# Nonlinear Interaction of Light with Transversely Moving Medium

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Interaction of a light beam with matter reveals extreme sensitivity to transverse motions. Spatio-temporal delocalization of interaction and diffusive transport of light-induced perturbations is revealed through a new family of diffraction patterns. The response of the medium to the transverse motion of the light enhances near the second order phase transition point. The phenomenon has large application potential for the study of motions that are beyond the power of Doppler-effect-based methods, and suggests fundamental problems about critical processes with nonstationary influences. [S0031-9007(96)01407-X]

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Nonlinear interaction of liquid crystals (LC) with laser beams has become one of the most interesting parts of the nonlinear physics [1–5]; see also the monographs [6,7]. This interaction is distinguished by a number of unique quantitative and qualitative features.

Quantitatively, large reorientation of LC molecules ($\theta \sim 1$ rad) can be achieved in the beam of low-power lasers (P $\sim 10-100$ mW), and can have relaxation times of the order of seconds. Given high optical anisotropy of LC, such a reorientation of its optical axis leads to remarkable changes in the phase of the beam ($\delta\Phi \sim 100$ rad) which, therefore, can be easily measured. The most straightforward and most popular method of determination of the phase shift is the calculation of the number of the fringes of such an "intrinsic" interference, $N = \delta\Phi/2\pi$ [6,8]. This simplest method provides remarkable accuracy due to a typically large number of fringes. Essentially better accuracy of phase measurements is achieved by registration of the intensity on the axis of the outgoing beam [9,10].

Qualitatively, the state of LC is described by two angles determining the orientation of its optical axis (the director). These angles allow strong modulations in space giving rise to a large number of spatial modes of deformation. The character and strength of excitation of these modes depend essentially on the geometry of the experiment.

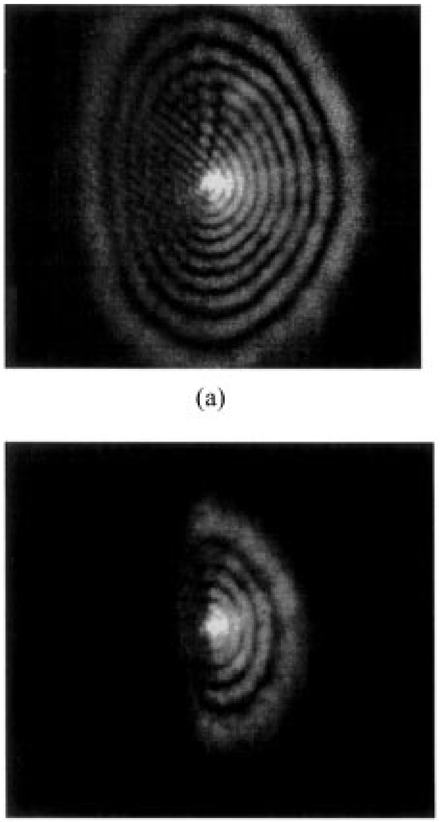
Such a multitude of control and behavior parameters, in addition to the strength of interaction, leads to the observation of many interesting nonlinear phenomena with small modifications of the experimental geometry [6]. Thus, for a homeotropically oriented nematic LC (NLC) (the molecules are perpendicular to the boundaries of the cell), the reorientation is proportional to the power density of the radiation for an obliquely incident beam of extraordinary polarization. At normal incidence of a linearly polarized beam, the reorientation acquires the features of a phase transition of a second order; it takes place above a threshold intensity of the radiation. With a normally incident elliptically polarized beam, the orientation of NLC reveals complex nonlinear dynamics like precession and nutation [3]. At small angles of incidence of an ordinary polarized beam, an intriguing unique scenario to chaotic oscillations of the director has been reported [5].

