Effect of a narrow light beam on a smectic liquid crystal

A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, and L. Csillag

P. N. Lebedev Physics Institute, Russian Academy of Sciences

(Submitted 15 December 1992)
Zh. Eksp. Teor. Fiz. **103**, 1609–1618 (May 1993)

A memory of director-field distortions caused by a narrow light beam has been observed in an octylcyanobiphenyl smectic liquid crystal with a good homeotropic orientation. A very narrow light beam of high power density distorts the smectic layers in a region with dimensions thousands of times the cross-sectional dimensions of the beam. The distortion pattern is axisymmetric and annular. It contains various textures, including polygonal, and also a Helfrich–Yuro periodic modulation of the layers.

A light beam produces optical inhomogeneities in the smectic liquid crystal octylcyanobiphenyl (OCBP) near the transition to the nematic phase.[1] These inhomogeneities may "freeze" in the crystal and remain there for a long time (this is a memory effect).[1)] An extremely wide variety of structures may freeze in the crystal, depending on the intensity of the light beam and the proximity of the crystal temperature to the phase-transition point. This diversity is evidence of the complexity of the processes which are induced in the smectic liquid crystal by the light beam.

The experiments of Ref. 1 were carried out at a crystal temperature 0–2° away from the temperature of the smectic-nematic phase transition. It was not possible to extend this range substantially in Ref. 1, although on occasion an effect of the light on the crystal was also observed at temperatures 5–7 °C below the transition temperature.

The effect of laser light on a crystal depends on the light intensity. An upper limit is set on this intensity by the particular laser at the disposal of the experimentalist. However, it is a simple matter to increase the power density by using a short-focal-length lens to focus the light into the crystal. We used a lens with a focal length ~ 20 mm in the present experiments. Below we report a study of the effect of the narrow light beam formed by this lens on the smectic phase of the OCBP crystal at room temperature.

As in Ref. 1, the experiments were carried out with homeotropically oriented samples of OCBP crystals. The sample thicknesses were $L_1=120$ and $L_2=40$ μm.

The beam from a Carl Zeiss ILA-120 argon laser, with a wavelength $\lambda=4880$ Å, was focused on the crystal. Behind the crystal, in the beam path, was a film analyzer. On a screen behind the analyzer we watched the changes which occurred in the beam as it interacted with the liquid crystal. The dynamics of the changes occurring in the crystal itself could also be monitored on the screen with the help of a $\sim 400\times$ microscope system.

The structures which were induced in the crystal by the beam, and which froze in the crystal when the beam was cut off abruptly, were studied with the help of a Polam L-213 polarizing microscope.

Let us summarize the results.

I. OCBP SAMPLE WITH A THICKNESS $L_1=120$ μm

1. We were unable to observe any effect of the beam on the crystal at a laser power $P<200$ mW (at a power density $P\sim 250$ kW/cm^2).

2. An effect of the laser light on the smectic liquid crystal becomes noticeable at $P>200$ mW. The primary effect is a change in the beam divergence and in the complexity of the beam structure. Several systems of rings appear on the screen at a time T_d after the light is first applied to the crystal. The first system is a system of rings of an orientational aberrational self-focusing (which accompanies a Fréedericksz transition in the field of the light wave). Immediately after the appearance of the first system of rings, and against this first system as a background, we observe the appearance of a system of three or four broad, equidistant rings (the maximum divergence of the beams forming this system is 20–25°; the angular size of the first ring is $\sim 2°$). Other systems are also observed. The time T_d depends on the power of the light beam, P, and on the sharpness of the beam focusing in the crystal. It ranges from 10 s to several minutes.

