# Observation of light-induced hydrodynamic instabilities in a nematic liquid crystal in a single-mode laser beam

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Light-induced hydrodynamic instabilities have been discovered and studied in a nematic liquid crystal which is initially in a state of uniform orientation. The instabilities occur when a single-mode laser beam excites convective flows and dynamic orientation structures with a complex (and controllable) topology. These structures lead to a self-induced diffraction and to a scattering of light with an optical bistability. These effects stem exclusively from a multiple internal feedback which arises in a strongly absorbing nematic liquid crystal. These effects are observed in an all-optical experiment, without any additional quasistatic fields or initial temperature gradients.

**1.** A specially selected dichroic dye was added to a sample with a thickness $d \approx 120$ $\mu$m, in which the molecules of the nematic liquid crystal were in a homeotropic orientation (the director was in the orientation $\vec{n}_0 \| z$; the plane of the substrates was the $x,y$ plane). This dye imparts a pronounced optical nonlinearity to the material (the absorption coefficients are $\alpha_\perp \approx 35$ cm$^{-1}$ and $\alpha_\| \approx 50$ cm$^{-1}$ at $T \approx 17$ °C) when the crystal is pumped with a single-mode cw beam from an $Ar^{3+}$ laser ($\lambda = 0.51$ $\mu$m). An unsaturated composition is obtained at dye concentrations $\leqslant 10\%$. This composition exhibits a positive dichroism of the absorption ($I_\perp / I_\| \sim 1.2$, where $I_\perp$ and $I_\|$ are the intensities of the waves of the $o$ and $e$ polarizations transmitted through the cell), because of a "guest-host" effect.

The experimental geometry is shown in Fig. 1. For normal incidence of the light ($\vec{k} \| \vec{n}_0$), the cell was oriented horizontally. For oblique incidence, it was rotated through an angle $\alpha \equiv \vec{k}\hat{}\vec{n}_0$, where $\vec{k}$ is the wave vector. The diameter of the focused beam was $2a \approx 74$ $\mu$m. [At the intensities used in these experiments, laser heating did not cause a transition of the nematic liquid crystal to the isotropic phase (more on this below), nor did the sample undergo a bleaching as the result of a saturation of the absorption.]

**2.** Let us look at the basic results of these experiments.

*Normal incidence ($\alpha = 0$).* As the intensity of the light incident on the cell is raised smoothly, a threshold $I_{thr1}$ is reached such that at $I > I_{thr1}$ the angular width $\Delta\Theta$ of the transmitted light increases in an avalanche fashion (by about an order of magnitude). The light is in the form of concentric rings (dozens of them) moving outward from their center. An equilibrium pattern is established in $\tau_1 \sim 10$–$15$ s. This pattern is dynamic, however, as can be seen in the temporal fluctuations and the strong scattering of the transmitted light.[1)] The length scale of the variations decreases with increas-

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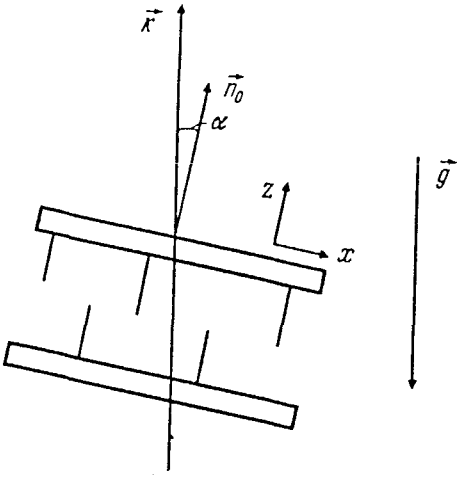


FIG. 1. Experimental geometry. $\vec{n}_0$—Director of the unperturbed nematic liquid crystal (this director is along the normal to the surfaces of the substrates, i.e., along the $z$ axis); $\vec{k}$—wave vector of the light; $\alpha$—angle through which the cell deviates from a horizontal orientation ($\alpha = \vec{n}_0\vec{k}$); $\vec{g}$—acceleration due to gravity.

ing $I$. The time scales of the fluctuations also decrease. At a level $I \sim 2I_{\text{thr1}}$, the nematic liquid crystal locally undergoes a transition to an isotropic phase. This effect can be identified easily (it can be seen under a microscope or in the angular distribution of the transmitted light). We carried out all our measurements before this transition occurred. Against the background of the main system of rings we observe another set of two or three rings. This double picture can be seen clearly when the pump light is blocked. The rings disappear with different relaxation times (in a probing beam).

