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Phase grating created by a shear flow induced instability+

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Abstract

We consider the optical study of the phase grating produced in a convective shear flow instability in a nematic film, using the diffraction pattern of a parallel laser beam, incident normally or oblically on the L.C. cell. In normal incidence, using classical description for phase gratings in optics, we describe quantitatively the time variation of the vertical component of the nematic distortion when a periodic shear is applied to the film. The oblique incidence study leads to the determination of both the vertical component and the horizontal one in the plane of the cell.

Réseau de phase créé par une instabilité de cisaillement dans un nématique

Nous étudions optiquement le réseau de phase produit dans une instabilité de cisaillement d'un film nématique planaire, en utilisant la diffraction d'un faisceau laser qui arrive sous une incidence normale ou oblique sur le film. Dans le cas de l'incidence normale, la variation temporelle de la distortion, sous l'effet d'un cisaillement alternatif, est décrite quantitativement en utilisant des résultats classiques de réseaux de phase. L'étude en incidence oblique permet aussi une détermination de la composante horizontale, le long du cisaillement, de la distortion.

I - Introduction ; convective instabilities in nematics

Nematic liquid crystal films offer a rich variety of convective instabilities, which develop in the presence of a large enough destabilizing agent : an alternating electric field, a temperature gradient or a shear applied between the parallel plates which contain the aligned material[1]. The local average direction of the molecular axis is characterized by a unit vector $\vec{n}$, the director. $\vec{n}$ is also the optical axis of this positive uniaxial material. A regular roll instability can be obtained due to circular flow patterns across the cell. Due to the coupling between the molecular orientation and the flow, a periodic distortion of the molecules takes place, and an optical study of this regular structure is appropriate. Figure 1 gives the velocity and distortion pattern for a typical thermal or electrohydrodynamic instability.

Several authors have considered the diffraction pattern produced by a beam incident on such a grating obtained in electrohydrodynamic instabilities. The study by Carroll[2] extended by Kashnow and Bigelow[3], concerned the low frequency field limit (Williams regime) with rolls having a wavelength Λ comparable with thickness ($d \sim 10$ to 100 microns). The work of Galerne[4] applied to the so called "chevron" regime in the presence of a high frequency field ; the wavelength Λ can be continuously varied to values much smaller than d. He clearly pointed out, from his qualitative study, the existence of two types of effects : one due to the electromagnetic field radiated by the dipoles along $\vec{n}$ and involving a spatial period Λ and one due to the phase grating produced by the phase modulation of $\vec{n}$ of spatial period $\Lambda/2$.

In this work, we apply these notions to a quantitative study of the periodic distorted structure involved in a shear flow of a planar nematic ($\vec{n}$ is strongly aligned, at the boundaries, along the x direction parallel to the limiting plates $z = \pm d/2$). The shear gradient $s = \frac{\partial v_y}{\partial z}$ is constant across z. A detailed discussion of the instability has been given elsewhere and will not be reviewed here. We will concentrate on the optical aspect of the characterization of the distortion which involves both a component of $\vec{n}$ out of the plane xy, n_z ($\ll 1$) leading to the phase grating in normal incidence, and a component n_y ($\ll 1$) along the flow direction.

An alternating shear is applied by displacing periodically the upper glass plate while keeping the lower one fixed as well as the spacing between the plates where the liquid crystal is maintained by capillarity. The periodic resulting distortion of the director calculated in (5) is given in terms of the variation of n_y and n_z with time on figure 2. The curves are characterized by the ratio (T_y/T_z) between the time constants for spontaneous relaxation of the distortions n_y and n_z (in the absence of the coupling between these variables, which triggers the instability). These variations correspond to the so called Y mode (only n_y goes to zero through a period of the shear. A geometrical description of the variations of $\vec{n}$ inside the shear cell is given schematically in figure 3 for the Y mode.

II - Experimental

A schematic view of the shear flow cell is given on figure 4. A narrow parallel laser beam (5 mw He-Ne ; $\lambda = 0.63\mu$) is incident perpendicular (P) or oblically (O) on the cell. In each case we observe, on a screen normal to the beam, two series of diffraction spots, imaging the regular grating formed by the rolls aligned along y, around the central spot (I_o). A first intense series (I_1, I_2, ...) is separated half way between them by a much weaker one (i_1, i_2, ...). The diffraction angle ψ is related quite generally to the period of the diffraction structure P by :

$$\sin\psi = n\lambda/P$$

the wavelength of the laser beam $\lambda = 0,63\mu$

n is the order of the spot.

The intense series corresponds to a period equal to the width of a roll (which is nearly equal to the thickness of the liquid crystal film).