3. The pattern on the screen is not static. The divergence of the beams forming the system of rings increases and decreases; the rings themselves become distorted, but they exist for only a finite time (T_l), 1–5 min. The rings then contract toward their center and disappear ("collapse"). After this event, yet another (the last) system of equidistant rings appears. The intensity at the center of the beam transmitted through the crystal is quite low. Small bright spots are observed over the entire field on the screen. From time to time, these bright spots shift slightly and vary in intensity. Sometimes they complicate efforts to observe the last system of rings, but no substantial changes involving the beam occur from this point onward.

4. If the laser beam is blocked for 2–3 min after the collapse of the rings, we find changes in the original orientation of the director over a huge region (~ 7 mm in size) around the zone of the light beam. These changes depend strongly on the distance from the center of the beam. A complex annular structure forms in the crystal (Fig. 1). The details of this structure can be seen under a micro-

 1063-7761/93/050788-06$10.00

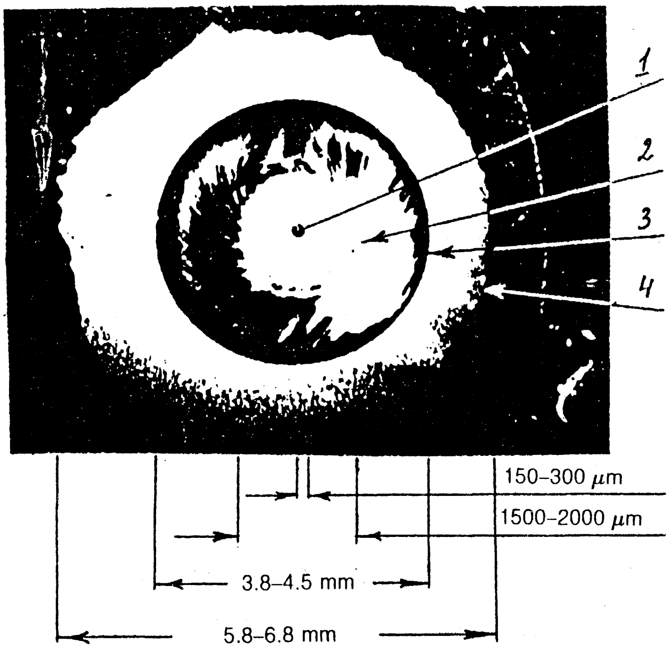

FIG. 1. Annular structure which is formed by a narrow light beam and "frozen" in an OCBP crystal ($L_1 = 120\,\mu$m). *1*—Black ring-spot; *2*—ring of "polygonal" texture; *3*—striated ring; *4*—"grid ring." (This photograph was taken by the dark-field method.)

scope (these observations were carried out 10–20 s after the illumination of the crystal was ended; Figs. 2 and 3). In crossed polarizers (Fig. 3a) one can see a dark spot at the center. The dimensions of this spot range from 150 to 300 μm in different experiments. In this spot we can see part of a dashed bright ring, which is evidence that the dark spot is not uniform. In parallel polarizers, the dark spot is ring-shaped (Fig. 3b; the characteristic dimensions can also be seen here). This dark ring-spot (depending on whether the polarizers are parallel or crossed) is enclosed by a ring 1500–2000 μm in diameter (1.5–2 mm), which has an approximately polygonal texture (frame 2 in Fig. 2), then by a "striated" ring (frame 3 in Fig. 2; the characteristic dimensions of this ring are 3.8–4.5 mm), and, finally, by a ring with an outer diameter of 5.8–6.8 mm, consisting of a fine-scale square grid (frame 4 in Fig. 2; the dimensions of a cell are 6–7 μm). The inner boundaries of all the rings except the striated one are very sharp. As time elapses, the rings shrink. The rate of this shrinkage depends on the duration of the illumination of the crystal.

5. An increase in the duration of the illumination of the crystal after the collapse leads to slower changes in the characteristic dimensions of all the rings which remain in the memory of the crystal, except those of the dark ring-spot. This feature converts fairly rapidly (within 1–3 h) into a blurred "star" structure against the background of a polygonal texture. All the remaining parts of the pattern can then persist for a long time—several days.

