Light scattering by structures created by a laser beam in an OCBP liquid crystal near the smectic-nematic phase transition; memory effect

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The interaction of a laser beam with a homeotropically oriented OCBP liquid crystal has been studied near the smectic-nematic phase transition. The light beam gives rise to a region which is optically inhomogeneous and anisotropic. This region "freezes" in the crystal when the crystal is cooled (i.e., there is a memory effect). A sufficiently intense light beam can drive the director in the crystal into stochastic motion. The original orientation of the director is disrupted in a region whose dimensions are considerably greater than the beam diameter.

The interactions of laser light with liquid crystals unquestionably represent a promising direction for future research on the physics of liquid crystals.[1-3] Several interesting effects have already been detected: the light-induced Fréedericksz transition, the aberrational self-focusing which accompanies this transition in a nematic liquid crystal,[4] self-diffraction,[5] light-induced square periodic structures in a cholesteric liquid crystal,[6] instabilities of various types, etc.[2,7] Of particular interest in the nonlinear optics of liquid crystals are effects which occur near phase transitions, where the crystals are particularly susceptible to external agents.[8-11]

Among the first studies in this field was a study[12] of the scattering of light by "holes" produced by a laser beam in a nematic liquid crystal, in particular, the nematic liquid crystal OCBP (octyl cyanobiphenyl) near the nematic-(isotropic liquid) phase transition. The OCBP crystal also exhibits a smectic-nematic phase transition.

In this paper we are reporting a study of the interaction of a laser beam with OCBP near this phase transition.

EXPERIMENTAL CONDITIONS AND RESULTS

We studied a homeotropically oriented OCBP crystal with a thickness $L = 150\ \mu$m. The crystalline, smectic, nematic, and isotropic phases of this material exist in the following temperature ranges:

$$t_{cr} \leqslant 21\ ^\circ\text{C} < t_{sm} < 32.5\ ^\circ\text{C} < t_n < 40\ ^\circ\text{C} < t_{iso}.$$

The cell with the liquid crystal was held in a constant-temperature chamber. The light beam from a cw argon-krypton ion laser (ILM-120) or a single-frequency argon laser (ILA-120, with a Fabry–Perot etalon) was focused by an objective with a focal length $f = 170$ or 330 mm into the cell. A double Fresnel rhombus was placed in the beam path in front of the crystal in order to rotate the polarization plane of the incident light. A film analyzer was positioned behind the crystal. On a screen behind this analyzer, ≈ 2 m away from the crystal, we photographed the observed pattern. A Polam L-213 polarizing microscope was used to study the changes which occurred in the crystal during exposure to the laser beam.

The experiments were carried out at a temperature $t \simeq t_{sm\text{-}n}$, where $t_{sm\text{-}n}$ is the temperature of the smectic-nematic phase transition, when the crystal was illuminated with light with a wavelength $\lambda = 5145$ Å. For illumination with a wavelength $\lambda = 5145$ Å, the experiments were instead carried out for a value of t 1.5–2 °C below $t_{sm\text{-}n}$. The radiant power incident on the crystal was less than 30 mW for $\lambda = 6471$ Å; it was 180–240 mW for $\lambda = 5145$ Å.

Let us summarize the results.

λ=6471 Å

1. Near $t_{sm\text{-}n}$, with the temperature regulator operating, and with crossed polarizers, a "cross" appears on the screen (Fig. 1). This cross lasts for 1–3 s or, in isolated cases, 5–10 s. If the temperature regulator is turned off when the cross appears, the cross disappears in 1–3 s, but it reappears 2–3 min later and then persists for a long time: tens of minutes or even hours. The cross is said to be "frozen" in this case.

2. When the polarization plane of the incident light is rotated, the frozen cross rotates through the corresponding angle.

3. A study of this frozen cross revealed that a synchronized rotation of the polarizer and the analyzer results in a rotation of the cross through the corresponding angle. The brightness of the cross increases with increasing power of the light incident on the crystal.

4. When the light is incident obliquely on the crystal (if the angle between the wave vector and the director is $\alpha \leqslant 20°$), we again observe a frozen cross, but it is now asym-

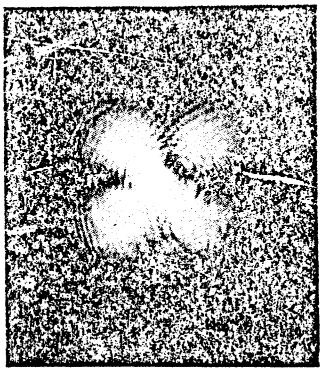

FIG. 1. Diffraction pattern (a cross) in crossed polarizers obtained as the result of the application of red light ($\lambda = 6471$ Å) to the liquid crystal.