We will present here the results of experimental and theoretical investigations of a new physical situation which puts forward new fundamental problems, nonlinear interactions, and phase transitions in nonstationary complex media, and suggests new opportunities in laser velocimetry. We provide evidences that transverse motion of LC relative to the light beam modifies essentially their interaction and leads to new nonlinear phenomena: diffusive transport of light-induced perturbations, temporal instability, formation of a new family of self-induced diffraction patterns. Some simplest features of the observed phenomena are rather universal for all materials which exhibit nonlinear response to one or other influences (optical, acoustical, etc.).

In our experiments, the radiation of an $Ar^+$ laser (Stabilite 2017 of Spectra-Physics, the wavelength of the radiation $\lambda = 0.514$ $\mu$m, the beam radius $b = 0.75$ mm $1/e^2$ of intensity) is focused with a lens of focal length $f = 20$ cm upon a homeotropically oriented NLC-cell (E7 of BDH) of thickness $L = 50$ $\mu$m. The cell is positioned on a motorized moving stage. The speed of motion of the stage can be chosen in the range 0.5–40 $\mu$m/s with 0.1 $\mu$m/s accuracy. Both the speed of motion and the position of the cell are controlled by a computer. An image processing system was used to capture and analyze the far field profile of the beam.

The typical pattern of self-phase modulation consisting in a system of annular rings is formed in the far field zone when the stage is still. When the stage is brought into motion, the pattern is deformed, Fig. 1(a), and the number of rings decreases. During relaxation, half of the pattern on the opposite side of the motion may completely shrink leaving a weak flickering shadow of a system of spots,



(b)

FIG. 1. The patterns formed at the output beam for different states of motion: (a) deformation of rings for slow motions, (b) the arc pattern for faster motion. The arcs are directed towards the direction of motion of the NLC-cell with respect to the laser beam.

Fig. 1(b). Formation of these patterns is accompanied by huge changes in the divergence of the beam which may become of the order of 0.1 radians resulting in tens of cm linear sizes of the patterns at a distance from the NLC-cell of about 1 m.

With changing speed of motion $\Delta v$, the changes in the number of fringes $\Delta N$, as well as changes in their form and divergence angle, are so drastic that they can be used to characterize the NLC reorientation. Registration of the number of fringes $N$ is a sufficiently accurate method (10%) only for a large number of fringes ($N \sim 10$); however, being interested in qualitative characterization of the effects, we used $N$ in all experiments described in the present paper. By that, the strength of the influence of the speed on the interaction of light with NLC is characterized with $\Delta N / \Delta v$ for a fixed power level $P$.

The results of measurements in the nonthreshold geometry that is realized when the laser beam impinges upon the sample at an oblique angle and has extraordinary type of polarization are summarized in Figs. 2 and 3. The strongest dependence of $N$ on speed is achieved at approximately $v = 10$ $\mu$m/s. At that speed, $\Delta N / \Delta v \sim -0.2$ $(\mu\text{m/s})^{-1}$ for $P = 722$ mW (power density at the focus $I = 24$ kW/cm$^2$).

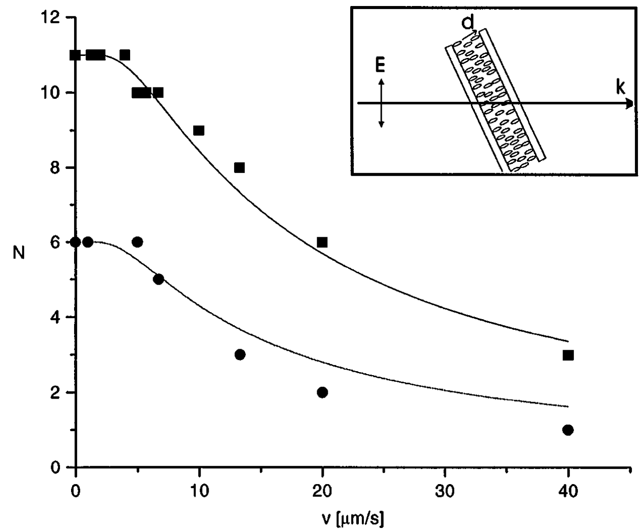



FIG. 2. The number of fringes $N$ of intrinsic interference vs the speed of transverse motion $v$ for different values of the light power $P$ in case of nonthreshold interaction of light with NLC (incidence angle of beam upon the NLC $\alpha = 25°$). Fit of experimental data with the aid of simplified theory for two values of radiation power $P = 722$ and 375 mW.