6. If the beam is cut off before the rings collapse or

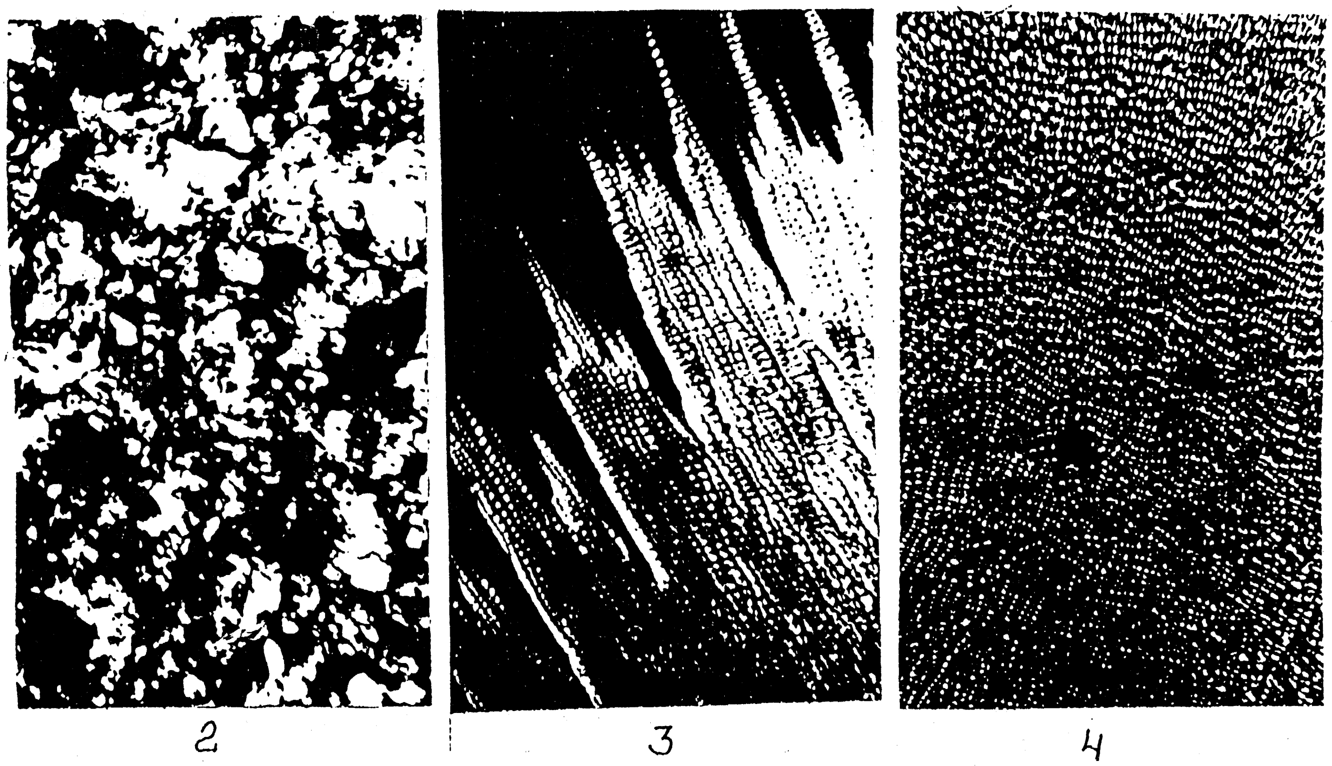

FIG. 2. Fragments of rings frozen in a crystal. *2*—Polygonal texture; *3*—outer edge of striated ring; *4*—grid. (The photographs were taken through a Polam L-213 microscope.)