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metric. This asymmetry becomes more obvious as α is increased.

5. If the crystal is instead rotated through an angle after the cross has already been frozen, then at small values $\alpha < 3°$ we still see a good cross, while for $\alpha > 3°$ the pattern does not look much like a cross.

6. Illumination of the crystal with circularly polarized light with $\lambda = 6471$ Å results in the formation of the same frozen structure as is produced by linearly polarized light. This conclusion follows from the observation that, after we remove the $\lambda/4$ plate in crossed polarizers, we observe a clearly defined cross, just as in the case of linear polarization.

7. In the green light from the ILM-120 laser, we observe not a cross but something which is closer to a square, with a hint of a cross shape. If this frozen square is illuminated by light with $\lambda = 6471$ Å, we see a good red cross. In the green light from the single-frequency ILA-120 laser, we always observe a cross. The square, rather than a cross, which is observed in the green light from the ILM-120 is evidently associated with the mode structure of this laser. Most of the experiments with the green light were carried out with the single-frequency ILA-120 laser.

$\lambda = 5145$ Å

1. At a temperature $t \approx t_{\text{sm-n}}$ and a beam power $P < 40$ mW, a green cross can be observed only in regions of the crystal with very good orientation. (The quality of the orientation can be tested by examining the conoscopic pattern observed on a little screen placed near the crystal, as proposed in Ref. 13.)

2. At $P \sim 40$–50 mW, as the temperature t approaches $t_{\text{sm-n}}$ from below, the cross is observed at essentially all times. It exists for several seconds. After the cross disappears, and the temperature regulator is turned off, the cross reappears in 2–3 min and exists for several minutes.

3. At higher power levels of the green light, $130 < P < 180$ mW, when the temperature reaches $t \approx t_{\text{sm-n}}$ as the crystal is being heated, a Fréedericksz transition usually occurs, and a characteristic pattern of aberrational self-focusing, with a large number of rings ($N \geqslant 34$), appears. A green cross can be observed on occasion before this self-focusing pattern appears. However, in essentially all cases the cross appears when the crystal is cooled (after the temperature regulator is turned off) at the time at which the aberrational pattern "collapses." This instant is preceded by a gradual decrease in the angular dimensions of the pattern (a decrease in the number of rings). Figure 2a shows a typical pattern of the green cross. The lifetime of the cross in this case ranges from tens of seconds to 1–2 min.

4. The longest-lived green crosses are found at a crystal temperature 1.5–2 °C below $t_{\text{sm-n}}$ for $180 < P < 240$ mW (we did not carry out experiments with $P > 240$ mW). The lifetime of the crosses in this case is a matter of hours or even days (the observations were not pursued for more than 2–3 days).

5. At this temperature, which was maintained continuously by the temperature regulator, and for $P \geqslant 180$ mW, a Fréedericksz transition occurs, and an aberrational pattern appears. If, with the temperature regulator turned on, the laser power is reduced to 120 mW at the instant at which the aberrational pattern has formed completely [$N(t)$ = const], and if the temperature regulator is then turned off, the nonlinear scattering begins to slowly decrease, and the process terminates in the appearance of a cross. This pattern can be observed repeatedly if P is reduced as soon as $N(t)$ reaches a constant value. If the beam power $P = 180$ mW is maintained for 3–4 sec at $N(t)$ = const, on the other hand, a decrease in P does not result in the appearance of a cross. If P is not reduced at all, there is a high probability for the onset of self-excited oscillations (discussed below), which still terminate in the appearance of a long-lived cross.

