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Light-Induced Pattern Formation in Nematic Liquid Crystals

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We present a theoretical study of the Light-Induced Fréedericksz transition in two geometries where dynamical phenomena induced by the laser light have been observed. We consider a spatially extended system, i.e. we assume the laser beam to be much wider than the thickness of the cell. We suggest that pattern formation phenomena in the plane of the cell entirely due to the light-director interaction should be possible to observe.

Keywords: Light-Induced Fréedericksz transition; Pattern formation; Dynamical phenomena

INTRODUCTION

The fact that intense laser light can induce a Fréedericksz transition in nematics has been known for two decades and this so-called Light-Induced Fréedericksz Transition (LIFT) has been widely studied [1]. LIFT is more interesting than the usual Fréedericksz transition induced by a low frequency field, because in some geometries interesting dynamical phenomena can be observed such as constant precession or precession and nutation of the director [2, 3] and even chaotic director oscillations [4, 5]. These dynamical phenomena arise

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as a result of the interaction between the electric field of the light-wave \vec{E} and the director \vec{n} . On one hand a dielectric torque acts on the director, on the other hand the orientation of the director influences light propagation via the dielectric tensor $\epsilon_{ij} = \epsilon_{\perp}\delta_{ij} + \epsilon_a n_i n_j$ that appears in Maxwell's equations. These phenomena have been studied in the past only in low aspect-ratio systems, i.e. the diameter of the laser spot-size has always been the same order of magnitude as the thickness of the cell.

The question that arises is the following: what if we now consider a large aspect ratio system, i.e. we assume that the cell is being illuminated homogeneously over an area whose size is much larger than the thickness of the cell? The physical system in this case can be considered to be translationally invariant in the $x - y$ plane (the plane of the cell) and thus all solutions of the equations that have been derived by assuming the physical quantities to depend only on the z -coordinate (the coordinate across the cell) remain valid solutions. However, the solutions may be unstable. Then modulations and patterns appear spontaneously in the plane of the cell, as is the case for numerous physical systems such as fluid layers heated from below. A hint that this probably must be the case comes from the knowledge, that for some ranges of ratios of the Frank elastic constants, the magnetic field induced Fréedericksz transition leads to a transversally modulated distortion of the nematic [6, 7]. Another hint comes from the observation that if the local ($x - y$ independent) dynamics of the system is chaotic, invariance in the plane of the cell is not likely to be preserved.

A theoretical investigation of this question can proceed along the following lines: First one can assume the director to depend only on the z -coordinate and seek to derive simple models to describe the dynamical phenomena that have been observed. The 1D PDE-s that describe the system can be reduced using a Galerkin expansion to a few coupled nonlinear ODE-s for the relevant reorientation mode amplitudes (0D model). Then this model can be generalized by allowing a slow dependance of the amplitudes (compared to the wavelength of the light) on the in-plane coordinates x and y . This $x - y$ dependance can be usually neglected in Maxwell's equations and only the elastic torques will give us terms that turn the system of ODE-s of the 0D model into a set of 2D PDE-s. The resulting set of equations can then be used to test the stability of homogeneous stationary states

against periodic perturbations. (Homogeneous in the $x - y$ plane, but not necessarily in the z direction.) An instability with a finite critical wave vector \vec{k}_c means that periodic modulations will appear if the system is carried across the instability. Above the instability, in the weakly nonlinear regime, universal amplitude equations can be derived that supply information on the types of patterns that may appear. The spatial evolution of the phase of oscillatory states can be investigated through universal phase equations.

In what follows we apply these methods to two geometries in which time dependent dynamical phenomena occur. We seek to identify situations where pattern formation in the plane of the cell can arise.

DIRECTOR OSCILLATIONS INDUCED BY OBLIQUELY INCIDENT LIGHT

One geometry where interesting dynamical phenomena have been observed is the case of a linearly polarized light incident on a cell of homeotropically aligned nematic at a slightly oblique angle (Figure 1). The direction of polarization is perpendicular to the plane of incidence. As the intensity of the light is increased, simple stationary LIFT is followed by regular and later chaotic director oscillations [4, 5]. The Galerkin expansion $\varphi(z, t) = A_1(t)\sin(\pi z/L) + A_2(t)\sin(2\pi z/L)$, $\theta(z, t) = B_1\sin(\pi z/L)$ leads to a set of three coupled nonlinear ODE-s for the distortion amplitudes A_1, A_2 and B_1 [8]. The dynamical behaviour that this relatively simple model shows is consistent with existing experiments.