At a fixed speed, the number of fringes $N$ increases with power $P$, Fig. 3. The slope of the dependence of $N$ on $P$ decreases with increasing speed, and there appears a threshold power below which no pattern is being formed in the beam at the particular speed.

The effect of the transverse motion is even more peculiar in the geometry where the laser-induced reorientation of NLC shows critical behavior, presenting, actually, a second order phase transition; that is, the case of normal incidence of the linearly polarized light beam on NLC. Figure 4 shows the decrease in the number of fringes with increasing speed of motion at a fixed power. In this case,

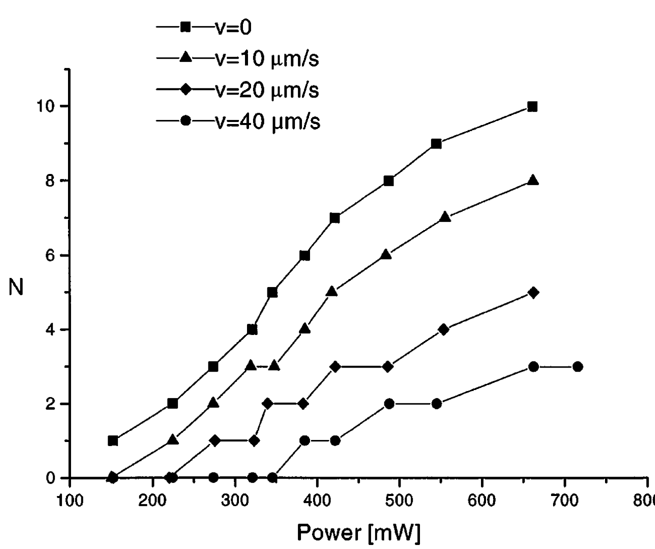



FIG. 3. Nonthreshold interaction of light with NLC (incidence angle $\alpha = 25°$): the number of fringes $N$ vs laser radiation power $P$ for different values of the speed of motion. The lines are guides for the eye.

the sensitivity to the motion, $\Delta N/\Delta v \sim 1\ (\mu\text{m/s})^{-1}$, turns to be remarkably stronger (about 5 times) than that in the case of noncritical (nonthreshold) interaction. The motion with a speed as small as 1 $\mu$m/s reduces the number of fringes by 1, and a speed of 8 $\mu$m/s suppresses 7 fringes obtained in still NLC for a given power level $P =$ 527 mW (17 kW/cm$^2$). The effect of motion is stronger near the critical power. However, only a small number of fringes is created near the threshold, and the method of counting fringes is not sufficiently accurate in that case. At a fixed speed, the threshold power giving rise to the fringe pattern increases with the speed of motion.

The measurement time in our experiments was typically about 10 s to ensure the establishment of the steady state when starting the motion or stopping it. At the nonthreshold geometry, the steady state was achieved in 6–7 s when stopping the motion, and was largely independent on the initial state. However, at the critical geometry of interaction, and in a particular range of the laser power and the transverse speed, the number of fringes underwent temporal oscillations and the steady state could not be reached during remarkably larger time periods. These oscillations were rather irregular, and will become a subject of future detailed studies. We could not attribute them to an inhomogeneity of the NLC-cell properties: the transverse shifts of the NLC-cell during the measurements were usually less than 0.4 mm, and the parameters of the NLC-cell were homogeneous in much larger scales (~5 mm) which could be checked by comparing the results obtained in different spots.

