Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006

Mol. Cryst. Liq. Cryst., Vol. 320, pp. 1–9
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A New Interferometric Technique for the Measurement of Elastic Anisotropy of Nematic Liquid Crystals

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It is well known that a continuous wave laser beam impinging on a nematic liquid crystals film can induce a change in the orientation of the molecular director. In the case of homeotropic alignment and for a p-polarized obliquely incident beam, the reorientation process has no threshold and the sample exhibits a giant optical nonlinearity due to the laser-induced change in refractive index of the medium. In the small distortion approximation the phase change is governed by a linear differential equation whose coefficients depend on the ratio between the elastic constants. In this paper we describe an interferometric technique based on high precision two-dimensional spatial fringe analysis Fourier method for measuring the elastic constants of nematic liquid crystals. Since the determination of the elastic anisotropy relies on the measurement of the ratios between the elastic constants, this technique provides a powerful and high accuracy method for characterizing nematic liquid crystals.

Keywords: LIQUID CRYSTALS, NONLINEAR OPTICS

INTRODUCTION

Interferometric techniques for measuring the refractive index of a sample are performed by means of an optical set-up in which the optical path

length of a laser beam is modified by the presence of the investigated sample. For wavefront division interferometers the interference fringes are visible everywhere in the zone where object and reference beams overlap. That is why they are called unlocalized fringes.

In this paper we describe a new interferometric technique for measuring the elastic anisotropy of nematic liquid crystals. This technique employs a division wavefront interferometer previously developed for measuring the refractive index of homogeneous transparent materials [1, 2]. In the case of homeotropic alignement of the liquid crystal molecules and for normal incidence of the pump-beam, no reorientation can be induced until the laser beam intensity reaches a characteristic threshold value. This phenomenon is known as the Optical Freedericksz Transition (OFT). At oblique incidence, the reorientation process has no threshold and the sample exhibits a Giant Optical Nonlinearity [3, 4, 5]. When no reorientation is induced, the interference pattern recorded by a CCD camera (reference interference pattern) is a set of parallel straight fringes whose pitch depends on the angle between the two interfering diffraction orders as shown in Fig. 1.

When the liquid crystal sample is irradiated the resulting change in the refractive index produces a local variation in the interference fringe pitch (modified interference pattern). The two-dimensional phase distribution of the modified interference pattern has been determined by means of a fully automated fringe pattern processing system based on Fourier transform method (FTM) for phase retrieval [6].

The elastic anisotropy of a nematic liquid crystal film as given by the ratios between the Frank's elastic constants has been determined by fitting the experimental phase values to the numerically computed ones. This technique allows to perform measurement of the elastic anisotropy with high accuracy and spatial resolution. The ultimate resolution of the method is limited by the spatial resolution of the CCD camera.

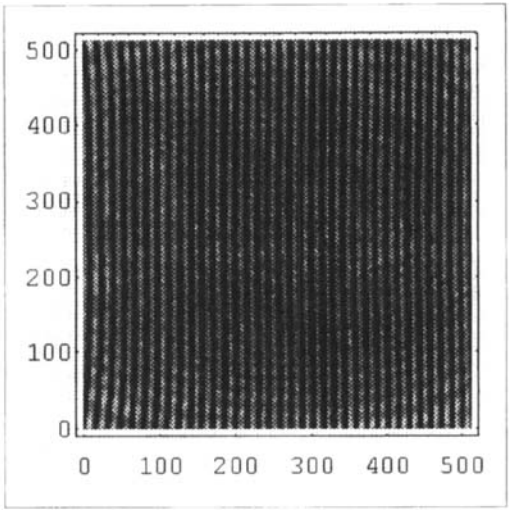

FIGURE 1 Interference pattern for an undistorted liquid crystal sample digitized into a 512x512 pixels array.