6. The crosses are usually encircled by arcs (Fig. 2b). The number of arcs is usually only one to three, but it can be larger. In the absence of the analyzer we observe a bright spot surrounded by half-rings in this case. Figure 2b shows the right side of the pattern observed in the absence of the analyzer. When the light incident on the crystal is polarized horizontally, these half-rings do not close into a ring along the vertical line. In the case of vertical polarization, they do not close along a horizontal line. The maximum number of half-rings observed in our experiments was nine. Figure 3a shows the pattern observed in crossed polarizers under a microscope in this case. We see a ring with a dark cross at its center. The outer diameter of the ring is 52–58 μm, and the inner diameter is 35–45 μm (depending on how sharply the laser beam is focused on the crystal). Figure 3b shows the

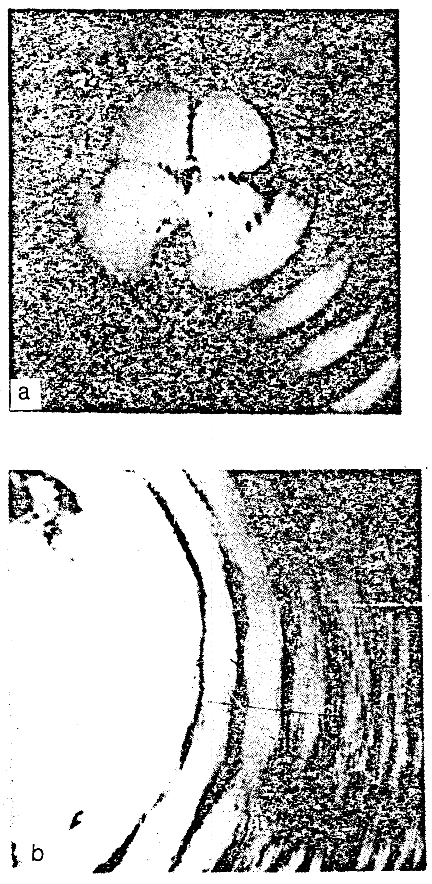

FIG. 2. Diffraction patterns (a) in crossed polarizers and (b) without the analyzer resulting from the application of green light ($\lambda = 5145$ Å) to the liquid crystal.

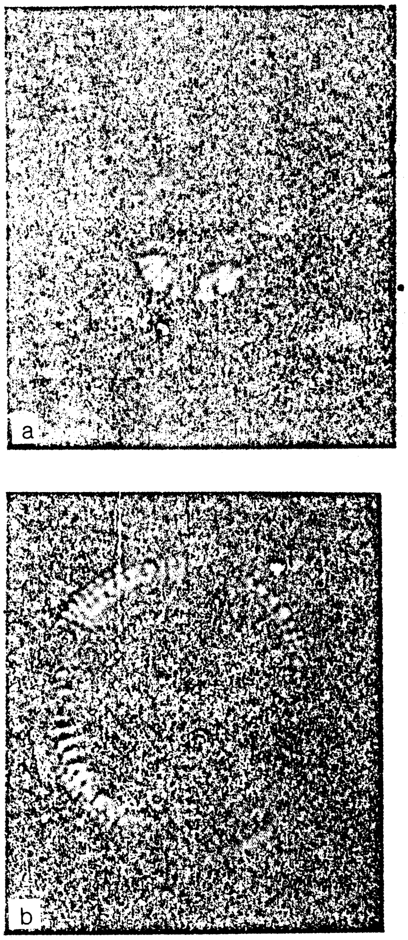

FIG. 3. Photographs (in crossed polarizers) of the distortions which are produced in the liquid crystal by laser light with $\lambda = 5145$ Å. a—The focal length of the lens is $f = 170$ mm; b—$f = 330$ mm.

ring which is found with a longer-focal-length lens ($f = 330$ mm). The outer diameter of this ring is 140 μm, and its thickness is 6 μm.

7. At a temperature 2 °C below $t_{\text{sm-n}}$ we frequently observe yet another interesting effect. The conditions required for the occurrence of this effect are not totally clear, but it usually occurs under the conditions described in paragraph 5. It consists of a pronounced instability of the aberrational pattern which arises upon the Fréedericksz transition (the rings collapse sharply and rapidly, reappear, and collapse again; this periodic process continues for several minutes). Random oscillations in the aberrational pattern can arise. In the process, there may be sharp changes in the shape of the rings. These random oscillations usually cannot be reduced in size or eliminated either by raising the cell temperature 0.5–1 °C or by changing the beam power to 120 mW, or even by blocking the beam for a fairly long time (1–10 min). These oscillations may persist for tens of minutes. There may also be some quieter oscillations in the aberrational pattern, with a periodic change of two to five aberration rings in the scattering. They sometimes appear when an attempt is made to produce a frozen cross in a part of the crystal in which a cross has existed previously and which persisted for several hours, i.e., in a part of the crystal in which there were, or are, defects in the homeotropic structure. In order to stop these self-excited oscillations, it is necessary to reduce the beam power sharply (to 40–50 mW).