We are dealing with a highly complex phenomenon: critical behavior induced by moving perturbation in a medium with diffusive nonlinearity. Its theoretical description, especially in case of the critical effects, is related with remarkable difficulties. Let us consider the nonthreshold geometry of interaction within the framework of several standard assumptions such as description of the orientation state with the aid of only one angle $\theta$, strong anchoring at the cell boundaries ($z = 0$ and $z = L$), and a Gaussian incident beam. Assuming the beam is moving relative to the NLC along the $x$ axis with a speed $v$, the problem can be reduced to a solution of the following linearized variational equation for the reorientation angle $\theta(x, y, z) = \theta_m(x, y)\sin(\pi z/L)$:

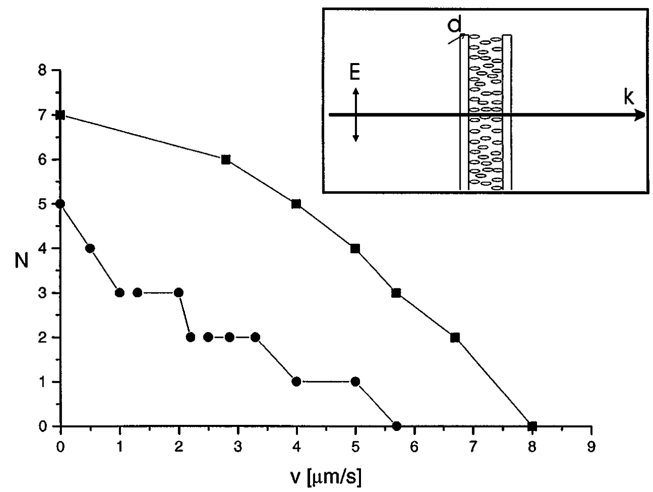



FIG. 4. The number of fringes $N$ vs the speed of transverse motion $v$ for the threshold interaction of light with NLC at different power levels. The lines are drawn as guides for the eye.

$$\frac{\partial\theta_m}{\partial t'} = \frac{\partial^2\theta_m}{\partial^2 x'} + \frac{\partial^2\theta_m}{\partial^2 y'} - \theta_m - \xi \exp\left[-2\frac{(x' - v't')^2 + y'^2}{(w')^2}\right]. \quad (1)$$

Here $x', y'$ are the Cartesian coordinates in the plane of the NLC-cell, and $w'$ is the beam waist radius, both normalized to $L/\pi$; $t' = t/\tau$ is the time in units of the characteristic relaxation time of orientation. The translation velocity is normalized to the "speed of relaxation" $v' = v\pi\tau/L$. The parameter $\xi$ is proportional to the intensity of incident radiation and depends on the geometry of interaction. Fourier transformations allow one to present the stationary solution of Eq. (1) ($t \gg \tau$) in the form

$$\theta_m(x, y, t) = -\xi\frac{(w')^2}{4\pi}\int\int\frac{\exp[-\frac{(w')^2}{4}(p^2 + q^2) - ip(x' - v't') - iqy]}{1 + p^2 + q^2 + ipv'}\,dp\,dq\,. \quad (2)$$

Equation (2) determines the reorientation magnitude of the director as a function of the speed of motion and the beam waist radius. Numerical study of Eq. (2) shows that the profile of reorientation is shifted in the maximum with respect to the beam axis ($x = vt, y = 0$) due to the motion. Treatment of the experimental results would consist in determination of this maximal reorientation as a function of the speed, beam waist size, power, and intensity. The obtained Eq. (2) allows numerical treatment only; we will present derivation of Eq. (2) and detailed study of its consequences elsewhere.

To gain more physical insight into the problem, we have to sacrifice with the accuracy and fullness of the description. Assume that the material experiences the influence of the moving laser beam as only a local change of radiation intensity in time. This corresponds to neglecting the effects of transverse distribution of the intensity, the spatial nonlocality of the material response, and diffusive transport of perturbations. In such an oversimplified model, the reorientation of NLC will effectively take place during the time when the beam is moved across the NLC-cell over a distance equal to its diameter $2w$: $T = 2w/v$. Let us describe the dynamics of the nonthreshold process with a simple exponential law of relaxation $\theta(t) = \theta_s[1 - \exp(-t/\tau)]$, where $\theta_s$ is the stationary value of the orientation achieved at $t \gg \tau$, and $\tau$ is the characteristic relaxation time. Thus, the magnitude of maximal reorientation of NLC which can be achieved in the moving beam can be evaluated as

$$\theta(t) = \theta_s\left[1 - \exp\left(-\frac{2w}{\tau v}\right)\right]. \quad (3)$$

