Laser-induced hydrodynamic instabilities and optical bistability in a liquid crystal: Theory and experiment

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Light-induced hydrodynamic instabilities in a nematic liquid crystal have been studied experimentally and theoretically. They were excited by single-mode laser light. The excitation involves the appearance of convective flows and dynamic orientation structures, which give rise to self-induced diffraction and scattering of the light. The dynamics of these effects and the hysteresis seen in them (an optical bistability) result exclusively from internal feedback, which in turn arises because of an interaction and a competition among various nonlinearity mechanisms: thermal, hydrodynamic, and orientational. This occurs in a strongly absorbing nematic liquid crystal (with dichroism), in the absence of any auxiliary quasistatic fields or initial temperature gradients.

1. INTRODUCTION

The time-varying and stochastic processes and instabilities which arise because of wave propagation in a strongly nonlinear medium are presently attracting considerable interest. The very first papers on nonlinear optics contained several discussions of the problem of the instability of nonlinear wave interactions. Those studies, however, were based on a weak nonlinear response of the medium. In actuality, strong nonlinear effects arose in these cases because of a buildup of nonlinear properties transversely across the medium.

Media which exhibit a pronounced intrinsic nonlinearity, e.g., liquid crystals, are now used widely. Because of the pronounced anisotropy of the molecules and their collective behavior (orientation) under the influence of an external field, some qualitatively new effects (not seen in conventional Kerr liquids) are induced in a liquid crystal by an external field. These effects are characterized in particular by the nonlocal nature of the response of the inhomogeneous medium to an external perturbation.[1]

The most interesting of these effects involve optical bistability (or multistability) in wave processes with internal feedback. In particular, a pronounced orientational nonlinearity in a liquid crystal, involving a threshold, leads to photoinduced (nonthermal) structural phase transitions with hysteresis (as the control parameter of the problem is varied). In these transitions, the feedback occurs specifically because of the nonlocal nonlinear response of the medium to the laser light.[2] Fluctuations of molecules play a fundamental role in liquid-crystal systems. It is because of these fluctuations that orientational effects are possible, and there may exist singular (critical) points near which intense nonlinear scattering of light occurs.

On the other hand, effects which result from laser heating play an important role in the interactions of laser light with liquid crystals. These effects lead in particular to a dependence of the material parameters of the medium (and thus the parameters of the photoinduced effects) on the temperature (here we will not take up some "coarser" effects—local transitions of the liquid crystal to an isotropic phase by virtue of the laser heating—which can easily be identified experimentally; Ref. 3, for example).

A mixture of a nematic liquid crystal with a resonantly absorbing (dichroic) dye, which we are examining in the present study, is a convenient experimental model in which effects stemming from the nonuniformity of heating and convective motion may compete with orientational nonlinear effects. In this case we are actually dealing with a dynamic nonlinear system with a small number of degrees of freedom in which, at comparatively low intensities of the cw laser light, the wave process becomes stochastic.

The high efficiency of effects of this sort is completely obvious for liquid crystals.[1-5] Research on the hydrodynamics of liquid crystals dates back a fairly long way. In particular, Dubois–Violette[6] studied the hydrodynamic instability induced in a nematic liquid crystal by a temperature gradient which was uniform in the plane of the liquid crystal. Laser-induced hydrodynamics in isotropic media—liquids and gases—was discussed in Ref. 7, among other places. Still, the photoinduced hydrodynamics of liquid crystals remains a field which has received little study.

In the present paper we are reporting a further study of the photohydrodynamic instabilities which were observed in Ref. 8 and of the accompanying nonlinear-optical effects caused in nematic liquid crystals by laser light. These studies were carried out in an all-optical experiment (i.e., without any auxiliary quasistatic fields or initial temperature gradients in the sample) in a highly nonlinear system: a composite mixture of a nematic liquid crystal and a dichroic absorbing dye. An important point is that a single-mode (TEM_{00}-mode) laser beam was applied to a nematic liquid crystal in an initially uniform orientation.

2. PHYSICS OF THE PHOTOHYDRODYNAMIC INSTABILITY IN NEMATIC LIQUID CRYSTALS; NUMERICAL ESTIMATES

The physical basis for the effects which we are discussing here is the appearance of convective instabilities in a liquid at rest under conditions such that a temperature gradient is set up in the liquid. We are essentially dealing with the classic Rayleigh–Bénard problem of the stability of a liquid layer of thickness d between two horizontal planes, the upper of which is at a temperature (T_1) lower than that of the lower plane (T_2): $T_1 < T_2$ (Ref. 9). Heat evolution further

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redistributes thermal energy through diffusion and transport of heated matter as the result of thermal expansion.

It is thus easy to find some simple estimates of the threshold for the onset of the photohydrodynamic instabilities, by analogy with the case of ordinary (isotropic) liquids. These estimates, however, are based on the strong assumption that the vertical temperature gradient (along the z axis) plays a dominant role (this point is the subject of a special discussion below).