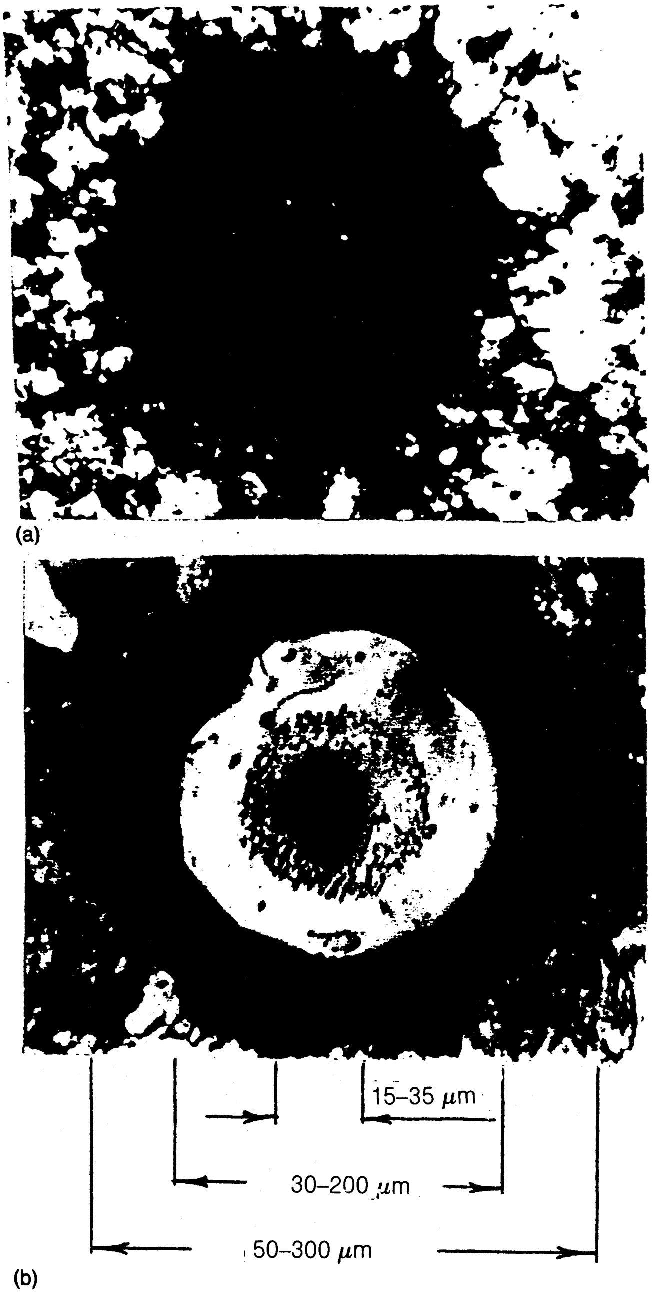

FIG. 3. Photographs (taken through a Polam L-213 microscope) of the black ring-spot. a—In crossed polarizers; b—in parallel polarizers.

immediately after they collapse, the pattern differs primarily in dimensions. These dimensions are much smaller. (If the laser beam is cut off after 15 s, the diameter of the grid ring is 1800, that of the striated ring 900, and that of the polygonal-texture ring 500 μm. If the beam is cut off after 30 s, the dimensions of these rings are 2500, 1200, and 600 μm, respectively.) Furthermore, the "striated" ring is no longer striated. It has no striations in either crossed or parallel polarizers, and it differs in no way from the rest of the crystal which have not been affected by the beam. The ring-spot is usually either a black spot or a black point (with minimum dimensions of 3 μm). Less often, it is a black ring. The most noticeable changes which occur as time elapses are in the dimensions of the ring with the grid pattern. It contracts at a rate of 70–150 μm/min to its inner diameter.

7. After a few days, spots 100–400 μm in diameter (depending on the duration of the illumination) remain in the places affected by the laser beam. These spots indicate that irreversible changes have occurred in the smectic layers in these places. These spots do not disappear even if the crystal is heated into the isotropic phase in a constant-temperature chamber and then cooled slowly, i.e., even under conditions such that all the distortions of the homeotropic orientation would ordinarily be erased.

