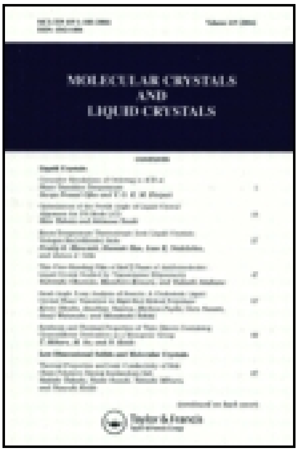

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Measurements of the Orientational Elastic Constants of the Nematic Liquid Crystal by A Four-Wave Mixing

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MEASUREMENTS OF THE ORIENTATIONAL ELASTIC CONSTANTS OF THE NEMATIC LIQUID CRYSTAL BY A FOUR-WAVE MIXING

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Abstract: The diffraction efficiency of an optically induced diffraction grating was measured in a homogeneously aligned cell of a commercial nematic mixture $ZLI1738$. Both self-diffraction of the pump beams and the diffraction of a probe beam were studied. The diffraction efficiency was calculated analytically with some reasonable approximations. In the calculation the finite beam sizes were taken into account. From the experimental data bend and splay orientational elastic constants were calculated.

INTRODUCTION

The giant orientational optical nonlinearity of nematic liquid crystals (NLC) manifests itself in many non-linear optical effects such as self-focusing, self-phase modulation, phase conjugation, optical bistability and self-diffraction of crossed laser beams[4,5,6,7]. Due to the low necessary optical power density needed, these effects are interesting mainly because of the possible use in optical devices. At the same time they provide also the information on the fundamental physical properties of the nematic phase.

Self-diffraction of two crossed laser beams was already successfully used to study the orientational elastic constants of a nematic sample[2]. In a recent paper[1] the splay and bend orientational elastic constants were measured by using an additional weaker beam to probe the induced diffraction grating, formed by two crossed pump beams. In that paper the diffraction efficiency was calculated with a plane wave approximation.

In this report we present a more thorough calculation of the diffraction efficiency for the Gaussian beams and show, that the corrections obtained are quite large and have to be taken into account in the analysis of the experiment. We also report on the measurements of the diffraction efficiencies for both self-diffraction and the diffraction of a probe beam. The dependence of the diffraction efficiency on the angle of incidence of the probe beam on the diffraction grating was measured as well. The position of the maximum of the diffraction efficiency confirms the need for the finite beam sizes to be included in the calculation. From the experimental data splay and bend orientational elastic constants were determined.

THEORY

Grating Formation

The formation of the diffraction grating is discussed in accordance with the geometry shown on fig. 1. Two linearly polarized coplanar laser beams with the wave vectors $\mathbf{k}'_1$ and $\mathbf{k}'_2$ are incident on the homogeneously aligned nematic cell. Their polarization is in the xz-plane. Due to the coupling between the NLC and the optical field, the director axis reorients in the xz-plane by an angle θ with respect to the initial orientation along $\mathbf{n}_0$. The coupling between the director $\mathbf{n}$ and the optical field can be described by the continuum elastic theory of nematics[3]. Its contribution to the free energy of the NLC is $-\frac{1}{2}\epsilon_0\epsilon_a(\mathbf{E}\cdot\mathbf{n})^2$, where $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ and $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the components of the dielectric tensor parallel and perpendicular to the director $\mathbf{n}$.

In deriving the reorientation angle θ the calculation done in the previous paper of I. Drevenšek and M. Čopič[1] is followed with the Gaussian shape of the beams being taken into account.

The time average of the coupling term between the director and the optical field is given by

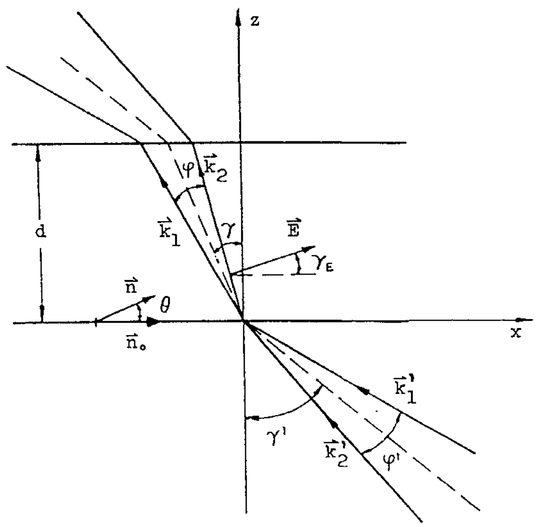

Figure 1: *Experimental geometry.* The pump beams are incident to the homogeneously aligned NLC cell of thickness *d*. The angle φ' between the beams is small compared to γ'.