EXPERIMENTAL SET-UP

The principle of operation of the interferometer is illustrated in Fig. 2. An expanded and collimated He-Ne laser beam is reflected downward onto the measuring arrangement. This arrangement consists of a tiltable 1200 *lines/mm* reflection diffraction grating G, mounted with a mirror M on a compact base plate. The direction of the grid rulings is perpendicular to the drawing plane. Part A of the illuminating beam strikes the grating directly. This part of the beam serves as an object beam when the sample is interposed on its path. Part B of the illuminating beam impinges the grating via mirror M. This second bundle with undeformed wavefront serves as reference. The angle of the incidence of the illuminating beam is chosen such that the +1 and -1 diffraction orders of the object and reference light beams, respectively, are directed along the grating normal towards the CCD camera. This condition is obtained when the grating and the mirror are perpendicular to each other. When the mirror is slightly tilted with respect to the grating normal an interference fringe pattern,

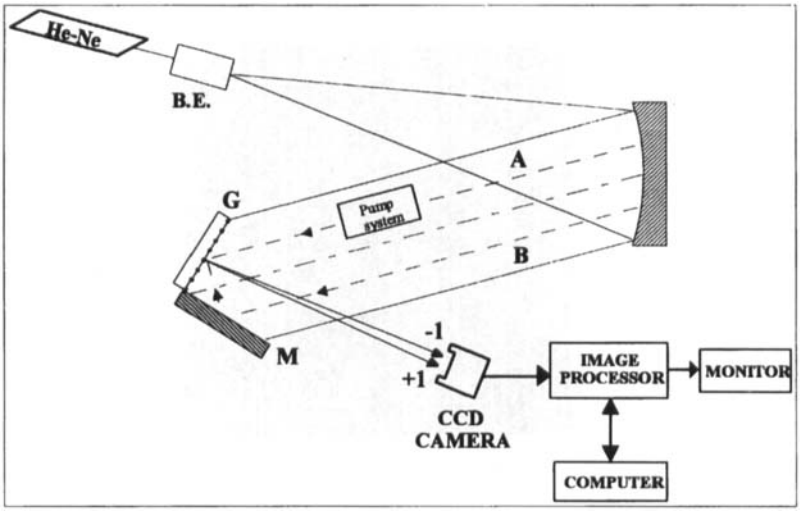

FIGURE 2 Lay-out of the interferometric set-up.

that results from the superposition of the two light diffraction orders, is detected by the CCD (array size: 756x581; pixel size: $11\mu m$). The reference interference pattern is a family of high contrast parallel straight fringes with a spatial frequency $\delta = \sin(\alpha/\lambda)$, where $\lambda = 632.8nm$ is the laser wavelength and α is the angle between the two interfering diffraction orders. The interferometer provides the possibility of easily adjusting the pitch of the reference interference pattern to select the proper spatial frequency in order to apply FTM[6, 7]. The Pump system block in the lay-out of the interferometer represents a pump-probe apparatus for the characterisation of microscopic samples (Fig. 3). It lies in a plane perpendicular to the interferometer. A pair of convergent lenses $L1$ and $L2$, in a confocal arrangement, is interposed in the A arm of the interferometer to focus the object beam onto the sample. As explained before, in this condition the reference interference pattern is recorded by the CCD. The intensity of the pump laser is attenuated continuosly by means of two polaroid films $D1$ and $D2$. A lens system mounted on a rotating mount focuses the p-polarized pump beam, coming from an optic fiber, onto the sample at

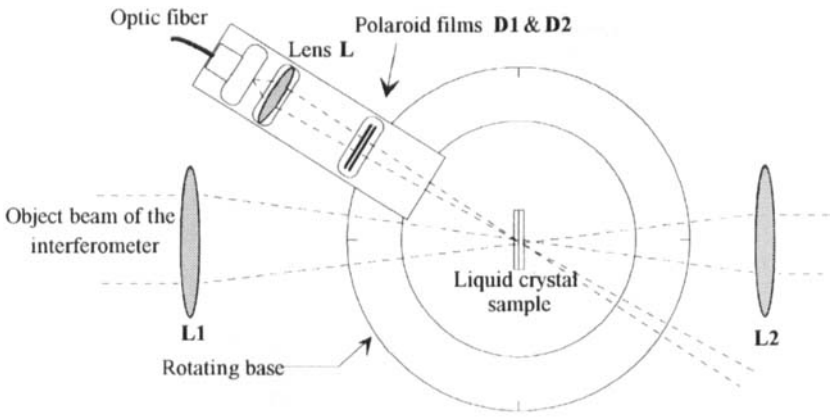

FIGURE 3 Scheme of the pump-probe system.