We turn now to the picture seen under a microscope in the case of the self-excited oscillations (Fig. 4).

If the self-excited oscillations are quiet, we observe through the microscope—in addition to the ring described in paragraph 6, or a dark spot which sometimes appears in its place—either one ring (with an outer diameter of 230–250 μm) or two rings (with outer diameters of 80–100 μm and 170–200 μm) of a small-scale lattice (the length scale of this lattice is 5–6 μm).

When the self-excited oscillations are instead stochastic, the scattering varies in time, and in addition the aberration rings repeatedly break up and are replaced on the screen by some rapidly flickering bright spots. The pattern observed under the microscope after the end of these oscillations usually consists of two rings. In the ring adjacent to the

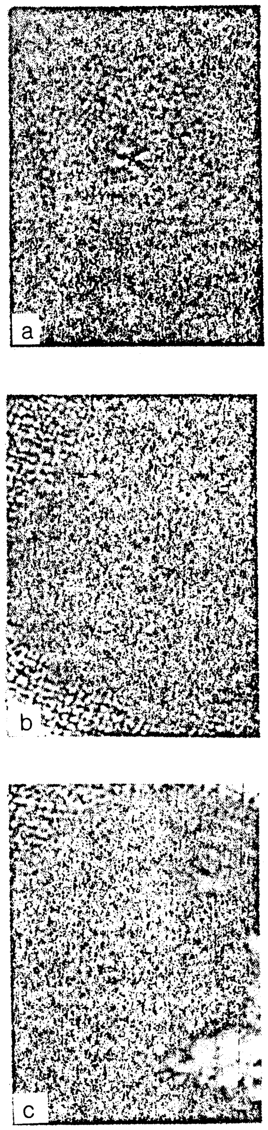

FIG. 4. Photographs of the distortions in the crystal corresponding to the onset of self-excited oscillations.

dark spot at the center of the pattern we observe no structure. The outer ring, in contrast, is a lattice (with a length scale of 5–6 μm). The diameters of the spot and the rings can vary over broad ranges: The diameters of the dark spot is 50–250 μm, the outer diameter of the inner ring is 250–550 μm, and the outer diameter of the outer ring is 800–900 μm.

DISCUSSION OF RESULTS

1. These experimental results indicate that an irregularity in the orientation of the crystal arises near the light beam near the smectic-nematic phase transition. This irregularity freezes in the liquid crystal when the latter is cooled.

The process by which this irregularity forms is apparently as follows. The absorption of light in the beam zone causes additional heating of the crystal and a transition of the crystal from the smectic phase to the nematic phase. This interpretation is indicated through the appearance of the characteristic intense scattering by fluctuations of the director. The absorption of green light by liquid crystals is known to be slightly stronger than the absorption of red light. Therefore, during illumination with green light at a sufficiently high laser power (at which the heating in the OCBP crystal is significant, despite the low absorption of blue-green light by this material[12]), the part of the crystal in the beam zone can undergo a phase transition at a crystal temperature (as set by the temperature regulator) lower than that corresponding to the phase transition, $t_{\text{sm-n}}$. The power of the red light in our experiments was much lower than the power of the green light, and the absorption was also weaker. The transition to the nematic phase thus occurred only at a crystal temperature close to $t_{\text{sm-n}}$.

The heating of the liquid crystal by the light beam is of course nonuniform: relatively great at the beam axis and relatively weak at the periphery. If, as the crystal is cooled, the temperature decreases more rapidly than the light-induced distortions relax in a region in which the crystal temperature is very close to the point of the phase transition (and in which the properties of the crystal have a substantial spatial nonuniformity), then the irregularity which arises in the crystal becomes frozen.

The picture observed during synchronized rotation of the polarizer and the analyzer implies that the irregularity has axial symmetry.

2. The light beam produces a spatial irregularity in optical properties and is diffracted by it. The freezing structure produces several orders of diffraction in the form of half-rings, which are observable without the analyzer, if the power of the green light is sufficiently high.

We can obviously use the diffraction pattern to estimate the size D of the irregularity responsible for the diffraction: $D=\lambda/\Delta\theta$, where $\Delta\theta$ is the angular distance between the diffraction orders. A calculation yields $D=50$ μm (for $f=170$ mm) or $D=100$ μm ($f=330$ mm). These results agree in order of magnitude with the waist size of the light beam focused by the lens.