8. Examination with the microscope system provided further information. It showed that the laser beam first contracts at a time T_d after the beginning of the illumination of the crystal, and a small bright ring appears on the screen. In crossed polarizers, one usually observes a cross which indicates the onset of an anisotropic formation in the crystal.

The changes in the illumination of the screen which then occur are evidence of a restructuring of the crystal around the light beam. This restructuring terminates in the formation of a black ring (in observations without an analyzer) or a black spot (in crossed polarizers). Outside the ring-spot, a wavefront with a double boundary propagates at a velocity which gradually decreases. After 10–50 s (depending on the beam power) this double boundary essentially comes to a halt. In crossed polarizers, the region enclosed by this boundary becomes noticeably darker. A "frozen" ring of polygonal texture is observed in the crystal if the beam is cut off abruptly under these conditions. The outer diameter of this ring is 20–25% smaller than the diameter of the wave front. We were unable to study the dynamics of the striated ring or that of the grid-pattern ring, since observation of these rings was obstructed by the intense parasitic scattering of the laser light, in the crystal itself and also by all the optical elements of the experimental apparatus.

The stability of the pattern frozen in the crystal improves with increasing duration of the illumination. If the crystal is illuminated for a few minutes, the frozen structures live a long time (days or even weeks). The only exceptional case is that of the black spot-ring, as we mentioned earlier. During repeated application of the light beam to the frozen formations, a slight change occurs in the dimensions of the black ring-spot and in those of the polygonal texture. This change depends on the relation between the illumination time and the time during which the crystal is not illuminated. If the intensity of the light beam is very slowly reduced after the illumination, on the other hand, we observe either the "erasure" of all the rings except the black ring-spot or the erasure of the rings of the polygonal and striated texture, while the grid-pattern ring survives and retains its dimensions. The latter events occur when there is simply a black spot rather than a ring-spot at the center of the pattern. A grid may also arise in the position of the polygonal and striated texture.

9. Estimates of the size of the formations which diffract

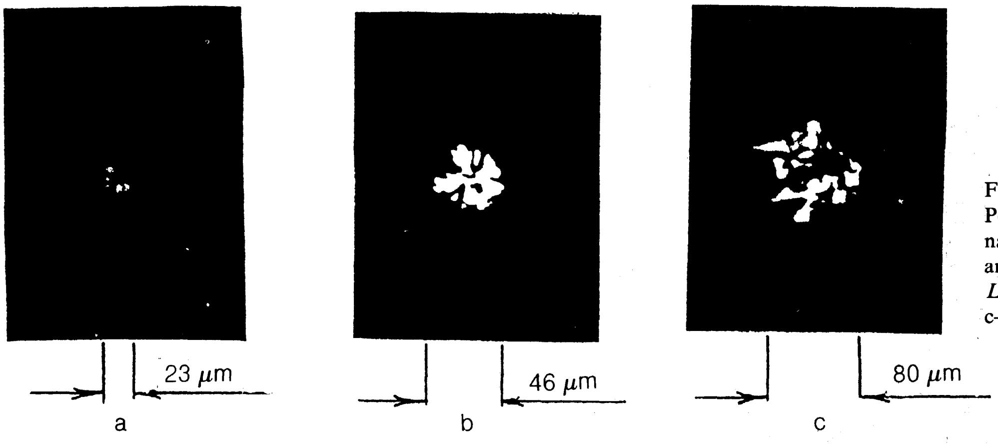

FIG. 4. Photographs (taken through the Polam L-213) of distortions caused by a narrow light beam in the smectic phase of an OCBP liquid crystal (with a thickness $L_2=40$ μm). a—$P\sim200$ mW; b—~270; c—~380 mW (crossed polarizers).

the beam found on the basis of the pattern of equidistant rings yield ~3 μm for the system of wide rings and ~200 μm for the system which is the last to appear.