As $I$ is lowered, the inverse transition to the original steady-state pattern occurs at a different intensity, $I_{\text{thr2}} < I_{\text{thr1}}$. The relaxation time is noticeably longer: $\tau_2 \sim 30$–$40$ s.

There is thus a clearly defined hysteresis as $I$ is sequentially raised and lowered (there is an optical bistability). The width of the hysteresis loop, $\Delta I \equiv I_{\text{thr1}} - I_{\text{thr2}}$, and the value of $I_{\text{thr1}}$ depend strongly on the overall sample temperature $T$ (Fig. 2).

These instabilities are not observed in a nematic liquid crystal without the dye, under otherwise the same conditions, at intensities up to levels $I > I_{\text{thr}} \approx 2I_{\text{thr}}$, at which we observe the familiar Fréedericksz transition (in the orientation $\vec{E} \perp \vec{n}_0$). The latter transition involves a threshold and occurs by an orientation mechanism.[5] For this transition, however, the ring structure, which determines the nonlinear phase shift for the transmitted light,[1] forms in a fairly smooth process and is easily distinguishable from the structure described above.

As the concentration of the dye dissolved in the liquid crystal is raised, and when saturation is reached (in this state, the undissolved particles serve as tracers), one can

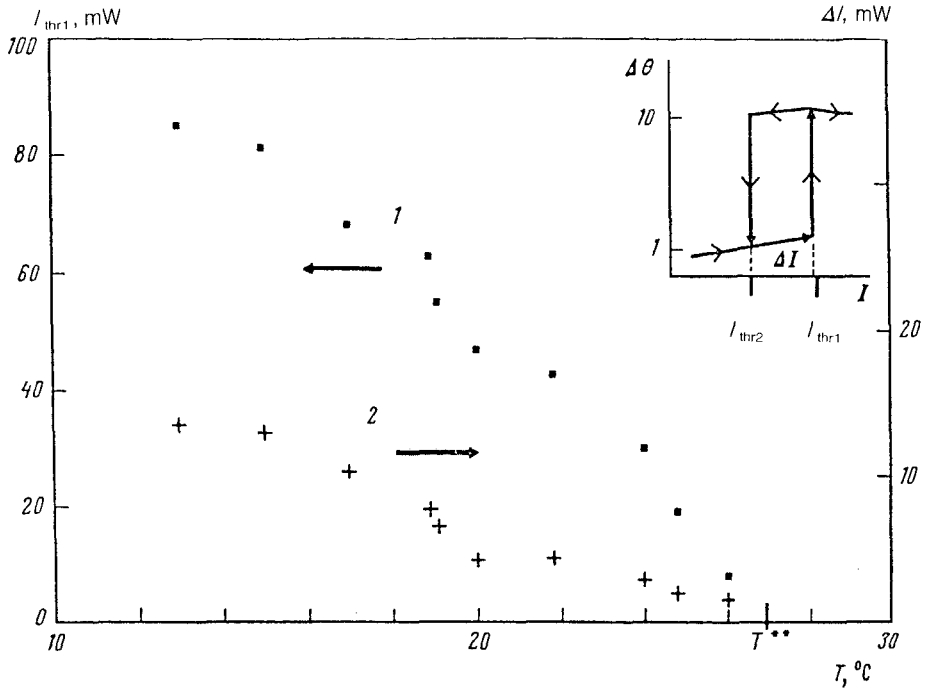


FIG. 2. The threshold intensity for the excitation of the light-induced hydrodynamic instability, $I_{thr1}$ (curve 1), and the width of the hysteresis loop, $\Delta I = I_{thr1} - I_{thr2}$ (curve 2), versus the sample temperature $T$, in Celsius degrees. Here $T^{**}$ is the temperature at which the nematic liquid crystal undergoes a phase transition to an isotropic state. The inset shows the hysteresis in the plot of the angular width of the transmitted light, $\Delta\Theta$, versus the intensity of the incident light, $I$($I_{thr2}$ is the intensity at which the light-induced hydrodynamic instability "turns off").