3. The experimental results indicate that the irregularity which arises has a transverse anisotropy. The depolarization of the light, i.e., the appearance of crosses in crossed polarizers, may be caused by either a transverse anisotropy, in which case the position of the optical axis changes in the beam zone (there is a deformation of the layers of the crystal), or a depolarization of the diffracted rays which form the conoscopic pattern (or which are deflected as a result of nonlinear refraction). However, the experimental results show that the angular distance between the diffraction orders is much smaller than that between the rings of the conoscopic pattern. Furthermore, a cross should be observed in parallel polarizers for the conoscopic pattern, but in fact it is not.

4. If there is a radially symmetric transverse optical anisotropy, the optical axis at each point in the perturbed part of the crystal should lie in the same plane as the beam axis and should make an angle with the beam axis which depends only on the distance ρ from the axis. In this case, an extraordinary wave can propagate through the crystal (the polarization of this wave is directed along the perpendicular to the beam axis and lies in the plane in question), as can an ordinary wave (with a polarization directed along the perpendicular to the polarization of the extraordinary wave).

Let us attempt to calculate the light field on the screen. For this purpose we introduce a Cartesian coordinate system xyz, whose x axis runs parallel to the linear polarization of the incident beam, and whose y axis is parallel to the beam axis. At an arbitrary point in the crystal, characterized by the cylindrical coordinates ρ and β in the xz plane, the light field incident on the crystal, $\mathbf{E}(\rho,\beta,y)$, can conveniently be expressed as the superposition of an extraordinary wave and an ordinary wave, with the amplitudes

$$E_e(\rho,\beta,0)=A(\rho)\cos\beta,\quad E_o(\rho,\beta,0)=A(\rho)\sin\beta, \qquad (1)$$

where $A(\rho)=A_0\exp(-\rho^2/w^2)$, and w is the waist size of a Gaussian beam. At the exit from the crystal ($y=L$), the components of the light field are evidently

$$E_x(\rho,\beta,L)=A(\rho)\{\exp(iS_o)+[\exp(iS_e)-\exp(iS_o)]\cos^2\beta\},$$

$$E_z(\rho,\beta,L)=A(\rho)[\exp(iS_e)-\exp(iS_o)]\sin\beta\cos\beta, \qquad (2)$$

where $S_e(\rho)$ and $S_o(\rho)$ are the phase shifts for the extraordinary and ordinary waves. The light field in the Fraunhofer diffraction zone (on the screen) is described by the Kirchhoff formula

$$\mathbf{E}(R,\gamma,y)=\frac{k}{2\pi iy}\exp\left[ik\left(y+\frac{x^2+z^2}{2y}\right)\right]\int_0^\infty \rho\,d\rho\int_0^{2\pi}d\beta$$

$$\times\mathbf{E}(\rho,\beta,L)\exp\left[\frac{ikR\rho}{y}\cos(\beta-\gamma)\right]. \qquad (3)$$

Here $k=2\pi/\lambda$ is the wave vector, and R and γ are the polar coordinates in the plane of the screen (the angle γ is reckoned from the polarization direction of the incident light). Substituting (2) into (3), and introducing the diffraction angle $\theta=R/y$, we find

$$E_z=-C\sin 2\gamma\int_0^\infty \rho J_2(k\theta\rho)A(\rho)[\exp(iS_e)-\exp(iS_o)]\,d\rho, \qquad (4a)$$

$$E_x=E_{x1}+E_{x2}, \qquad (4b)$$

where

$$E_{x1}=C\int_0^\infty 2\rho A(\rho)J_0(k\theta\rho)\exp(iS_0)\,d\rho,$$

$$E_{x2}=C\int_0^\infty \rho A(\rho)[\exp(iS_e)-\exp(iS_0)][J_0(k\theta\rho)-J_2(k\theta\rho)\cos 2\gamma]\,d\rho,$$

$$C=\frac{k}{2iy}\exp\left[ik\left(y+\frac{x^2+z^2}{2y}\right)\right], \qquad (4c)$$

and F_n are Bessel functions.

The intensity of the diffracted light in crossed polarizers is $I\sim E_z^2\propto\sin^2 2\gamma$. This angular distribution corresponds to the observed cross. The light fields in parallel polarizers and also in the absence of an analyzer have complex angular distributions.

5. However, the following (extremely simplified) model leads to results which agree satisfactorily with the experimental data.