In this case, assuming for simplicity that the volume of the liquid layer heated by the laser beam is a vertical cylinder of height d and cross-sectional radius b, and assuming that the wall of this cylinder is heat-insulating, we find the following estimate of the critical Rayleigh number R_{cr}, which determines the threshold for the onset of convective flows:[9]

$$R_{cr} \equiv \frac{g\beta(\Delta_z t)_{cr} b^4}{d\gamma\chi_{\parallel,\perp}} = 68. \qquad (1)$$

Here b is an effective radius for the steady-state heated volume of the medium. This radius depends on the thermal conductivity of the medium (it is usually legitimate to assume $b \gg a$, where a is the radius of the laser beam), g is the acceleration due to gravity, $\beta = -\rho^{-1}\cdot(\partial\rho/\partial T)$ is the coefficient of thermal expansion of the liquid, ρ is the density of the liquid, $\gamma \equiv \eta/\rho$ is the kinematic viscosity (η is the dynamic viscosity), $\chi_{\parallel,\perp} \equiv \varkappa_{\parallel,\perp}/\rho c_p$ is the thermal diffusivity, $\varkappa_{\parallel,\perp}$ is the thermal conductivity, c_p is the specific heat, and $(\Delta_z T)_{cr} \equiv T_2 - T_1 > 0$ ($T(z) = T_2 - (\Delta_z T)z/d$). If we use the parameter values given in Ref. 6 for a nematic liquid crystal [$\beta \sim 10^{-3}$ K^{-1}, $\rho c_p \sim 1$ J/(cm^3·K), $\chi_{\parallel,\perp} \sim 10^{-4}$ cm^2/s, and $\eta \sim 1$ P], and if we assume $g \sim 10^3$ cm/s^2, $d \sim 10^{-2}$ cm, and $b \sim 10^{-1}$ cm, we find that condition (1) is satisfied at $(\Delta_z T)_{cr} \approx 1$ K.

An independent estimate of the initial temperature gradient (i.e., that which prevails before an equilibrium is reached) between thin layers (of thickness Δz) in the entrance and exit planes of the sample—the gradient which arises because of the absorption (the fastest process) of light, of intensity I—yields

$$\Delta_z T = \alpha_\perp I(\Delta z)^2[1 - \exp(-\alpha_\perp d)]/(\pi^2 \rho c_p \chi_\parallel), \qquad (2)$$

where $\alpha_{\parallel,\perp}$ are the absorption coefficients associated with the anisotropy (with the dichroism). In our case we have $\alpha_\perp \approx 35$ cm^{-1} (Sec. 4). This value of $\Delta_z T$ corresponds to the value $\Delta_z T = 1$ K found from (1) if we use $\Delta z \approx 5$ μm and $I \sim 2\cdot 10^2$ W/cm^2.

We thus reach the conclusion that the excitation of photohydrodynamic instabilities in an absorbing nematic liquid crystal can be extremely effective and can occur in the field of a comparatively low-power (cw) laser. However, there are several important features which distinguish our case, that of the excitation of photoinduced instabilities by a laser beam, from the classic Rayleigh–Bénard problem.

In the first place, in addition to the vertical temperature gradient $\Delta_z T$, which results from the propagation of the laser light through the medium and the associated exponential attenuation of the light intensity, $I(z) = I(z=0)\exp(-\alpha_\perp z)$, there is a horizontal gradient $\Delta_r T$, which is determined by the finite size (in the transverse direction) of the Gaussian laser beam $I(r) = I(r=0)\exp(-r^2/a^2)$, where a is the beam radius.

Second, because of the anisotropy of the medium, the laser light may give rise to strong photoinduced reorientation of the director of the nematic liquid crystal in the medium (this reorientation is nonuniform along z and r).[10] These reorientations lead in turn to temperature gradients (along z and r, respectively) in the sample, because of the positive dichroism of the absorption (cf. Ref. 11). Photoinduced orientational effects can serve as a trigger for photohydrodynamic instabilities, for example; they lead to relative heating of the central layers of the liquid (in the z direction), for which the reorientation is at its greatest in the sample[1] and thus for which the absorption is strongest. In other words, there is an increase in $\Delta_z T$. The efficiency of these nonlinear processes (in particular, the threshold for their occurrence, I_{thr}, for the case in which light is incident normally on the sample, i.e., at an angle of incidence $\vartheta = 0$; Fig. 1) depends strongly on the temperature (I_{thr} decreases as $T \to T^{**}$, where T^{**} is the temperature at which the nematic liquid crystal undergoes a transition to the isotropic phase[12]).

Third, for an oblique orientation of the sample (along x; $\vartheta \neq 0$), i.e., for special boundary conditions (cf. Ref. 9), there is an additional mass flux along the oblique plane (the component of the gravitational acceleration $g \sin\vartheta$ comes into play and contributes to the pressure in the medium). This flux is unstable with respect to the excitation of periodic structures of the form $\exp(iqx)$, where q is the corresponding reciprocal-lattice vector.[4,6,13] As a result, "roller" structures should be induced (with a length scale $\Lambda \sim \pi/q$), and there should be a diffractive spreading of the beam in the plane of incidence.

Fourth, the establishment of the temperature field in our system and thus the values of the parameters of the transient processes are determined by multiple feedback channels. In addition to the standard mechanism, which involves fluctuating hydrodynamic flows (which implement an exchange of energy between different layers of the medium), there are some additional feedback channels. The latter result from the interdependence of the light-induced orientational-hydrodynamic mechanisms and thermal mechanisms (the latter operating because of the dichroism of the absorption).

As a result, there may be hysteretic effects in the sys-

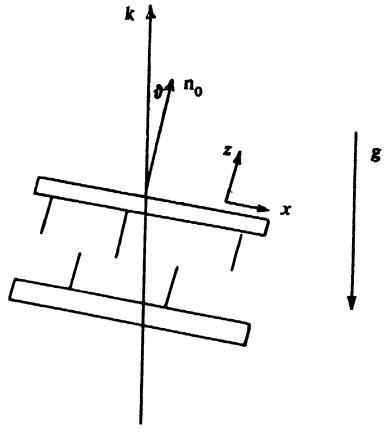

FIG. 1. Experimental geometry. $\mathbf{n}_0$—Director of the unperturbed nematic liquid crystal (which is along the normal, z, to the surfaces of the substrates); k—light wave vector; ϑ—angular deviation of the cell from a horizontal position g—acceleration due to gravity.

tem. An exact quantitative analysis of this problem thus requires the simultaneous solution of a three-dimensional heat-conduction problem with nonuniform distribution of the heat source (the laser beam) and the system of nonlinear nematodynamic equations in the case in which light has an orienting effect. It might seem that if we were to take account of all these factors (and also their relaxation times), which lead to a nonlinear dependence of the material parameters of the medium (the viscosity, etc.), we would be obliged to use self-similar approximations to solve the problem (cf. Refs. 13 and 14). Nevertheless, a rather simple theoretical analysis of the problem turns out to be possible. This analysis, which yields a qualitative description of the effect, is the subject of the following section of this paper (Sec. 3).