clearly see[2)] (either under a microscope or in the transmitted light) that the instabilities which arise are accompanied by a hydrodynamic transport of material. In other words, we are seeing light-induced hydrodynamic instabilities. The angular rotation velocity of these flows increases with increasing $I$.

*Oblique incidence ($\alpha \leqslant 56°$).* Although the picture which we observe in the case of oblique incidence is generally the same (in particular, a threshold is still involved), there are some distinctive features.

First, a reorientation of the director occurs (in a process which does not involve a threshold) in the field of an $e$-polarized incident wave, before the light-induced hydrodynamic instabilities are excited.[5,6] Interestingly, the system of rings associated with the light-induced hydrodynamic instability is seen on a screen to be spatially shifted away from the system of orientation rings.

Second, after the threshold for the light-induced hydrodynamic instabilities is reached, a special direction for scattering arises, in the plane of incidence (some fairly stable scattering "whiskers," drifting slowly, are observed).

Third (this result is of fundamental importance), some strictly periodic irregularities appear over the cross section of the beam in the medium. These irregularities can

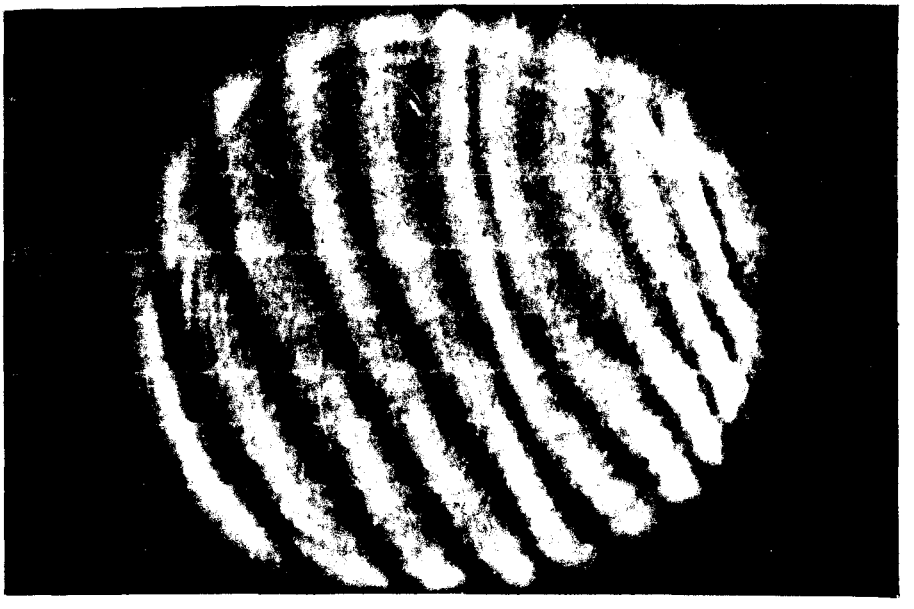


FIG. 3. Pattern of the periodic perturbations of the nematic liquid crystal over the cross section of the laser beam (the $x$ axis runs across the bands). This photograph was taken through a microscope at 800×. Also shown here is a schematic drawing of the corresponding structure of "roller" domains.

be seen clearly under a microscope [their period $\Lambda$ decreases with increasing $I$; at $I \approx 2 \times 10^3$ W/cm$^2$, for example, we have $\Lambda \sim 10\,\mu$m (Fig. 3)]. At sufficiently high $I$, a structure which is small in scale (grainy) but still fairly regular forms (there may be a superposition of several gratings of perturbations). The constituent modes of this structure exhibit a complex dynamics and compete with each other.