If we assume that the weaker series also contribute to the intense peaks of even order spots, we find that it corresponds to a spatial period $P \sim 2d$ twice as large as the former one, We will discuss the origin of these two series in III,

The intensity of the diffracted spot is read using a photo cell linear in intensity and is recorded together with the displacement of the cell along y. A typical recording of the intensity of the central spot I_o and of I_1, together with the displacement D is given as a function of time on figure 5. If the shear rate $s = 2\ D_{max}/T$, where T is the period of the flow, is small enough (curves a), the intensity I_o is constant and the intensity of the diffracted peaks is zero as no convective instability is present.

Just above threshold (curves b) a diffracted intensity I_1 function of time appears, and I_0 also varies with time. Point B where the shear changes sign corresponds to the maximum distortion of the liquid crystal pattern whereas point A is a point of minimum distortion n_z (see figure 2). When the shear is further increased (curves c) the variation of the distortion also increases but the variation of I_1 becomes more complex. This point will be analysed in part III.

III - Diffraction study

1. Periodicity

The existence of two series of diffraction patterns has already been discussed in the case of electrohydrodynamic instabilities in liquid crystals (3) (4) and we summarize these results.

a) First order series : for the weak spots, the spatial wavelength is Λ. This implies that the effect of the director component n_z is opposite in points a and b of figure 1 and should be of first order in n_z. This diffraction pattern is due to the uniaxial character of the medium of finite thickness and can be understood as due to the emission by elementary dipoles along the average molecular direction. In this case, the distortions $n_z(x)$ and $n_z(x + \Lambda/2)$ give different contributions to the diffraction pattern. In the limit of small distortions, the order of magnitude of the amplitude of the two first diffraction spots, as obtained in the electrohydrodynamic case is proportional to $n_z\psi$ ($n_z^2\ \psi^2$ for the intensity). Such a dependence was also obtained in (3) by considering the optical path of oblique (in plane xz) parallel light. The difference between the optical path for rays passing through regions a and b contains a first order contribution proportional to $n_z\psi$.

b) Second order series : The intense spots (I_1, I_2, ...) are associated to a phase grating structure of spatial period $\Lambda/2$, caused by the variation of birefringence of the nematic as n_z varies. The order of magnitude of this series is given by n_z^2. This second order term also gives rise to the formation of focal lines (stars on fig.1) of period $\Lambda/2$[6], formed alternatively above and below the plane of the layers, which are sometimes used to characterize the instability threshold.

In the experiment by Galerne on the "chevron" regime of the electrohydrodynamic instability, the wavelength of the roll structure could be made much smaller than the thickness $d \sim 30\ \mu$. The angle ψ is relatively large and, near threshold, the first order spot, i_1 is more intense than the second order one I_1. The first order effect is not reported in the work by Carroll[2] because it is weak, except very near threshold, as soon as the wavelength of the periodic pattern becomes large. This is also the case in our work where $\sin \psi \simeq 1/300$ and the first diffracted peak i_1 much weaker than the I_1, I_2, ... series except possibly very near threshold. For this reason, we will consider only the series of more intense peaks and examine only the phase grating mechanism.

2. Phase grating diffraction pattern[7][8]

We present theoretical and experimental considerations on the phase grating diffraction. The cases of normal and oblique incidence will be studied.

a) Normal incidence-Fourier formalism

If a plane wave of unit amplitude is incident on a grating, the optical field distribution just past the grating may be calculated as a function $u(x)$. The diffraction pattern of the grating at infinite distance is the Fourier transform of $u(x)$. An equivalent approach is to calculate

u(x) as a superposition of plane waves. Each spot of the diffraction pattern can be shown to be the Fourier transform of one of these plane waves.
The main point of the diffraction calculus is then the determination of u(x), generally known as"the amplitude transmittance of the grating".

In the case of normal incidence, following (13), we express u(x) as :

$$u(x) = u_o \exp\left[ik \int_{-d/2}^{d/2} n_{eff.}(z)\,dz\right]$$

with

$$k = \frac{2\pi}{\lambda}$$

λ being the incident wavelength.
That expression may be calculated using the usual definition of the effective index n_{eff}

$$\frac{1}{n^2_{eff}} = \frac{\cos^2\theta}{n_e^2} + \frac{\sin^2\theta}{n_o^2}$$