Before we take up that analysis, however, we wish to make two assertions of fundamental importance.

First, the transport of heated matter in a liquid crystal is known to be opposed by the orientational order of the nematic liquid crystal:[6] If the hydrodynamic flow (in the presence of a velocity gradient) tends to rotate the director (if there is a torque $\Gamma_{\text{hyd}}\gamma \propto (\partial \mathbf{v}/\partial \mathbf{r})$, where $\mathbf{v}$ is the flow velocity and $\mathbf{r}$ is the coordinate), then elastic forces create a torsional moment (which works opposite the hydrodynamic moment) because of the stiffness of the boundary conditions. The magnitude of this moment is $\Gamma_{\text{el}} \propto K\partial^2\Phi/\partial r^2$, where K is the elastic constant, and Φ is the angle through which the director is reoriented. It is the balance struck between these two moments which actually determines the threshold for the onset of photohydrodynamic instabilities.

Second, the presence of a transverse temperature gradient turns out to be extremely important for the occurrence of a hydrodynamic flow in a nematic liquid crystal ($\mathbf{v}\neq 0$); it actually determines the onset of the process. As in the laser excitation of threshold orientational structures in a nematic liquid crystal, forced convection in them is fluctuational in nature. It starts from thermal fluctuations of the director which exist in the medium. As was shown in Ref. 6, it is specifically because of these fluctuations that transverse temperature gradients arise (by virtue of the anisotropy of the thermal conductivity). As a result, these fluctuations are responsible for the transverse component v_x of the mass transport velocity in the liquid crystal. Hence hydrodynamic flows may be excited.

In our case, this temperature gradient $\Delta_r T$ exists at the onset and is determined by the geometry of the problem (the finite dimensions of the laser beam). It is this gradient (and not fluctuations of the director, as in Ref. 6) which determines the "start" of the process and which is responsible for the high efficiency of the process. The physics of the effect can be summarized as follows: The gradient $\Delta_r T$ creates a pressure drop and displaces a heated element of liquid into a less-heated region, in the peripheral part of the laser beam. Thereafter, the process reduces to the usual Rayleigh–Bénard mechanism.

The value of $\Delta_r T$ in the geometry of Fig. 1 can be found in the manner in which we found the estimate (2):

$$\Delta_r T = \alpha_\perp I(\Delta r)^2[1 - \exp(-r^2/a^2)]/(\pi^2 \rho c_p \chi_\perp)\,. \qquad (3)$$

For $\Delta r \sim 3\ \mu$m we have $\Delta_r T \sim 3$ K.

If we estimate $\Delta_r T$ from the fluctuation mechanisms (the case discussed in Ref. 6) we find values smaller by more than three orders of magnitude. Correspondingly, the efficiency of the heat-convection process is much lower (i.e., the conditions for the beginning of this process are more stringent). The situation can be illustrated in the following way.

It is easy to compare the excitation of photohydrodynamic instabilities and that of electrohydrodynamic instabilities in a nematic liquid crystal (the latter instabilities have been studied thoroughly). According to this analogy, when the role of the electric potential is played by the temperature[15] we find the following estimate (cf. Ref. 6) for the threshold temperature gradient $(\Delta_z T)_{\text{cr1,2}}$:

$$(\Delta_z T)_{\text{cr1,2}} \approx \left| \frac{K_{3,1}}{\rho g \beta} \frac{\eta_{5,4}}{\gamma_{3,2}} \frac{\varkappa_{\parallel,\perp}}{\varkappa_a} \frac{2\pi^4}{d^3} \right| . \qquad (4)$$

The anisotropy of the medium has been taken into account here (in the indices of the parameters); $\varkappa_a \equiv \varkappa_\parallel - \varkappa_\perp$ is the anisotropy of the thermal conductivity; and the two values $(\Delta_z T)_{\text{cr1,2}}$ correspond to the two original orientations of the director of the nematic liquid crystal in the sample, i.e., homeotropic (the molecules are oriented perpendicular to the plane of the substrates) and planar (along the substrates), respectively. We see from (4) that there is an inverse cubic dependence of $(\Delta_z T)_{\text{cr1,2}}$ (and thus of the threshold for the photohydrodynamic instabilities) on the sample thickness d. In this case we find that, with $d = 1$ mm, the threshold temperature difference between the substrates (for a homeotropic orientation of the liquid crystal) required for the occurrence of a convective instability is $(\Delta_z T)_{\text{cr}} = 5$ K. This result agrees with the experimental results of Ref. 6 and with the estimates in (2) and (3).

If we take that approach (which, in our case, essentially corresponds to the wide-beam approximation: $a \gg d$), however, we would need a huge temperature difference, $(\Delta_z T)_{\text{cr}} \sim 5000$ K, for the thickness of the liquid-crystal cell under consideration here, $d \sim 0.1$ mm. Such temperature differences are obviously unrealistic. On the other hand, we know that in practice laser-induced convection is distinguished by a particularly low threshold (Ref. 7, for example). This contradiction is resolved by the fact that in a laser experiment a dominant role is played by the transverse temperature gradient, which exists initially by virtue of the finite size of the beam, while the derivation of (4) was based on $\Delta_r T$ due to fluctuations of the director.