$$\overline{(\mathbf{n}\cdot\mathbf{E})^2} = \Big\{|E_1||E_2|(\cos^2\gamma_E + \theta\sin 2\gamma_E)\cos(\mathbf{qr}) + \\ +\frac{1}{2}\left(|E_2|^2+|E_1|^2\right)\left(\cos^2\gamma_E+\theta\sin 2\gamma_E\right)\Big\}\cdot \\ \cdot\exp\left\{-2\left((x\cos\gamma+z\sin\gamma)^2+y^2\right)/w_p^2\right\} \quad , \tag{1}$$

where $|E_1|^2$ and $|E_2|^2$ are the beam intensities at the centre of the first and the second beam, and w_p is the beam waist in the NLC cell and is connected to the beam waist w_p' outside the cell by $w_p = w_p' \cos\gamma/\cos\gamma'$, where γ and γ' are the angles between the z-axis and the propagation direction inside and outside the cell. Because NLC is an optically uniaxial material, the angle of the polarization direction γ_E in the cell is not equal to the angle of the propagation direction γ. The relation between the two angles is

$$\tan\gamma_E = \frac{\epsilon_\parallel}{\epsilon_\perp}\tan\gamma \ . \tag{2}$$

The grating vector $\mathbf{q}$ is (see fig. 1)

$$\mathbf{q} = \mathbf{k}_2 - \mathbf{k}_1 = (q_1\ ,\ 0\ ,\ q_3) =$$

$$= \; k_p n_p \varphi (\cos\gamma \, , \, 0 \, , \, \sin\gamma) = q (\cos\gamma \, , \, 0 \, , \, \sin\gamma) \; , \qquad (3)$$

where k_p is the wave vector magnitude of the pump beams outside the cell, q is the magnitude of the grating vector and n_p is the index of refraction.

The reorientation of the director axis can be obtained by taking the ansatz in the form

$$\theta = \sin(pz) \left[u_0 + u_1 \cos(\mathbf{q} \cdot \mathbf{r}) \right] \exp \left\{ -2 \left((x \cos\gamma + z \sin\gamma)^2 + y^2 \right) / w_p^2 \right\} \; . \qquad (4)$$

It is assumed that the profile of the change in $\mathbf{n}$ has the same spatial dependence as the intensity of the pump beams. Strong anchoring at the bounding plates is assumed as well. Therefore p should be equal to π/d, so that the boundary conditions $\theta(z = 0) = 0$ and $\theta(z = d) = 0$ are satisfied.

By putting θ into the expression for the free energy and minimizing the energy over the sample volume, parameters u_0 and u_1 are obtained:

$$u_0 = \frac{\epsilon_0 \epsilon_a \left(|E_2|^2 + |E_1|^2 \right) \sin(2\gamma_E)/\pi}{K_1 \left(p^2 + 2 \sin^2 \gamma / w_p^2 \right) + K_3 \; \left(2 \cos^2 \gamma / w_p^2 \right)} \qquad (5)$$

and

$$u_1 = \frac{2 \epsilon_0 \epsilon_a |E_1| |E_2| \sin(2\gamma_E)/\pi}{K_1 \left(p^2 + q_3^2 + 2 \sin^2 \gamma / w_p^2 \right) + K_3 \left(q_1^2 + 2 \cos^2 \gamma / w_p^2 \right)} . \qquad (6)$$

The beam waists of the pump beams appear in the denominators of u_0 and u_1. They become important, when w_p is comparable to or smaller than the cell thickness or the grating period.

Diffraction Efficiency

The periodic part in the spatial dependence of θ represents the diffraction grating. The pump beams themselves are diffracted on this grating (self-diffraction). However, the diffraction of a weaker probe beam can also be studied. The use of the probe beam provides much more experimental freedom than the studies of self-diffraction. As already pointed out by I. Drevenšek and M. Čopič the grating cannot be considered as a thin phase grating. Because the sample is thick compared to the wavelength of light, the coupled wave theory should be used.

In deriving the diffraction efficiency one starts from the wave equation for an anisotropic medium

$$\nabla \times (\nabla \times \mathbf{E}) = k_0^2 \left(\underline{\epsilon} + \delta \underline{\epsilon} \right) \mathbf{E} \;\; , \qquad (7)$$

where $\mathbf{E}$ is the electric field of the probe beam that is incident to the NLC cell at an angle α' with respect to the z-axis. $\mathbf{E}$ can also be the electric field of the pump beam when self-diffraction is considered. In that case $\alpha' = \gamma'$. $k_0 = \omega/c_0$ is the magnitude of the wave vector of the probe or the pump beam outside the cell.