When the laser beam is blocked, there are sequential changes in the mode composition of the perturbations: The grains grow and convert smoothly into clearly defined periodic gratings with a varying wave vector $\vec{q}$ ($|\vec{q}| = \pi/\Lambda$). Before the overall transient (relaxing) picture disappears, we can make out a high-contrast grating which is the one of largest scale, with $\Lambda \sim d$. The spreading effects and the scattering whiskers described above arise from a multiple diffraction of light by these gratings (cf. Ref. 6).

These effects of the light-induced hydrodynamic instabilities depend rather weakly on the polarization of the incident light, but they depend noticeably on $a$, $d$, the rate at which $I$ is switched, etc.

Some special experiments with a cell in a planar orientation ($\vec{n}_0 \| x$) showed that the difference ($I^o_{\text{thr}} - I^e_{\text{thr}}$) between the thresholds for the excitation of the light-induced hydrodynamic instabilities in the fields of pure $o$ and $e$ waves is essentially the same as the value of $\Delta I$ for a homeotropic cell.

**3.** The physics of the effects observed here involves a convective instability of a liquid at rest in which a temperature gradient is set up. The case which we are discussing here has several features which distinguish it from the classical Rayleigh-Bénard problem[7]—of the stability of a liquid layer of thickness $d$ between two horizontal plates, the upper of which is held at a temperature $T_1$ lower than that of the lower plate, $T_2$.

First, in addition to the vertical temperature gradient $\Delta_z T$ which arises from the passage of the laser light through the medium [$I(z) = I(z=0) \cdot \exp(-\alpha_{\|,\perp} z)$] there is a horizontal gradient, $\Delta_r T$, which is determined by the Gaussian shape of the beam [$I(r) = I(r=0) \cdot \exp(-r^2)/(a^2)$].

Second, the light-induced reorientation effects which occur by the purely orientational mechanism [these effects are nonuniform along $z$ and $r$ (Ref. 8); cf. Ref. 9] lead in turn to temperature gradients (along $z$ and $r$, respectively) in the sample, because of the positive dichroism in the absorption. The effectiveness of these nonlinear processes depends strongly on $T$ (Ref. 6).[3)]

Third, when the sample is tilted (along $x$)—this orientation imposes some special boundary conditions in the problem (cf. Ref. 7)—an additional flow of material occurs along the inclined plane (the component $g \cdot \cos\alpha$ of the acceleration due to gravity is operating in this case). This flow is unstable with respect to the excitation of periodic "roller" structures of the $\exp(iqx)$ type.[2,4,7]

Fourth, the establishment of the temperature field in the system and, correspondingly, the characteristics of the transient processes are determined by a multiple internal feedback: In addition to the standard mechanism involving the operation of hydrodynamic flows (which bring about an exchange of energy between different layers of the medium), there are some additional, nonlinear mechanisms. The latter mechanisms cause a mutual coupling of the light-induced orientation-hydrodynamic and thermal effects (for example, they cause an increase in the heat evolution after the instability threshold is reached, because of the positive dichroism in the absorption). These effects are responsible for the hysteresis in the system.

Since the condition $a < d$ holds under these experimental conditions, the gradient $\Delta_r T$ plays an important role in the onset of the light-induced hydrodynamic instabilities. It creates a pressure drop and displaces a heated part of the liquid into a cooler region at the periphery of the laser beam (the process subsequently reduces to the ordinary Rayleigh–Bénard process). On the other hand, the light-induced orientational effects may serve as the trigger for the light-induced hydrodynamic instabilities. They cause a relative heating of the liquid layers which are the central layers along the $z$ direction. The reorientation is at a maximum for these layers.[6] The absorption is therefore also at a maximum (because of the positive dischroism); i.e., $\Delta_z T$ is intensified.