3. THEORY

In this section of the paper we restrict the discussion to a qualitative theoretical analysis, and we estimate the threshold for photohydrodynamic instabilities. Taking the approach of Ref. 6, which also assumes that the medium is weakly anisotropic (the single-constant approximation in terms of the elastic parameters of the medium), and ignoring the role of the vertical gradient in comparison with that of the horizontal (transverse) gradient, we find the following system of equations to describe the steady-state convective instability in a nematic liquid crystal caused by laser heating (the meaning of the parameters introduced here is defined below):

$$
\begin{aligned}
&-\frac{\partial}{\partial z}\delta P+\eta\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)v_z+\rho g\beta\delta T=0,\\
&-\frac{\partial}{\partial x}\delta P+\eta\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)v_x=0,\\
&-G_x v_x-\varkappa\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)\delta T-\varkappa_a G_x\frac{\partial}{\partial x}\delta\Phi=0,\\
&-K_F\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)\delta\Phi-\alpha_2\frac{\partial}{\partial z}v_x=0,\\
&\operatorname{div}\mathbf{v}=0,
\end{aligned}
\tag{5}
$$

Here K_F is the Frank elastic constant, and α_2 is the Leslie viscosity. As in Ref. 6, we are assuming that the problem is uniform along the y axis. We thus have only a transverse temperature gradient G_x, due to the laser heating of the medium to the threshold for mechanical transport of mass and heat:[9] $G_x=(\partial/\partial x)T(x)$, where $T(x)$ is found from the heat-conduction equation $\varkappa_\perp\Delta T+\alpha_{\parallel,\perp}I=0$, $\alpha_{\parallel,\perp}$ is the average light absorption coefficient, and Δ is the Laplacian. Again, we wish to stress that we are assuming $G_z\ll G_x$.

We seek elementary excitations (above the threshold for the convective instability) for the velocity, the pressure, the temperature, and the director reorientation angle (v, δP, δT, and $\delta\Phi$, respectively) in the form

$$
\begin{aligned}
\mathbf{v}&=\left(1,0,-\frac{q_x}{q_z}\right)\cdot v\exp(iq_z z+iq_x x),\\
\delta P&=P\exp(iq_z z+iq_x x),\\
\delta T&=T\exp(iq_z z+iq_x x),\\
\delta\Phi&=\Phi\exp(iq_z z+iq_x x)
\end{aligned}
\tag{6}
$$

Substituting into Eqs. (5), and carrying out some simple manipulations, we find an expression for the threshold value of G_x:

$$
G_x\approx\frac{1}{\delta}\frac{(1+\delta^2)^4}{(1+\delta^2)+\alpha_2\varkappa_a/K_F}\frac{q_z^4\varkappa\eta}{\rho g\beta},
\tag{7}
$$

where $\delta\equiv q_x/q_z\approx d/b$.

Let us examine the functional dependence $G_x(\delta)$ for the following values of the parameters: $d\approx 100$ μm ($q_z\approx\pi/d=3.14\cdot 10^2$ cm^{-1}), $g\approx 10$ cm$\cdot$s^{-2}, $\rho\approx 1$ g$\cdot$cm^{-3}, $\beta=10^{-3}$ K^{-1}, $\varkappa=5\cdot 10^{-4}$ cm$^2\cdot$s^{-1}, $\varkappa_a\approx 10^{-4}$ cm$^2\cdot$s^{-1}, $K_F\approx 5\cdot 10^{-7}$ dyn, and $\alpha_2\approx 0.8$ P. For water (for the essentially isotropic phase of the nematic liquid crystal), the value of η is[9] 0.01 P. For a nematic liquid crystal in the nematic phase at room temperature, we would have[15] ~ 1 P. Near the transition to the isotropic phase, which is the situation which usually prevails in experiments on nematic liquid crystals, we can assume $\eta\sim 0.1$ P. From (7) we then find

$$
G_x\approx 5\cdot 10^5\frac{1}{\delta}\frac{(1+\delta^2)^4}{(1+\delta^2)-160}.
\tag{8}
$$

Figure 2 shows a plot of $G_x(\delta)$. We see that the behavior of the threshold for the photohydrodynamic instabilities as a function of the geometric parameters of the problem (the cell thickness and the beam width) is generally not monotonic: After a sharp decay (in the region $\delta\lesssim 1$), $|G_x(\delta)|$ begins to increase slowly with increasing δ (cf. Ref. 14).

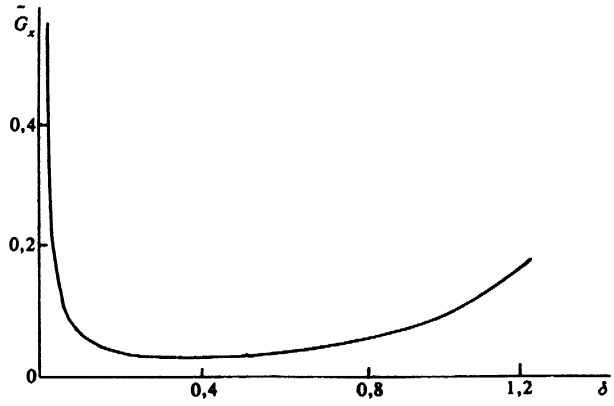

FIG. 2. Theoretical threshold temperature gradient G_x, normalized to $q_z^4\varkappa\eta/\rho g\beta$, i.e., $|\tilde{G}_x|\equiv G_x\rho g\beta/q_z^4\varkappa\eta$ (see the text proper regarding the notation) as a function of the parameter δ, which is the ratio of the sample thickness d to the effective width of the heated region, $b(\delta=d/b)$.