II. OCBP SAMPLES WITH A THICKNESS $L_2=40$ μm

The application of a narrow light beam to a homeotropically oriented smectic phase of the OCBP crystal with a thickness a third of L_1 leads to some completely different results. We should first mention the pronounced sensitivity to the precision of the focusing of the beam in the crystal with L_2. Also comparatively prominent is the dependence of the result of the illumination on the intensity of the light beam. Figure 4 shows the intensity dependence of the result of the application of a narrow beam. We see that the dimensions of the region in which the director field is altered are small. According to our estimates, the size of the beam waist formed by a lens with $f\sim20$ mm is ~10 μm. Consequently, the changes which occur in the director field in the crystal with $L_2=40$ μm occur in a region which differs in size from the waist by a factor of 6–20, in contrast with the factor of several hundred in the case of the crystal with $L_1=120$ μm.

The pattern observed on the screen is usually as follows. Immediately after the beginning of the illumination, the analyzer field brightens slightly. After 10 s to several minutes, a cross appears. The dimensions of this cross gradually decrease, and two or three systems of arcs appear around the cross.[1] In the absence of an analyzer, these arcs represent two or three rings.

As in the experiments with the crystal with L_1, we observe a system of equidistant rings. This system usually arises immediately after the decrease in the dimensions of the cross. The distance between the rings decreases as time elapses; it reaches a constant value after 1–2 min. Estimates of the size of the anisotropic region which does the diffraction yield 50–80 μm, depending on the beam intensity.

DISCUSSION OF RESULTS

The pattern observed on the screen tells us first that the crystal is heated in the beam zone, and is ultimately heated by at least 18–19°, i.e., into the isotropic phase ($t_{\text{n-is}}>40$ °C, $t_{\text{room}}\approx21$–22 °C). Our reasoning here is that the only possible explanation of the collapse of the rings of the orientational aberrational self-focusing which always concludes the process is a transition to the isotropic phase. However, the beam does not "burn through" the crystal as rapidly as one might expect if the effect were purely thermal. The time which elapses from the instant at which the beam is first applied to the collapse of the rings is at least a minute; at the lowest beam intensities at which traces of the effect of the beam remain in the crystal, this time is several minutes. One might suggest that the part of the crystal in the beam zone is first heated to the nematic phase. In this case there would of course by a Fréedericksz transition in the light beam, since we have $P>P_{\text{thr}}$ (in our experiments we have $P>200$ mW, in comparison with $P_{\text{thr}}\sim20$ mW, according to the results of Ref. 4). Evidence for this conclusion comes from the pattern of the orientational aberrational self-focusing. The overall nonlinear divergence of this pattern increases sharply in comparison with that in broader light beams (when longer-focal-lens are used),[6] in complete agreement with the theory of Ref. 5. The absorption of light by a nematic liquid crystal distorted by the Fréedericksz transition in the beam zone may be slightly higher than that of a smectic liquid crystal in a good homeotropic orientation. Furthermore, at the power density ($\mathscr{P}>250$ kW/cm^2) provided by the short-focal-length lens, chemical processes can also occur in the material of the crystal and in the orienting material on the walls of the cell. The result may be a further increase in absorption. The crystal is ultimately burnt through, and the process comes to a halt.

We should now understand why a pattern of such texturally different rings arises and remains in the crystal (why a memory effect occurs).

As we know, a thermal effect can alter the thickness of a smectic layer. Because of the stress which arises in the process, it may give rise to long-lived periodic deformations (a grid) in the crystal.[7]

By virtue of its absorption, a light beam heats a crystal and gives rise to a grid. As the heating proceeds, i.e., as the illumination time is increased, the grid arises in regions progressively farther from the beam axis. In the region close to the light beam the temperature reaches a value $t > t_{\text{n-is}}$ ($t_{\text{n-is}}$ is the temperature of the transition to the isotropic phase). The smectic phase persists slightly longer, to $t > t_{\text{sm-n}}$ ($t_{\text{sm-n}}$ is the temperature of the transition from the smectic phase to the nematic phase), at a certain distance from the axis of the light beam.