Three experimental facts indicate that anisotropy of the medium plays an impor-

tant role in the onset of the light-induced hydrodynamic instabilities: (1) The picture of these instabilities which is observed is identical in the cases in which the light is incident on the horizontally positioned sample from above and from below. (2) This is not true for the isotropic phase of this liquid crystal. (3) The values of $\Delta I$ are identified with $I^o_{\text{thr}} - I^e_{\text{thr}}$.

**4.** When all the factors mentioned above are taken into account—these factors lead to a nonlinear, time-varying dependence of the constitutive properties of the medium (the viscosity, etc.)—the problem can apparently be solved only by invoking some self-similar approximations (cf. Refs. 10 and 11). However, effects which are definitely observed involve an orientational order in the medium and a heat evolution, with a subsequent redistribution of thermal energy through diffusion and through transport of heated material as the result of thermal expansion.[7] The latter is known to be prevented by specifically the orientational order of a nematic liquid crystal:[4] If the hydrodynamic flow (in the presence of a gradient of the velocity $\vec{v}$) tends to rotate the director through an angle $\phi$ (the moment of force is $\Gamma_{hyd} \sim \gamma(\partial\vec{v}/\partial\vec{r})$, where $\gamma$ is the kinematic viscosity), then elastic forces create a torsional moment (which acts in the direction opposite the hydrodynamic moment) of magnitude $\Gamma_{el} \sim K(\partial^2\phi/\partial r^2)$, where $K$ is an elastic constant. The balance which is struck between these two moments determines the threshold for the light-induced hydrodynamic instabilities ($\phi \neq 0$; for periodic structures we would have $\phi \sim \cos \vec{q}\vec{r}$).

It is easy to find some simple estimates by assuming that $\Delta_z T$ plays a dominant role ($a > d$) and by ignoring the anisotropy of the medium. Assuming for simplicity that the volume of the heated layer of the liquid is a vertical cylinder of height $d$ with a cross-sectional radius $b$ and heat-insulating walls, we find the following expression for the critical Rayleigh number $R_{\text{cr}}$ in this case (this critical value determines the threshold for the excitation of convective flows):[7]

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

Here $b \gg a$ is some effective radius for the heated volume of the medium which is established as time elapses. This effective radius depends on the thermal conductivity of the medium. In addition, $\beta = -\rho^{-1}(\partial\rho/\partial T)$ is the coefficient of the thermal expansion of the liquid; $\rho$ is the density of the liquid; $\gamma \equiv \eta/\rho$; $\eta$ is the dynamic viscosity; $\chi_{\parallel,\perp} \equiv \kappa_{\parallel,\perp}/\rho c_p$ is the thermal diffusivity; $\kappa_{\parallel,\perp}$ are the thermal conductivities; $c_p$ is the specific heat; and $(\Delta_z T)_{\text{cr}} \equiv T_2 - T_1 > 0$ [$T(z) = T_2 - \Delta_z T z/d$]. Considering a nematic liquid crystal ($\beta \sim 10^{-3}$ deg$^{-1}$, $\rho c_p \sim 1$ J/cm$^3$·deg), $\chi_{\parallel,\perp} \sim 10^{-4}$ cm$^2$/s, and $\eta \sim 1$ P; Ref. 4), and assuming $g \sim 10^3$ cm/s$^2$, $d \sim 10^{-2}$ cm, and $b \sim 10^{-1}$ cm, we find that condition (1) holds with $\Delta_z T \approx 1$ deg. An independent estimate of the initial gradient $\Delta_z T$ between the thin layers (of thickness $\Delta z$) in the entrance and exit planes of the sample which arises from the absorption (the fastest process) of light of intensity $I$ leads to the value $\Delta_2 T \approx \alpha_\perp I \times \Delta z^2 [1 - \exp(-\alpha_\perp d)]/(\pi^2 \rho c_p \chi_\parallel)$. This value corresponds to[4] $\Delta_z T \approx 1$ °C for $I \sim 2 \times 10^2$ W/cm$^2$ and $\Delta z \approx 5$ $\mu$m.