an incidence angle $\alpha = 40°$. The sample surface is perpendicular to the propagation direction of the object beam. The sample used is a $50\mu m$ homeotropic layer of E7 produced by Merck, doped by an antraquinone dye AQ2 at 0.2% in weight. The measured pump-beam waist at $1/e^2$ of the maximum intensity is $(121 \pm 6)\mu m$.

EXPERIMENTAL RESULTS

The laser intensity used in the experiment is low, so that the resulting molecular distortion in the sample is very small. For such small distortion the linearized law governing the spatial modulation of the molecular director $\hat{n} = (\sin\theta, 0, \cos\theta)$ is given by [8]:

$$\left(\frac{k_{11}}{k_{33}}\right)\frac{\partial^2\theta}{\partial x^2} + \left(\frac{k_{22}}{k_{33}}\right)\frac{\partial^2\theta}{\partial y^2} + \frac{\partial^2\theta}{\partial z^2} + \left(\frac{\pi}{L}\right)^2\left(\frac{\sin\alpha}{\sqrt{\epsilon_o}}\right)\left(\frac{I}{I_{Fr}}\right) = 0 \qquad (1)$$

where

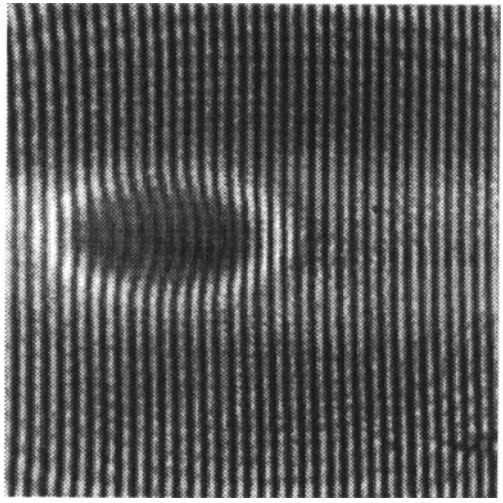

FIGURE 4 Interference pattern observed when a doped nematic liquid crystal sample is irradiated by an He-Ne laser beam (incident power 1.5 mW, spot diameter $(121 \pm 6)\mu$m)

$$I_{Fr} = \frac{c\epsilon_e k_{33}}{(\epsilon_e - \epsilon_o)\sqrt{\epsilon_o}} \left(\frac{\pi}{L}\right)^2 \tag{2}$$

is the threshold intensity for the OFT at normal incidence (c is the speed of light in the vacuum and *cgs* units have been used). In Eqs. 1 & 2 $\epsilon_e = n_e^2$ and $\epsilon_o = n_o^2$, where n_e and n_o are the extraordinary and ordinary refractive indeces, $k_{ii}(i = 1, 2, 3)$ denote the nematic elastic constants for splay, twist and bend, respectively, L is the thickness of the sample and α is the previously defined pump laser incidence angle. The intensity I appearing in Eq. 1 is the projection of the incident intensity profile I_0 on the input face of the nematic film. Since in the experiment we have used a dye doped nematic liquid crystal sample, all the reorientation theory still holds provided that I_{Fr} is substituted by $I_{Frdye} = I_{Fr}/\eta$, where in our case, $\eta \approx 20$[9, 10]. The η factor has been estimated measuring the lowering of the threshold intensity for OFT of our sample with respect to an analogous one without doping [11, 12]. By tak-