We can estimate the time over which the crystal is heated by the laser beam. The crystal thickness (120 μm) is much smaller than the thickness (3000 μm) of the plates of glass which form the cell holding the crystal. In a first approximation, we can thus take the crystal heating time to be the time over which the cell walls are heated. The time evolution of the temperature change $\Delta t = t - t_0$ at a point at a distance r from a point heat source is given by[8]

$$\Delta t(r,T) = \frac{q}{4\pi k} \frac{2}{\pi^{1/2}} \int_{\frac{r}{2aT^{1/2}}}^{\infty} e^{-\alpha^2} d\alpha, \tag{1}$$

where k and $a^2 = (k/c\rho)$ are the thermal conductivity and thermal diffusivity, respectively, q is the intensity of the heat source, and ρ and c are the density and specific heat. The relaxation time T_{rel} for a nearly steady-state temperature can be found from (1) by setting $\Delta t(r,T)/\Delta t(r,T = \infty) = 1 - \nu$ where ν is the error of the determination.

In this approximation we have

$$T_{\text{rel}} = 2r^2/\pi a^2 \nu^2. \tag{2}$$

For glass we would have[9] $k = 1.6 \times 10^{-3}$ cal/(cm · s · deg), $\rho = 2.5$ g/cm^3, and $c = 0.18$ cal/deg. If we set $\nu = 0.3$, then with $r = 0.5$ cm (the distance at which the grid usually appears) we find $T = 250$ s. This figure agrees in order of magnitude with the illumination time required for the appearance of a grid at this distance.

One might suggest that the black ring—a region with a greatly disrupted director orientation—is the boundary between the isotropic and nematic phases, while the boundary between the nematic and smectic phases is the double wavefront boundary. When freezing occurs in the region of the nematic phase, a polygonal texture arises in the region which is the region furthest from the axis of the light beam but still subjected to heating–the grid. In the region between them, a striated ring with a confocal texture arises. This texture may, as in Ref. 10, be due to some tilting of the director field with respect to the walls of the cell (in Ref. 10, this tilting was arranged by treating the cell walls in a special way; in our own case, the tilting results from a transition from a smectic region with a fairly good homeotropic orientation (the ring made up of the grid) to a region in which the director orientation changes from point to point (the ring of polygonal texture).]

The outer and inner diameters of the black ring depend on the intensity and duration of the illumination, as we mentioned earlier. If both the duration and the intensity are low, a ring does not appear at all. Instead we find either a black spot or a black point—a region which, like the ring, has a greatly distorted structure, since it is observed in both crossed and parallel polarizers. The subsequent dissipation (conversion into a "star" or relaxation) of the black ring, which occurs more rapidly than the dissipation of the overall system of rings, indicates that the most distorted layers relax more rapidly.

Let us compare the period of the grid with the period of the Helfrich–Yuro instability. The latter period is given by[11]

$$\Lambda = 2(\pi d L)^{1/2}, \tag{3}$$

where d is the reduced length, which is comparable in order of magnitude to the layer thickness p (d may be greater than p near a phase transition). For OCBP we have[12] $p = 30$ Å, and an estimate from (3) yields $\Lambda = 2$ μm. This figure is slightly smaller than the $\Lambda = 6$–7 μm found experimentally.

We turn now to the systems of equidistant rings which are observed on the screen. It is natural to suggest that one of these systems is the result of diffraction of the beam by a "hole" which is "burnt through"[2)] the crystal by the central part of the light beam. This conclusion is implied by the size (3 μm) of the inhomogeneity which causes the diffraction, as estimated from the angular distances between the rings. Further evidence for this conclusion is that this system of equidistant rings appears when the system of rings of the orientational aberrational self-focusing collapses, i.e., when the nematic phase in the central part of the beam zone begins a transition to an isotropic liquid.