With the aid of (3), one can evaluate $\Delta\theta/\Delta v \sim (\theta_s\tau/w)\eta^2 e^{-\eta}$ where $\eta = 2w/v\tau$. Thus, the largest response $\Delta\theta/\Delta v$ is achieved at the speed $v_0 = w/\tau$. In our experiment $v_0 = 15\ \mu$m/s since $w = 44\ \mu$m and $\tau = 6$ s. The minimal change in the speed which still can be sensed is $(\Delta v)_{\min} \sim (\Delta\theta_{\min}/\theta_s)v_0$ where $\Delta\theta_{\min}$ is the magnitude of the change in NLC orientation that can be registered with a particular method of measurement. One fringe accuracy ensures $\Delta\theta_{\min}/\theta_s = \Delta N/N_s = 0.1$ and, hence, $(\Delta v)_{\min} \sim 1\ \mu$m/s. The sensitivity to the motion when the initial stationary state is the state of rest is $v_{\min} = 2v_0/\ln(\Delta\theta/\theta_s) \sim v_0 \sim 10\ \mu$m/s in good accord with the measured values. The fit of experimental data with the expression (3) shows good coincidence, particularly for large power density and slow motions, Fig. 2.

Evaluation of the slope $\Delta N/\Delta v$ at the optimum speed $v_0$ gives $\Delta N/\Delta v = -0.5N_s\tau/w \sim -0.025N_s$, which is also in reasonable agreement with the experimentally obtained $\Delta N/\Delta v = -0.2\ (\mu\text{m/s})^{-1}$ for $N_s = 11$ ($P =$ 722 mW). These results reflect the circumstance that the smaller is the transverse size of the beam and the larger is the relaxation time of the medium, the stronger is the spatiotemporal decoupling of the beam and the medium for the given speed of translation.

For the geometry of the experiment when the reorientation takes place as a phase transition of the second order, the characteristic relaxation time is critically enhanced for the laser beam power density close to the threshold power density: $\tau_{\text{OFT}} = \tau/(P/P_F - 1)$. Therefore, in the light of the above discussion, the sensitivity of the interaction to the relative motion of the NLC with respect to the beam remarkably increases: $v_{\min} \sim (\Delta\theta/\theta_s)v_0(P/P_F - 1)$. Noting that electro-optical schemes allow registration of $\delta\Phi$ with much higher accuracy than that corresponding to one fringe of the intrinsic interference (the phase shift can be determined with much better accuracy than $2\pi$), one can anticipate the possibility of detection of extremely slow motions $v \sim 100$ nm/s.

Criticality leads also to qualitative peculiarities. Thus, the transverse modes of reorientation are essentially intercoupled due to spatial nonlocality of the laser beam influence. The dynamics of the process is thus described by a system of first order differential equations which usually reveals oscillatory behavior. One may expect "catastrophes," jumps and hysteresis, in the orientation state of the medium with smoothly varying control parameters since the speed of transverse motion proved to be as essential a control parameter as the power of radiation.

The phenomena under discussion are not only of fundamental interest but also have large application potential. Thus, it can be applied to visualization and registration of slow motions and rotations, to power and intensity measurements of laser beams, as well as to measurements of nonlinear optical properties of materials along with the well-known $z$-scan method [11]. Optical nonlinearity, particularly of thermal origin, is characteristic to actually all materials [12], and important applications of the results under discussion may become characterization of convective motions in liquids and in atmosphere; see [12,13] as well as references in [12]. It is interesting to note that the effect under discussion suggests new principles of "nonlinear laser velocimeters"' which are able to register motions of a *transparent* media *across* the beam: *a situation where Doppler-effect methods cannot be applied.*

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