In our case, θ is shown to be the director angle, and assuming a sinusoïdal variation across z it is described by :

$$\theta = \theta_o.\cos\frac{\pi z}{d} \cdot \cos q_x x$$

with

$$q_x = \frac{2\pi}{\Lambda} .$$

Suitable approximations give :

$$n_{eff} = n_e + \frac{n_e(n_e+n_o)}{2\,n_o^2} \cdot (n_o-n_e)\theta^2.\cos^2\frac{\pi z}{d} \cdot \cos^2 q_x x$$

and the expression of the amplitude transmittance of the grating :

$$u(x) = u_o \exp[ik\,n_e d]\,\exp[i\phi\cos^2 q_x x]$$

with

$$\phi = \frac{k}{2}\frac{n_e(n_o+n_e)}{2\,n_o^2}.(n_o-n_e)\theta_o^2.d$$

The diffraction pattern is given by the Fourier transform of $u(x)$, and can be calculated using the formula :

$$\exp[i\,m\cos\alpha] = \sum_{n=-\infty}^{+\infty} \exp[i\,n(\alpha+\frac{\pi}{2})]\,.\,J_n(m)$$

where J_n is the n^{th} order Bessel function.

One should note that, if the left side of this expression is considered as a complex-shaped wave, the right side gives its decomposition into an infinite superposition of plane waves of amplitude $J_n(m)$. Substitution in the expression of $u(x)$ gives :

$$u(x) = u_o \exp[i(k\,n_e d - \frac{\phi}{2})] \sum_{n=-\infty}^{+\infty} \exp[i\,n\frac{\pi}{2}]\exp[i\,n\,2q_x x]\;J_n(\frac{\phi}{2})$$

The n^{th} plane wave of that decomposition gives the n^{th} spot of the diffraction pattern whose amplitude is given by :

$$A_n = u_o \exp[i(kn_e\,d - \frac{\phi}{2})]\exp[i\,n\,\pi/2]\,J_n(\phi/2)$$

Its intensity is :

$$I_n = ||A_n||^2 = ||u_o||^2\,J_n^2(\phi/2)$$

$||u_o||^2$ is an unknown coefficient that is the same for any order. It is of no interest in our case, and intensities may be normalised in order to get unit intensity of the zeroth order diffraction spot in the absence of instability. The functions $J_0^2(\phi/2)$ and $J_1^2(\phi/2)$ are plotted on figure 6b. Their shape will provide qualitative explanation of further experimental results.

b) Normal incidence - Experimental

The time dependence of the intensity in the zero and first order spots of the diffraction patterns is plotted on figure 5 together with the displacement of the upper plate. Curves show typical shapes that can be qualitatively explained using $J_0^2(\phi/2)$ and $J_1^2(\phi/2)$ (see figure 6a).

On figure 5b, the shear is just above threshold. At point A, the intensity of the zeroth order spot is maximum and close to unity : Parameter ϕ is at its minimum value. It increases with time up to a maximum corresponding to point B. Intensity in the zeroth order spot is then minimum while it is maximum in the first order spot. At point B, the shear changes sign and ϕ starts decreasing again down to a minimum distortion corresponding to the initial one at point A.

When the shear is increased (Figure 5c) the variations of intensities may be more complicated. At point A, ϕ takes its minimum value. I_o is maximum and is nearly equal to one while I_1 is minimum. From this point ϕ increases with time. I_o decreases to a minimum close to zero at point C. I_1 increases to point D corresponding to the maximum of $J_1^2(\phi/2)$ and then decreases to point C.

At point C, the shear changes sign and $J_0^2(\phi/2)$ and $J_1^2(\phi/2)$ follow an opposite variation corresponding to a continuous decrease of ϕ to its minimum value.

A more quantitative work can be done on these curves, in order to recover the parameter ϕ from them. This determination is plotted on figure 6 b as a function of time, using the numerical values of figure 5 c.
One should note that the maximum normalized value of I_1, that should be 0.34 at point D, is experimentally found to be 0.28 in this experiment.
We have normalized the intensity values I_1 in order to get the correct value of the maximum rather than just to the maximum of I_0 (The difference can be due to "parasitic forward light scattering by small particles" mentioned in Carroll work. This effect was so large in his experiment that the central peak intensity could not be used to study the diffraction pattern).
On the same figure, we give the variations of n_z calculated from the data of fig.2 for a value of T_y/T_z=.74 evaluated independently for this experiment[9]. The agreement is good and shows well the assymetry between the down and up parts of the curves due to the difference between the time constants T_y and T_z.

On Fig.7, we have plotted a series of determinations of the parameter ϕ (proportionnal to n_z^2), using the two first diffraction orders and following the method described in Fig.6. The sample thickness was 150µ and the shear was progressively decreased down to the threshold value s_c. The linear variation is consistent with a "mean field" behaviour which should apply to the description of these instabilities. We have also indicated a value of the average distortion deduced from that of ϕ and from the knowledge of the refractive indices. This rather small value explains the good agreement between experiments and the Fourier theory which ignore strong deflections of the optical beams.