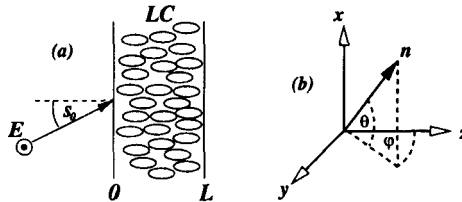


FIGURE 1: a) *Geometry of the setup: an ordinary wave incident at a slightly oblique angle upon a cell of nematic LC with homeotropic orientation.* b) *Definition of the angles describing the orientation of the director.*

Figure 2 shows the phase diagram calculated from our model. The basic state (homogeneous homeotropic orientation) is unstable in the area shaded gray in the figure. Up to a certain angle it loses stability in a stationary bifurcation, and above this in a Hopf bifurcation. The two regimes are separated by the line where the linear eigenvalues become complex (dashed line). The stationary distorted state that develops at lower angles also becomes Hopf-unstable at a slightly higher intensity (dash-dotted line). Beyond this line, the model shows a great variety of nonlinear behaviour [8], among them a very interesting route to chaos through a sequence of gluing bifurcations [9].

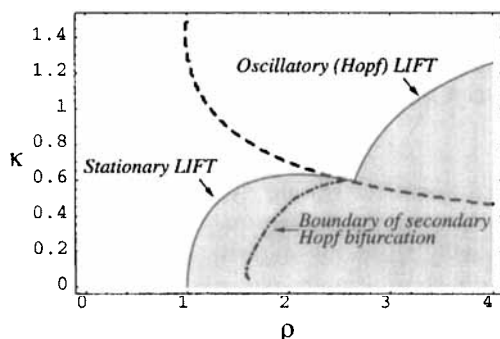


FIGURE 2: Phase diagram of LIFT induced by obliquely incident light. ρ is the intensity normalized by the threshold intensity of LIFT for perpendicular incidence, κ is a parameter that gives the phase shift between the ordinary and extraordinary waves in the cell in the absence of reorientation and is proportional to the square of the angle of incidence.

We may now consider an extension of our model by allowing the amplitudes to vary slowly in the plane of the cell, i.e. we now have $A_{1,2}(x, y, t)$, $B_1(x, y, t)$. Performing a linear stability analysis of the basic state with this extended model we found that the undistorted homeotropic state always loses stability in a spatially homogeneous bifurcation, i.e. $\vec{k}_c = 0$. This is true for both the stationary LIFT (lower region in Figure 2) and the oscillatory LIFT (upper region in Figure 2). For the case of the oscillatory LIFT we have derived the relevant complex Ginzburg-Landau equation which describes the

behaviour of the system in the weakly nonlinear regime. The linear dispersion parameter in this equation turns out to be zero, while the nonlinear dispersion parameter is in such a range that one expects stable plane wave solutions, spirals and - in 1D - stable hole solutions [11].

We have also carried out the linear stability analysis of the stationary distorted state that is stable between the primary bifurcation and the secondary Hopf bifurcation for small angles (lower region of Figure 2). We found that the secondary bifurcation which destabilizes it is homogeneous only if the Frank elastic constants are all equal ($K_1 = K_2 = K_3$), but is not homogeneous otherwise. In fact $\vec{k}_c \neq 0$ for any degree of anisotropy of the elastic constants (here reflection symmetry is broken by the primary transition, so $\vec{k}_c \neq 0$ is actually the generic case). This is contrary to the magnetic field induced transition, where there is a lower threshold to K_1/K_2 [6] or K_3/K_1 [7] below which $\vec{k}_c = 0$ and no stripes appear. Since the instability is nonstationary, a finite \vec{k}_c means the appearance of travelling waves. Figure 3 shows a contour plot of the linear growth rates of periodic perturbations of the homogeneously distorted state as a function of the wavevector in the $x - y$ plane. The largest growth rates (and hence the most unstable modes) are those in the middle of the light circles. The magnitude of \vec{k}_c grows with the anisotropy of the elastic constants and its direction (the direction of wave propagation) is roughly parallel to the in-plane component of the director \vec{n}_p .

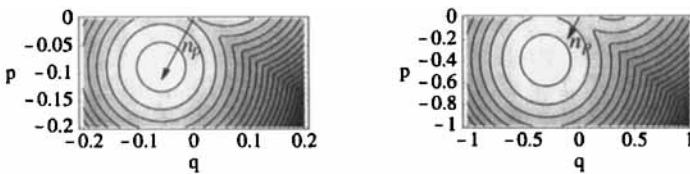


FIGURE 3: Contour plot of the growth rates of periodic perturbations of the homogeneously distorted state as a function of $p = Lk_x$ and $q = Lk_y$ for $K_1/K_3 = 2/3, K_2/K_3 = 1/2$ (left) and $K_1/K_3 = 0.4, K_2/K_3 = 0.2$ (right). For other material parameters see [8].