Since in our case, in contrast with Ref. 6, the threshold for the convective instabilities is exceeded by virtue of a previously specified transverse temperature gradient, rather than by fluctuations of the director, we can ignore those terms in Eqs. (5) which are responsible for fluctuations (i.e., which ignore the reorientation angle $\delta\Phi$). The minimum value of G_x is then found to be $G_x\sim 10^3$ K/cm (in the region $\delta\lesssim 1$). If we assume that the size of the heated region is $b\sim 2a\sim 0.01$ cm (this is fairly close to the actual values), we find $\Delta_r T\approx bG_x\approx 5$ K in agreement with the estimates in Sec. 2.

To determine the parameter b, we would obviously have to find a rigorous solution of the heat-conduction equation. However, since our calculations are only approximate, and they simply explain the experimental results reported in Sec. 4 below, we will content ourselves with a qualitative discussion. We believe that this discussion is completely satisfactory (we are actually suggesting that the parameter b be used as an adjustable parameter).

4. EXPERIMENT

Experimental conditions. A specially selected dichroic dye was added to a sample of a nematic liquid crystal (4-cyano-4-amylbiphenyl). As a result, there was a pronounced optical nonlinear of the entire complex, because of the absorption in the green part of the spectrum. An unsaturated composition was obtained at a dye concentration $<10\%$. It exhibited a positive absorption dichroism ($I_\parallel/I_\perp\sim 1{,}2$, where $I_{\parallel,\perp}$ are the intensities of the o- and e-polarized waves transmitted by the cell for a fixed input intensity). This composition leads to a good resonance (the absorption coefficients are $\alpha_\perp\approx 35$ cm^{-1} and $\alpha_\parallel\approx 50$ cm^{-1} at $T\approx 18$ °C) when a single-mode cw Ar^{3+} laser ($\lambda=0.51$ μm) is used as a pump.

The experimental geometry is shown in Fig. 1. The samples had a thickness $d=70$–220 μm. The molecules of the nematic liquid crystal in the mixture were homeotropically oriented [the director is in the orientation $\mathbf{n}_0\|z$; the plane of the substrates is the (x,y) plane]. Most of the experiments were carried out at room temperature. When the light was

incident normally ($\mathbf{k}\|\mathbf{n}_0$), the cell was positioned horizontally (the case of a vertical position will be pointed out separately). When the light was incident obliquely, the cell was rotated through an angle $\vartheta \leqslant 56°$, where $\mathbf{k}$ is the wave vector of the incident light. The diameter of the spot of the focused laser beam was $2a \approx 80\,\mu$m. In the course of the experiment, we checked to ensure that the liquid crystal did not undergo a transition to the isotropic phase (more on this below) as a result of the laser heating. We also made sure that the sample was not bleached because of a saturation of the absorption.

Let us summarize the experimental results.

Normal incidence ($\vartheta = 0$). As the intensity I of the light incident on the cell is raised smoothly, we see a threshold I_{thr1}, above which (at $I > I_{\text{thr1}}$) there is an avalanche increase (by about an order of magnitude) in the angular spectrum $\Delta\theta$ of the transmitted light. This effect is a consequence of the onset of photohydrodynamic instabilities in the medium. The angular spectrum in this case consists of concentric rings which are moving away from their center (the number of rings is a few tens; Fig. 3, d,e,f).

The time scale of the relaxation to an equilibrium pattern is $\tau_1 \sim 10$–15 s. This pattern is dynamic, however, as can be seen in temporal fluctuations and a strong scattering of the transmitted light. Just below the threshold ($I < I_{\text{thr1}}$) the laser beam broadens, and a single broad ring appears. We also see beats (in the diameter) of the ring against the background of a slight flickering of the scattered light, with time scales $\tau \sim 5$ s and $\tau \leqslant 0.1$ s, respectively.

A short distance above the threshold, we see, superimposed on the background of the main system of rings, another system (of two or three rings). This double pattern can be seen clearly by blocking the pump light. In this case the different systems of rings observed by means of a probe beam disappear with different relaxation times. If the temperature conditions are chosen appropriately, it becomes possible to observe the disappearance of one system of rings (that which relaxes more rapidly); then the other system of rings arises from the center of the beam and also undergoes relaxation, but far more slowly (over a time on the order of the relaxation time for orientational effects).

When the pump light is blocked, the fastest thermal mechanisms are turned off first, then the hydrodynamic mechanisms, and only then the orientational mechanisms. Because of the interplay among these factors, and because the decay times are approximately the same (for, say, the orientation and hydrodynamic mechanisms), the decay process (the collapse of the ring pattern) may be nonmonotonic at a local level. This is what is seen experimentally.

As the intensity is raised further (above the threshold, $I > I_{\text{thr1}}$), the system of rings begins to break up. Chaotic scattering becomes progressively more predominant (Fig. 3d). Even further above the threshold value of the intensity of the incident light, the angular spectrum of the transmitted (scattered) light becomes narrower, and it nearly reaches its original value (there is a local transition of the nematic liquid crystal into the isotropic phase). Later (for $I \gg 2I_{\text{thr1}}$),