Let us emphasize that we consider these experimental results as only preliminary. A more detailed study can be performed and the variation of the shape of the above curve easily obtained from the application of a high frequency electric field (which decreases T_z and possibly leads to an interchange of the n_y and n_z variations[5]) or a low frequency one (which increases T_z and would give variations of n_z more assymetric as well as of smaller amplitude.)

c) Oblique incidence

The laser beam is now oblically incident with respect to the plane of the rolls (fig.3) ; the plane of incidence being parallel to the rolls (outside the cell, the wavevector $\vec{k} = 0, k_o \sin\alpha, k_o \cos\alpha$). We use only the extraordinary component of light (polarization $\vec{p}$ along x). The oblique laser beam should be sensitive both to the n_y and n_z distortion. In the limit of small deformations and using a weakly oblique beam (α small), the z component of the wave vector within the cell is given by :

$$k_z = C - \frac{1}{\cos\delta} k_o \frac{n_e(n_e+n_o)}{2\,n_o^2} (n_o-n_e)(n_z \cos\delta + n_y \sin\delta)^2$$

where δ is the incidence angle within the layer $\delta \simeq \frac{\alpha}{n_e}$.

The phase shift of the extraordinary beam across the sample is

$$\Phi = \frac{k_o}{\cos\delta} \frac{n_e(n_e+n_o)}{2\,n_o^2} (n_o-n_e) \int_{-d/2}^{d/2} (n_z \cos\delta + n_y \sin\delta)^2 dz$$

If we assume, in agreement with the case of normal incidence, that n_y and n_z have nearly the same z dependence :

$$n_{y,z} = n_{y_m, z_m} . \cos(q_x x) \cos(\pi \frac{z}{d})$$

One gets :

$$\Phi(x) = \frac{n_e(n_e+n_o)}{2\,n_o^2} \frac{k_o}{2.\cos\delta} (n_o-n_e)\cos^2 q_x x\, (n_{z_m} \cos\delta + n_{y_m} \sin\delta)^2$$

The intensity in the n^{th} spot is then :

$$I_n = ||u_o||^2 . J_n^2(\phi/2)$$

with

$$\phi = \phi_o(1 + \frac{n_{y_m}}{n_{z_m}} \text{tg } \delta)^2 \cos\delta$$

where ϕ_o is the value of ϕ when $\delta = 0$ (normal incidence).
It is possible to estimate the ratio $\frac{n_y}{n_z}$ by comparing the intensity for $\delta = 0$ and that for a given small angle δ. Using a semitransparent mirror arrangement to split the beam into a normal plus an oblique one, the intensity corresponding to the two values of δ can be measured simultaneously. A typical recording of the central spots is given on figure 7. 8
We note that the variation for the oblique beam shows a strong assymmetry over two consecutive half periods as the two alternances n_y and n_z of the Y mode, used in the experiment, are no longer symmetrical with respect to the beam direction.
Using the relation between I_o (or I_1) and the corresponding Bessel function, one gets :

$$\phi_o/2 = 1.4 \text{ for } \delta = 0$$

$$\phi_o/2 = 1.02 \text{ and } 2.04 \text{ for } \delta = 5°.$$

this gives, using the above expression for ϕ, $n_{y_m}/n_{z_m} = 2.3$ and 1.7 for the two half periods. The average between these two values is of the order of magnitude of that calculated numerically elsewhere(10).

Conclusion

Thanks to the large birefringence of nematics, convective instabilities associated with a periodic distortion can be studied along the lines of ordinary phase gratings in optics[8]. This description leads to original informations concerning the hydrodynamics description of liquid crystals such as the quantitative description of the time variation of the distortion. We can in turn ask how much use can be these elements for applications in optics. The use of an hydrodynamic instability mode rather than the electrohydrodynamic one, where inhomogeneous distribution of charges cause strong inhomogeneities in the pattern, lead to a reasonably homogeneous and periodic grating. The amplitude of of the distortion and, for certain modes, the spatial periodicity can be adjusted at will. It is possible to use rather thin cells (∿ 30μ) where the absorption of light and the difficulties arising from finite thickness effects can be minimized. The obstacle due to the existence of moving parts can be overcome, at least for fundamental studies, in recent high frequency shear studies[11] [12] where the total displacement is limited to microns. We may evoke possible connections with applications in optics : beam deflectors, grating couplers[13] and distributed laser structures[14] in integrated optics (in the latter case it is known that the threshold of dyes is strongly decreased near the nematic transition). Materials which are only weakly absorbant and strongly birefringent in the nematic range are now available (tolanes). Very thin layers should be used in order to reduce the viscosity limited relatively long time constants of the devices. We also feel that the use of the hydrodynamic instabilities rather than the electrohydrodynamic one can lead to very regular grating structures over large areas (∿ cm^2).