DIRECTOR PRECESSION INDUCED BY CIRCULARLY POLARIZED LIGHT

If LIFT is induced in a cell of homeotropically aligned nematic with circularly polarized light (Figure 4), the director will not only reorient, but start to precess [2]. The Galerkin expansion $n_p(z, t) = A_1(t)\sin(\pi z/L)$ and $\varphi(z, t) = B_0(t) + B_1(t)\cos(\pi z/L)$ again leads to a set of three coupled nonlinear ODE-s for the amplitudes A_1 (magnitude of reorientation), B_0 (orientation of the distorted state) and B_1 (twist amplitude). The resulting equations describe small angle reorientation and precession of the director in excellent agreement with [2]. The model shows that the amplitudes A_1 and B_1 converge to stationary values, while B_0 will increase linearly with time. Its time derivative (the precession frequency Ω) depends only on A_1 . The basic state loses stability in a subcritical bifurcation, so there is a region of bistability. Figure 5 shows a plot of the stationary state values of the amplitudes and of Ω . There are two stable and two unstable branches which merge at bifurcation points.

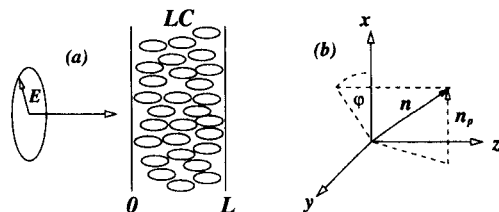


FIGURE 4: *Geometry of the setup: circularly polarized light incident perpendicularly on a cell of homeotropically aligned nematic. The director can be described by n_p , its component perpendicular to the z -axis and φ the azimuthal angle.*

This model can also be generalized to include an $x - y$ dependence of the amplitudes. Investigations then show that branches 1 and 2, which are stable in the 0D theory, remain stable against any finite k perturbation too. It is also possible to derive a phase diffusion equation for the precessing state that describes the spatial evolution of phase disturbances [10]. The general form of this equation is $\partial_t \psi = \alpha \nabla^2 \psi + \beta (\nabla \psi)^2$. In our case, the spatially dependent phase perturbation is $\psi = B_0 - \Omega t$ and the real constants appearing

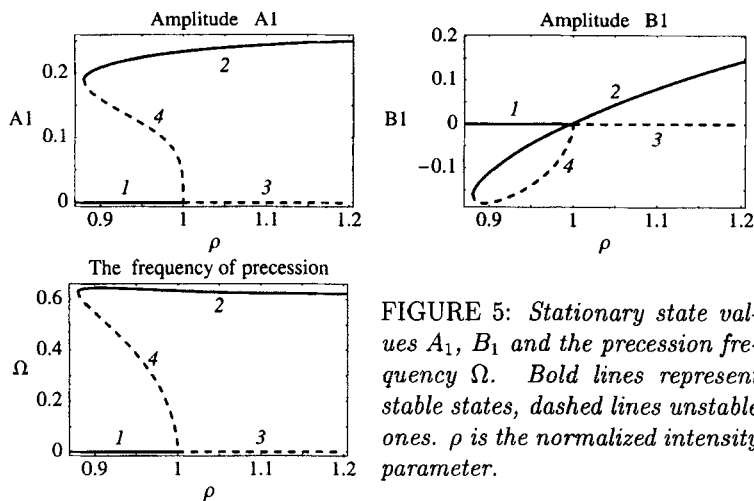


FIGURE 5: Stationary state values A_1 , B_1 and the precession frequency Ω . Bold lines represent stable states, dashed lines unstable ones. ρ is the normalized intensity parameter.

in the equation turn out to be: $\alpha = (K_1 + K_2)/K_3, \beta = 0$. This means that we have plane wave solutions ($B_0 \sim qx + py$), but without group velocity. These are the only attractors and presumably all solutions decay to such states apart from topological point defects. Vortex-like topological defects, whose core however is not described by this equation, should also exist.

We mention that in the case of a Fréedericksz distortion driven by elliptically polarized light, the director can show more complicated dynamical behaviour, various regimes of precession and nutation are possible [3]. It is also feasible to derive a 0D model for this case [12], although this model is much more complicated. It is expected that the phase diffusion equation mentioned in the previous paragraph will have $\beta \neq 0$ for this model. The plane waves then acquire a group velocity and the vortices become spirals.

Summary

As a conclusion, we can say that we have found evidence that the LIFT can lead to pattern formation phenomena in the plane of the cell if a large aspect ratio system can be realized. This can happen in several geometries where dynamical phenomena were observed previously in a low aspect ratio system.

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