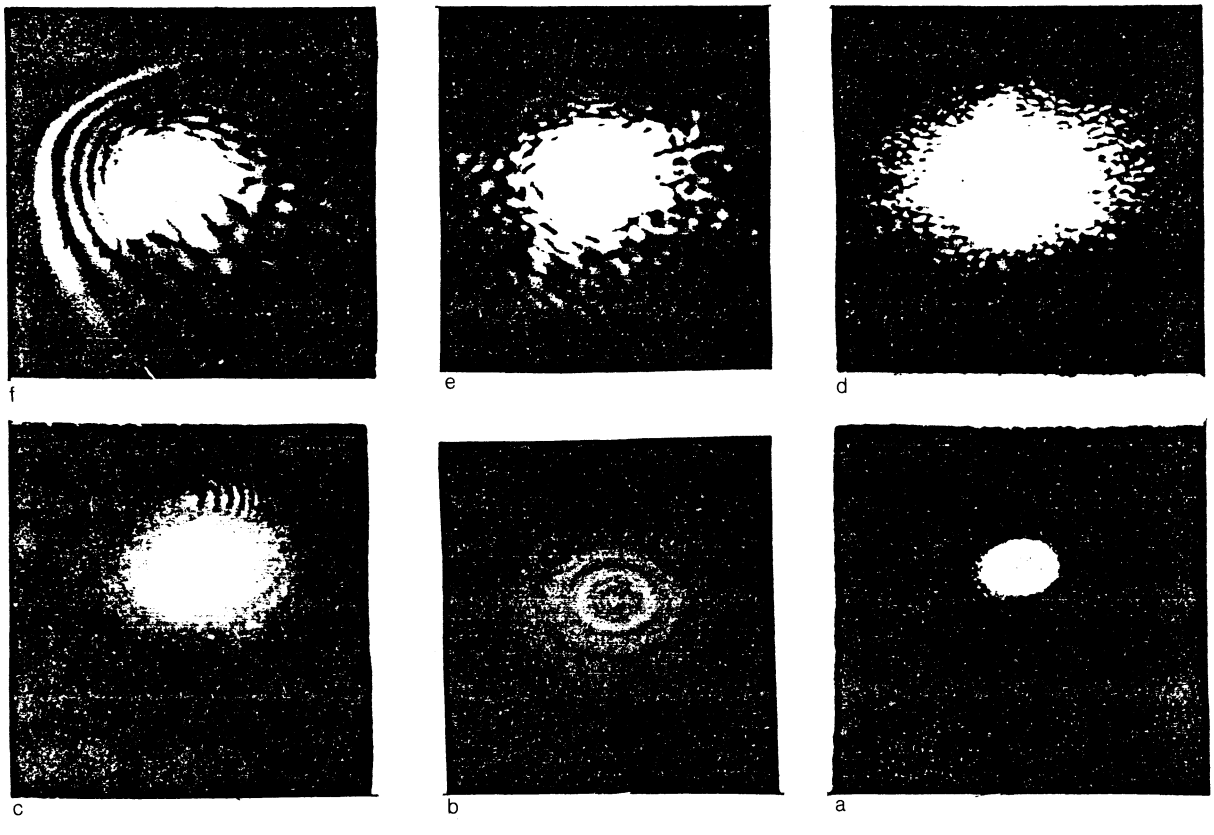

FIG. 3. Nature of the light transmitted through the sample in the far zone. a—Before the transition to the photohydrodynamic instability; b—ring structure immediately after the transition; c—breakup of the ring structure at higher intensities; d–f—formation of a grid pattern after the main beam is blocked (the photographs were taken at an angle $\sim 20°$ with respect to the screen).

the laser heating causes as thermal self-focusing of the beam in the isotropic phase.

When the changes in light intensity are repeated in the opposite direction (from the latter level), there is a temperature-induced transition back to the nematic phase. With a further decrease in I, the inverse transition to the original steady-state pattern in the transmitted light occurs at a different threshold intensity, $I_{thr,2} < I_{thr,1}$. The relaxation time (the time scale for the collapse of the ring structure in the transmitted light) is $\tau_2 \sim 30$–40 s, considerably shorter than the relaxation time.

As I is increased and reduced in succession, we thus see clearly defined hysteresis (an optical bistability). The width of the loop is $\Delta I \equiv I_{thr,1} - I_{thr,2}$. In addition, the value of $I_{thr,1}$ is itself a strong function of the temperature of the overall sample, T (Fig. 4).

For a pure nematic liquid crystal (without a dye), under the same conditions, the corresponding instabilities are not observed up to values $I > I_{thr} \approx 2I_{thr,1}$, at which the well-known photoinduced Fréedericksz transition occurs by an orientational mechanism (after the threshold is reached, in the case $\mathbf{E} \perp \mathbf{n}_0$).[16] For this transition, however, the ring structure, which determines the nonlinear phase shift for the transmitted light,[1] forms fairly smoothly[1,16] and is easily distinguishable from the pattern described above. We also note that hysteresis of the photoinduced Fréedericksz transition is not observed in this nematic liquid crystal (cf. Ref. 17).

When samples (of thickness 200–300 μm) with a saturated dye solution in a nematic liquid crystal are used (in this case, undissolved particles serve as tracer particles), the nature of the events which occur is qualitatively the same as above. In addition, the instabilities which arise in the laser-irradiated volume of the medium are clearly accompanied by hydrodynamic mass transport. In other words, we are actually talking about photohydrodynamic instabilities in this case. The time scales of these periodic fluxes (the angular rotation velocity) depend on I, decreasing with increasing I (the angular velocity increases in the process).

In a vertically oriented cell, the centers of hydrodynamic rotation of the material (the tracer particles) arise without any auxiliary conditions. In a horizontal cell, they can be observed after an addition spatial modulation of the light intensity over the beam cross section (a wire grid is placed in the beam).

Some special test experiments with a cell in a planar orientation ($\mathbf{n}_0 \| \mathbf{x}$) showed that the difference between the thresholds for the onset of the photohydrodynamic instabilities in purely *o*- and *e*-polarized waves is essentially equal to the value of ΔI for a homeotropic cell. The implication is that a dichronic absorption is playing an important role.