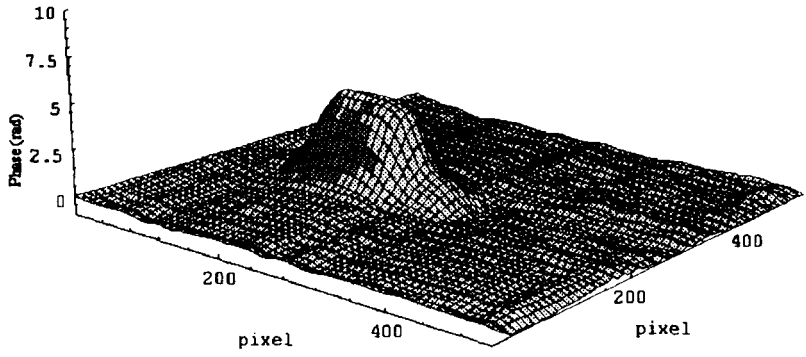

FIGURE 5 Unwrapped phase map relative to the interference fringes pattern of Fig. 4

ing into account that the phase change ψ due to reorientation is given by

$$\psi = \beta\langle\theta\rangle$$

$$\beta = \left[\frac{2\pi(\epsilon_e - \epsilon_o)L}{\epsilon_e \lambda}\right] \sin\alpha$$

where $\langle\rangle = L^{-1}\int_0^L dz$ denotes the spatial average along the sample, we rewrite Eq. 1 in the following form:

$$l_x^2 \frac{\partial^2\psi}{\partial x^2} + l_y^2 \frac{\partial^2\psi}{\partial y^2} - \psi + a\frac{I}{I_{Fr}} = 0. \tag{3}$$

Eq. 3 describes the distribution of the phase change $\psi(x, y)$ in the linear approximation. We have defined:

$$l_x = \frac{L}{\pi}\sqrt{\frac{k_{11}}{k_{33}}}$$

$$l_y = \frac{L}{\pi}\sqrt{\frac{k_{22}}{k_{33}}}$$

$$a = \frac{\sin\alpha}{\sqrt{\epsilon_o}}\beta$$

where l_x and l_y are the diffusion lengths describing the distortion decay along the x and y axis, respectively. In order to determine, from the recorded interference fringe patterns, the two-dimensional phase map $\psi(x,y)$, one dimensional fast Fourier transform has been applied along each row of the digitized images of the reference and modified interference patterns. The values of the reconstructed phase of both interferograms are wrapped in the range between $-\pi$ and π. From the wrapped phase the unwrapped phase values can be computed by using standard two-dimensional unwrapping procedure [6, 7]. The demodulated phase distribution is determined by subtracting from the phase of the deformed interferogram the phase of the reference one pixel by pixel. By fitting the experimental data to the theoretical model we determine the elastic anisotropy of the nematic liquid crystal in terms of the ratios k_{11}/k_{22} and k_{11}/k_{33} between the elastic constants. From the value $k_{33} = 15.3 \times 10^{-7}$ dyne, determined by measuring the OFT, we obtain the following results for the elastic constants k_{11} and k_{22}:

$$k_{11} = (12.10 \pm 0.05) \times 10^{-7} dyne$$

$$k_{22} = (6.53 \pm 0.05) \times 10^{-7} dyne$$

that correspond to diffusion lengths $l_x = 14.146$mm and $l_y = 10.400$mm.

We point out that these values are in good agreement with those reported in Ref. [8] obtained by using a different interferometric technique.

CONCLUSIONS

In this paper we have described a new interferometric technique for measuring the elastic constants of nematic liquid crystals. This technique employs an automated fringe pattern processing system based on Fourier

transform method to determine the laser induced phase change distribution over the irradiated sample. The technique allows to perform high accuracy measurements of the elastic anisotropy of nematic liquid crystal films.

Experimental results obtained by applying this technique to the measurement of elastic anisotropy of a dye-doped nematic liquid crystal film show clearly the capability of the method and the high spatial resolution it provides.

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