It is interesting to compare two effects: the excitation of light-induced hydrodynamic instabilities and that of electrohydrodynamic instabilities (which have been studied thoroughly) in a nematic liquid crystal. Pursuing the analogy in which the role

of the electric potential is played by the temperature, we find the following estimates for $I_{\text{thr}1,2}$ (cf. Ref. 4):

$$I_{\text{thr}1,2} \sim I_{\text{thr}}^{o,e} \sim (\Delta_z T)_{\text{cr}1,2} \sim \left| \frac{K_{3,1}}{\rho g \beta} \frac{\eta_{5,4}}{\gamma_{3,2}} \frac{\kappa_{\parallel,\perp}}{\kappa_a} \frac{(2\pi)^4}{d^3} \right|. \quad (2)$$

Here we have incorporated the anisotropy of the medium (in the indexes on the parameters), and $\kappa_a = \kappa_\parallel - \kappa_\perp$.

Using (2), we find $I_{\text{thr}1}/I_{\text{thr}2} \sim (K_3/K_1)\cdot(\eta_5/\eta_4)\cdot(\gamma_2/\gamma_3)\cdot(\kappa_\parallel/\kappa_\perp)\cdot(\alpha_\parallel/\alpha_\perp)^2$. However, the only way to reconcile expression (2) with the data in Fig. 2 [small values of $(\Delta_z T)_{\text{cr}}$] is to take account of the governing role played by $\Delta_r T$ in the excitation of the light-induced hydrodynamic instabilities. In our problem, this gradient is determined by the special geometry [cf. Ref. 4, where $\Delta_r T$ was related to fluctuations of the director; huge values of $(\Delta_z T)_{\text{cr}}$ are thus required for our $d$].

Furthermore, since we have $K \propto S^2(T)$; $\kappa_a$, $\kappa_\parallel$, $\eta$, $\gamma \propto S(T)$; and $\kappa_\perp \propto 1/S(T)$, where $S(T)$ is the nematic order parameter [$S(T) \propto (T^{**} - T)^\epsilon$, where $\epsilon \approx 0.15$–$0.3$; Ref. 4], we should indeed find $I_{\text{thr}1,2}$, $\Delta I \to 0$ when the sample is heated ($T \to T^{**}$).

For $\tau_2$ the dependence is also well known:[1] $1/\tau_2 \approx Kq^2/\gamma$. For the slowest mode ($\Lambda \approx d$), with $K \approx 4 \times 10^{-7}$ dyn and $\gamma \approx 1$ P, we find $\tau_2 \approx 25$ s, in agreement with the experiments.[5)]

We wish to thank R. S. Akopyan for discussions and A. V. Sukhov for assistance.

[1)]Just before the threshold is reached, we see some slight flickering and beats in the light, with time scales $\tau \leqslant 0.1$ s and $\tau \sim 5$ s, respectively.

[2)]The picture is particularly clear when the sample is in the vertical orientation.

[3)]The effectiveness also increases sharply near other special points of the system (e.g., $I_{\text{thr}1}$), except $T^{**}$.

[4)]The same estimate in the case of the transverse gradient leads to a value $\Delta_r T \approx (\alpha_\perp/\rho c_p \chi_\perp)(\Delta r/\pi)^2 I[1 - \exp(-r^2/\alpha^2)] \sim 3$ °C with $\Delta r \approx 3$ $\mu$m.

[5)]When the pump light is blocked, the fastest thermal mechanisms are the first to drop out of the picture; then the hydrodynamic mechanisms drop out; and only then do the orientational mechanisms drop out. Because of the interplay among these factors, the decay (the collapse of the ring pattern) may be locally nonmonotonic, as is seen experimentally.

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