The effects of the photohydrodynamic instabilities described here depend rather weakly on the polarization of the incident light (the threshold for their excitation in normally incident light remains essentially the same when we switch from, for example, linearly polarized to circularly polarized light). In addition, the observed effects are essentially the same when the light is incident vertically from above and below the sample. On the other hand, they depend strongly on the size of the laser beam, the cell thickness, the rate at which I is switched, etc.

For example, the dependence of the threshold intensity I_{thr1} on the beam size (for certain values of the cell thickness) is as shown in Fig. 5. We see that for the parameter values used here this is a monotonically decreasing dependence (as the thickness of the sample is increased, the threshold intensity also decreases).

Oblique incidence ($\vartheta \leqslant 56°$). Although the basic features of the observed picture are generally the same (e.g., there is still a threshold involved), there are some qualitative distinctions.

First, in the field of an incident *e* wave, the excitation of photohydrodynamic instabilities is preceded by reorientation of the director of the nematic liquid crystal in a process which does not involve a threshold.[12,16] Interestingly, the system of diffraction rings associated with the photohydrodynamic instability is spatially shifted with respect to the orientational system of rings.

Second, after the threshold for photohydrodynamic instability is reached in this case, a special scattering direction arises, and the transmitted beam spreads out in the plane of incidence (some fairly stable scattering "whiskers," drifting slowly, are observed; Fig. 6). As the cell is rotated around the beam axis, these whiskers follow the rotation, with a time lag ~ 2.5 s. Measurements of the times involved showed that while the relaxation time of the orientational rings before the excitation of the photohydrodynamic instability is ~ 5 s, the relaxation time of the whiskers (after the onset of the photohydrodynamic instability) is ~ 2.5 s. The purely thermal lenses, with a far shorter relaxation time, ~ 0.1 s, have evi-

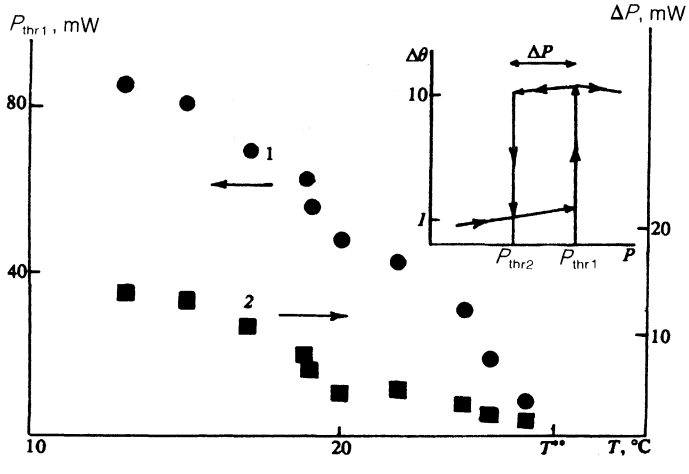

FIG. 4. Experimental results on the threshold power $P = I\pi\alpha^2$ for the onset of photohydrodynamic instabilities, P_{thr1} (*1*), and on the width of the hysteresis loop, $\Delta P \equiv P_{thr1} - P_{thr2}$ (*2*), versus the sample temperature T; Here T^{**} is the temperature at which the nematic liquid crystal undergoes a phase transition to an isotropic state. Inset: Hysteresis in the dependence of the angular spectrum of the transmitted light, $\Delta\theta$, on the power of the incident light, P (P_{thr2} is the intensity at which the photohydrodynamic instability "turns off").

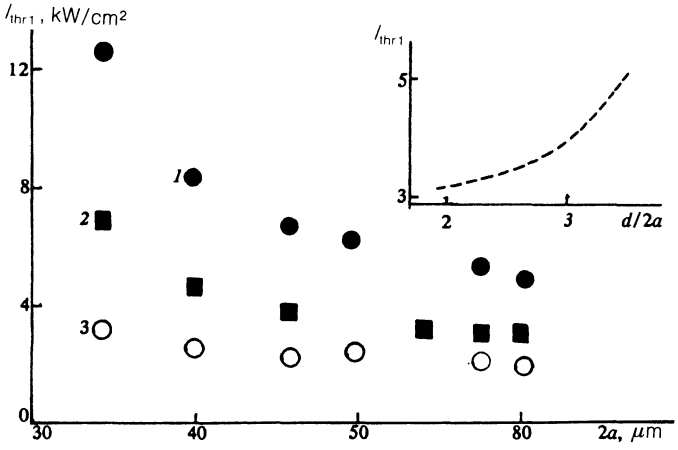

FIG. 5. Experimental threshold intensity I_{thr1} versus the beam diameter 2α for various sample thicknesses d: *1*—70 μm; *2*—130 μm; *3*—220 μm. Inset: Typical plot of I_{thr1} versus the parameter d, the ratio of the sample thickness d to the beam width 2α, for $d = 130$ μm (cf. the curve in Fig. 2 and $\delta > 1$).

dently been eliminated here.

As the main laser beam is turned on and blocked, the sequential changes (in several distinct steps) in the mode composition of the perturbations in the nematic liquid crystal cause corresponding (and also sequential) changes in the nature of the light scattering pattern (regardless of the orientation of the sample). This pattern is grainy. As the pattern is built up, the grains grow and convert into periodic lattices against the background of the ring structure (Fig. 3, b,c,a).

These spreading effects and scattering whiskers arise from a multiple diffraction of light by the lattice perturbations of the medium (cf. Ref. 12).

We thus have a fundamental result: In this special experimental geometry, it is possible to excite spatially periodic perturbations of a hydrodynamic nature in the medium by means of a single Gaussian beam.

5. DISCUSSION

It follows from the analysis of Secs. 2 and 3 that the physics of the observed effects described in Sec. 4 is indeed associated with the appearance, in an anisotropic medium (a nematic liquid crystal), of a convective instability of a liquid at rest as the result of light absorption under conditions such that a temperature gradient is set up in the medium.

