs in the form of streaming of the substance away from the hot region we suppose that in our case the observed stationary convective motion is caused predominantly by the Marangoni mechanism.

According to our estimates,² the temperature in slip bands consisting of edge dislocations increases by 0.3-0.8°C. A temperature gradient on the surface of the CLC layer induced by this heat release within a narrow band results in streaming of the substance away from the band and the formation

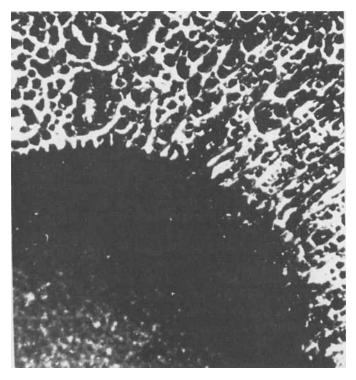


FIGURE 5 Radial streaming of CLC away from the hot region (heating with help of a light beam). Transmitted unpolarized light.

of rotating spirals situated symmetrically relative to "the heater" (Figure 2). The picture observed is similar to one visible when locally heating silicone oil.8 The geometry of local heating, however, appears to influence the character of the streaming process. In our experiments the heater is linear, and the CLC motion occurs predominantly in directions perpendicular to the heater and leads to the formation of rolls (Figure 2). In experiments with point heating using a laser beam (see Urbach et al.9), the NLC streams radially away from the hot point, but stationary motion structures are not generated. We have conducted experiments similar to those of Urbach et al.,9 for isotropic liquids and various types of liquid crystals and also have not obtained stationary motion structures. For all the substances studied, only streaming of the substance away in radial directions was observed (Figure 5) and, in the case of more severe heating, craters formed (Figure 6). After the local heating ceased, the craters in the layers of isotropic liquids and NLC disappeared and the surface of the layer relaxed to the initial state; in CLC and SLC layers the recovery did not occur. In NLC layers, interference rings have been observed in the areas where the layer thickness had decreased (Figure 7). In more

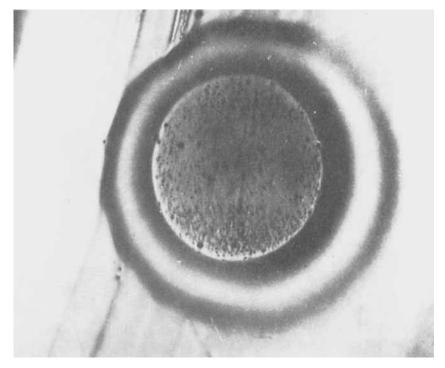


FIGURE 6 Craters in CLC layer. Transmitted unpolarized light.

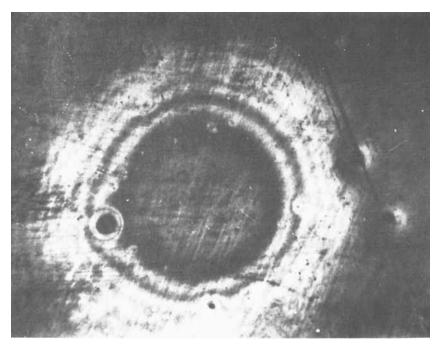


FIGURE 7 Interference rings in the region of local heating in NLC layer. Transmitted unpolarized light.

detail, these experiments were described elsewhere.¹³ Presently, the study of convection in CLC is in progress for CLC mixtures of low temperature sensitivity, having planar or distorted planar texture.

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