As the crystal is heated further, the zone occupied by the isotropic liquid becomes larger. When it reaches its maximum dimensions for the given intensity (a black ring appears on the screen at this point), we see the last diffraction pattern, which persists after the process ends. Estimates show that the dimensions of the region causing the diffraction are $\sim$200–400 μm; i.e., they correspond to the dimensions of the black ring-spot.

The apparent reason for the pronounced difference between the effects of the intense light beam on the crystals with $L_1 = 120$ and $L_2 = 40$ μm is the important stabilizing effect of the crystal walls in the case of the thin sample.

Let us summarize. Blue-green light of high power density, when turned off abruptly, remains in the memory of a smectic liquid crystal for a fairly long time, in the form of a complex system of rings which fill a region with dimensions thousands of times those of the light beam.

In a region with dimensions several tens of time those of the light beam, an irreversible distortion of the smectic layers occurs.

In thinner crystal samples, the dimensions of the region in which the layers are distorted by the light beam are smaller by a factor of hundreds.

We wish to thank D. B. Terskov for preparing the OCBP sample with a thickness of 40 μm.

[1)]A memory effect had been observed previously[2,3] in smectic liquid crystals in which measures had been taken to greatly intensify the absorption.

[2)]By "burnt through" we mean a heating of the liquid crystal in the region of the laser beam to the point that it goes into the isotropic phase, as in

Ref. 13, where this effect was observed in a broader light beam in an OCBP crystal near the phase transition from a nematic liquid crystal to an isotropic liquid.

[1] A. S. Zolot'ko, V. F. Kitaeva, and D. B. Terskov, Zh. Eksp. Teor. Fiz. **101**, 1827 (1992) [Sov. Phys. JETP **74**, 974 (1992)].

[2] F. J. Kahn, Appl. Phys. Lett. **22**, 111 (1973).

[3] G. N. Taylor and F. J. Kahn, J. Appl. Phys. **45**, 4330 (1974).

[4] L. Csillag, I. Janossy, V. F. Kitaeva, N. Kroo, and N. N. Sobolev, Mol. Cryst. Liq. Cryst. **84**, 125 (1982).

[5] A. S. Zolot'ko, V. F. Kitaeva, N. N. Sobolev, and A. P. Sukhorukov, Zh. Eksp. Teor. Fiz. **81**, 933 (1981) [Sov. Phys. JETP **54**, 496 (1981)].

[6] V. F. Kitaeva and A. S. Zolot'ko, Laser Research in USSR **10**, 275 (1989).

[7] R. Ribotta, J. Phys. (Paris) **37**, C3 (1976).

[8] A. N. Tikhonov and A. A. Samarskii, *Equations of Mathematical Physics*, Pergamon, New York, 1964.

[9] E. M. Voronkova, B. N. Grechushnikov, G. I. Distler, and I. P. Petrov, *Optical Materials for Infrared Technology*, Nauka, Moscow, 1965.

[10] N. J. Chou, S. W. Depp, M. H. Eldrige, G. J. Lee, A. Sporkel, and J. Brown, J. Appl. Phys. **54**, 1827 (1983).

[11] P. G. de Gennes, *The Physics of Liquid Crystals*, Oxford Univ. Press, London, 1974 (Russ. Transl. Mir, Moscow, 1977).

[12] Y. Ouchi, Y. Takanishi, H. Takezoe, and A. Fukuda, Jpn. J. Appl. Phys. **28**, 2547 (1989).

[13] V. F. Kitaeva, N. N. Sobolev, A. S. Zolot'ko, L. Csillag, and N. Kroo, Mol. Cryst. Liq. Cryst. **91**, 137 (1983).

Translated by D. Parsons