Three experimental facts indicate that the anisotropy of the medium plays an important role in the onset of the photohydrodynamic instabilities in this case: (1) The observed picture of the photohydrodynamic instabilities is identical in the cases in which the light is incident on a horizontally oriented sample from above and from below. (2) This is not the case (at least at the given efficiency level) in the isotropic phase of the nematic liquid crystal. (3) The value of ΔI has been identified with $I^0_{thr} - I^e_{thr}$.

The two values $I_{thr1,2}$ can be written as a sequence of proportions (on the basis of the simple arguments in Sec. 2):

$$I_{thr1,2} \sim I^{o,e}_{thr} \sim (\Delta_z T)_{cr1,2}, \tag{9}$$

where we have estimate (4) for $(\Delta_z T)_{cr1,2}$.

Using (9) and (4) we then find

$$\frac{I_{thr1}}{I_{thr2}} \sim \frac{K_3}{K_1}\frac{\eta_5}{\eta_4}\frac{\gamma_2}{\gamma_3}\frac{\varkappa_\parallel}{\varkappa_\perp}\left(\frac{\alpha_\parallel}{\alpha_\perp}\right)^2.$$

However, as we mentioned back in Sec. 2, relations (9) agree

FIG. 6. Diffraction pattern in the case of an oblique orientation of the sample. Scattering "whiskers" can be seen along the direction of the inclination.

with the data in Fig. 4 [small values of $(\Delta_z T)_{cr}$] only when the governing role of $\Delta_r T$ in the excitation of the photohydrodynamic instabilities is taken into account. In our case, the matter is determined by the special geometry of the problem [again, compare this situation with that of Ref. 6, where the existence of a $\Delta_r T$, incorporated in the derivation of (4), stems from fluctuations of the director and thus requires huge values of $(\Delta_z T)_{cr}$ with this value of d]. Nevertheless, this approach does yield an explanation of the temperature dependence found experimentally (Fig. 4). Specifically, since we have $K \propto S^2(T)$, $\varkappa_a, \varkappa_\parallel, \eta, \gamma \propto S(T)$, and $\varkappa_\perp \propto 1/S(T)$, where $S(T)$ is the nematic order parameter $S(T) \propto (T^{**} - T)^{\varepsilon}$, $\varepsilon \sim 0.15$–0.3 (Ref. 6), we should have $I_{thr1,2}$, ΔI when the sample is heated ($T \rightarrow T^{**}$).

For τ_2 the dependence is again well known:[1] $1/\tau_2 \approx Kq^2/\gamma$. For the slowest mode ($\Lambda \sim d$), with $K \sim 4 \cdot 10^{-7}$ dyn and $\gamma \sim 1$ P, we find $\tau_2 \approx 25$ s, in agreement with the experimental results in Sec. 4.

A more accurate description of the behavior of the threshold for the photohydrodynamic instabilities, which is determined by the transverse temperature gradient G_x, is given by Eq. (8).

The experimental behavior shown in Fig. 5 (see also the inset in Fig. 5) actually corresponds to the case in which the geometric factor of the problem has a value $\delta > 1$. In this case, an increase in δ is accompanied by an increase in the threshold for the onset of the instabilities (if we use the approximation $G_x \propto I_{thr}$). A study of the region $\delta \ll 1$, in which there may be a deviation from monotonic behavior, requires a far higher light intensity (in agreement with Fig. 2). We did not study this region in detail (at these values of I, a transition to the isotropic phase occurs; the theory in Sec. 3 is no longer applicable).

At high intensities we again observe hysteresis effects and the appearance of a ring diffraction pattern. However, these effects are not due to hydrodynamic flows; they are determined instead by a local transition of the nematic liquid crystal to the isotropic phase and diffraction of light by the isotropic "holes" which form.[3] In addition, complex effects arise upon various conformational conversions of the molecules of the nematic liquid crystals in the region of strong absorption of the incident light.[18] These effects may also be caused by the admixture of dye molecules (cf. Ref. 19). Furthermore, these impurities may, in sufficiently high concentration, stimulate an independent phase transition in the system because of short-range-order effects[1] and thus lead to an avalanche behavior of the optical parameters of the problem near a singular point of this sort. However, these cases can be identified easily. In particular, the latter effect is not seen under our conditions (far from saturation of absorption).

6. CONCLUSION

Resolving all aspects of the problem discussed in this paper will of course require more thorough theoretical calculations (certain approaches are pointed out in Refs. 13 and 14, for example). There is also a need for further, detailed experiments. The results presented here should be regarded as merely a first step toward a study of photohydrodynamic instabilities. However, it can be concluded from even these results that some effects of fundamental physical importance are being manifested in phenomena associated with photohydrodynamic instabilities: on the one hand, the onset of stochastic behavior in a highly nonlinear system, and, on the other, the development of self-organization processes in the system.[5,13]

Some well-known experimental advances have been made in this direction. In particular, advances have been made during the excitation of transverse structures and space-time chaos under conditions such that an effective two-dimensional (external) feedback is realized in a nonlinear optical system (Ref. 20, for example). In our case the multiplex feedback, determined by the interplay and competition among hydrodynamic, orientational, and thermal processes, is an internal property of the system itself. It constitutes a separate problem, no less interesting, with many possibilities for the realization of various regimes.

This system is even more attractive because it can be prepared rather easily by simply adding dye molecules to a nematic liquid crystal. It was predicted in Ref. 11, on the basis of the specific features of the system consisting of a nematic liquid crystal and a dichroic dye, that there might be an absorption configurational bistability, regenerative pulsations, and effects due to heat evolution (as in our case). Accordingly, in this field we can expect to find yet other interesting effects, which will require experimental and theoretical study.

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