It follows from the experiments that the transverse anisotropy arises in the crystal primarily in a narrow spatial region between two cylinders, whose axes runs parallel to the beam axis and whose radii are ρ_1 and ρ_2 (Fig. 3a). If we assume that the anisotropy is of constant magnitude in the region $\rho_1<\rho<\rho_2$ (between the cylinders), then the integrals in (4a) and (4b) become

$$E_z=-C'J_2(k\theta\bar\rho)\sin 2\gamma, \qquad (5a)$$

$$E_{x2}=C'[J_0(k\theta\bar\rho)-J_2(k\theta\bar\rho)\cos 2\gamma], \qquad (5b)$$

where

$$C'=C[\exp(iS_e)-\exp(iS_o)]\bar\rho A(\bar\rho),\quad \bar\rho=(\rho_1+\rho_2)/2.$$

[These relations are valid for diffraction angles $\theta<1/k(\rho_2-\rho_1)$.]

The total intensity of the diffractive light is given by

$$I=|E_x|^2+|E_z|^2. \qquad (6)$$

We make the further assumption that the field E_{x1} does not play any significant role in the diffractive scattering at angles well above the diffraction angle of the unperturbed Gaussian beam, $\theta_{\rm diff}=\lambda/\pi w$. We can make this assumption because the refractive index for the ordinary wave does not depend on the orientation of the optical axis, and its temperature dependence is far weaker than that of the refractive index for the extraordinary wave. The spatial variation of the function $S_o(\rho)$ in this case is of course much slower than that of the function $S_e(\rho)$. As a result, the expression for the total intensity of the diffracted light for $\theta_{\rm diff}<\theta<1/k(\rho_2-\rho_2)$ is

$$I=|C'|^2[J_0^2(k\theta\bar\rho)+J_2^2(k\theta\bar\rho)-2J_0(k\theta\bar\rho)J_2(k\theta\bar\rho)\cos 2\gamma]. \qquad (7)$$

Using the asymptotic expression

$$J_n(x)=(2/\pi x)^{1/2}\cos(x-\pi/4-n\pi/2),$$

for the Bessel functions, we finally find

$$I=(2|C'|^2/\pi ke\rho)(1+\cos 2\gamma)\cos^2(k\theta\bar\rho-\pi/4). \qquad (8)$$

Relation (8) shows that half-rings should be observed in the diffraction pattern without the analyzer; this is the result found experimentally.

6. With regard to the nature of the transverse anisotropy which arises in the zone of the light beam, we do not have a clear picture. It is, however, obvious that the transverse anisotropy is a consequence of a distortion of smectic layers at the boundary with the nematic liquid crystal (which arises in the beam zone).

7. The stochastic regimes in the behavior of the director are accompanied by a propagation of the effect of the light beam over a spatial region (Fig. 4) with dimensions considerably larger than the beam cross section, according to the observations with a microscope. The photographs in Fig. 4 correspond to observation of an unstable regime for 30–40 min, after which the laser was turned off. Throughout the observation time the temperature regulator was turned off, and the crystal was allowed to cool at its own rate. The temperature of the OCBP in the beam zone should in this case correspond to the smectic phase, and the elastic interaction between the illuminated region (where the light beam is continuing to have its effects) and the rest of the crystal does not come to a halt. It extends over a progressively larger region around the light beam.

We thus see that during the stochastic regimes in the behavior of the director the deformation of the smectic layers may be a result of the operation of elastic forces caused by a deformation of the director in the beam zone under the orienting influence of the field of the light wave.

The lattice observed under the microscope arises because of a wavelike distortion of the smectic layers.[14] Deformations of this sort are possible when a variety of agents operate on liquid crystals, as was shown in Refs. 15 and 16. In particular, such deformations are possible during heating, during cooling, and during the thermal effect of a laser beam. The period of the structure which arises was measured with the help of a microscope and found to be $\approx 5\,\mu$m. This figure agrees in order of magnitude with an estimate of the period based on the relation given in Ref. 14.

In summary, this study has established the following results.

1) A light beam gives rise to an optically nonuniform, anisotropic spatial region in a smectic liquid crystal which is near the point of a transition to the nematic phase.

2) This anisotropic region may persist in the crystal for a long time (there is a memory effect).

3) A sufficiently intense light beam can give rise to a stochastic motion of the director in the liquid crystal. In this case the original orientation of the director is disrupted in a region whose size is much larger than the diameter of the light beam.

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