The electric field of the probe beam can be written as a sum of spatial Fourier components

$$\mathbf{E} = \int_{-\infty}^{\infty} \mathbf{e}_q \psi_q(z) \exp\left\{i\left(qx + \beta_q z\right)\right\} dq \ , \tag{8}$$

where $\mathbf{e}_q$ is the polarization direction and ψ_q are the amplitudes that change slowly with z. They are obtained by the inverse Fourier transformation. Because the medium is uniaxial, the transverse wavenumber q and the longitudinal propagation constant β_q of the probe beam inside the cell are connected by

$$\frac{\beta_q^2}{\epsilon_{\parallel}} + \frac{q^2}{\epsilon_{\perp}} = k_0^2 \ . \tag{9}$$

The ansatz for θ can also be written as a sum of spatial Fourier components

$$\theta_p = \int_{-\infty}^{\infty} \tilde{u}_1(\tilde{q}, z) \exp\left\{i\tilde{q}x\right\} d\tilde{q} \ . \tag{10}$$

Only the periodic part of the reorientation angle is written in (10), because only this part is responsible for the formation of the diffraction grating. By putting θ_p in the wave equation (7), taking the projection of the equation on the polarization direction and keeping only the term for the diffraction peak around $q_0 + q_1$, where q_0 is the central transverse wave number of the probe beam and q_1 is defined in equation (3), one obtains

$$\begin{aligned} \frac{\partial \psi_q(z)}{\partial z} &= \frac{AB\sin(pz)}{S(q)} \int_{-\infty}^{\infty} dq'' \left[\psi_{q''}(z) \exp\left\{-\frac{w_p^2}{8\cos^2\gamma}\left(q'' + q_1 - q\right)^2\right\} \cdot \right. \\ & \left. \cdot \exp\left\{i\left(\beta_{q''} - \beta_q\right) z\right\} \cdot \exp\left\{i\left\{q_3 - \left(q_1 + q'' - q\right)\tan\gamma\right\} z\right\}\right] \ , \end{aligned} \tag{11}$$

$$A = \frac{ik_0^2}{2}\epsilon_a \sin 2\alpha_E \ , \tag{12}$$

$$B = u_1 w_p / \left(4\sqrt{2\pi}\cos\gamma\right) \ , \tag{13}$$

$$S(q) = \beta_q \cos^2\alpha_E - \frac{1}{2} q \sin 2\alpha_E \ . \tag{14}$$

α_E is the angle between the direction of polarization and the direction of propagation of the probe beam.

To be able to calculate the above integral analytically further approximations have to be made. It is assumed that the divergence of the diffracted and the transmitted beams is smaller than the diffraction angle, so that the contributions to the diffracted peak with the transverse wavenumber q result only from the nondiffracted part of the probe beam. It is also assumed that the probe beam is not significantly depleted upon passing through the cell. Then $\psi_{q''}(z)$ in the expression (11) is independent of z and is equal to

$$\psi_{q''}(z=0) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp\left\{-\frac{x^2\cos^2\alpha}{w^2}\right\}\exp\left\{i\left(q_0 - q''\right)x\right\}\, dx \ , \tag{15}$$

where w is the beam waist of the probe beam. By putting $\psi_{q''}(z=0)$ into (11) and integrating the expression over z, one obtains

$$\begin{aligned}\psi_q(d) &= \int_0^d \frac{\partial\psi_q(z)}{\partial z}\, dz = \\ &= \frac{ABC}{S(q)}\int_{-\infty}^{\infty} dq'' \left[\frac{p\left(\exp\{i\xi d\}+1\right)}{p^2-\xi^2}\exp\left\{-\frac{w^2}{4}\left(q_0 - q''\right)^2\right\}\cdot\right. \\ &\quad \left.\cdot\exp\left\{-\frac{w_p^2}{8}\left(q'' + q_1 - q\right)^2\right\}\right] \ ,\end{aligned} \tag{16}$$

where

$$\xi = q_3 + \beta_{q''} - \beta_q - \left(q_1 + q'' - q\right)\tan\gamma \ , \tag{17}$$

$$C = w/\left(2\sqrt{\pi}\cos\alpha\right) \ . \tag{18}$$

In the first approximation it can be assumed that the factor

$$\frac{p\left(\exp\{i\xi d\}+1\right)}{p^2-\xi^2} \tag{19}$$

does not change significantly over the region where the exponential part of the integrand in (16) significantly differs from zero. Then this factor can be put in front of the integral and is evaluated at $q = q_0 + q_1$ and $q'' = q_0$. By a straightforward integration first $\psi_q(d)$ is obtained and then the diffraction efficiency, which is

$$\frac{I_{scatt}(d)}{I_{in}(0)} = \frac{\int_{-\infty}^{\infty}|\psi_q(d)|^2\, dq}{\int_{-\infty}^{\infty}|\psi_{q''}(0)|^2\, dq''} =$$

$$= \frac{w_p^2\cos\alpha}{\sqrt{2w^2\cos^2\gamma + w_p^2\cos^2\alpha}\cdot\sqrt{2w^2+w_p^2}}\cdot \tag{20}$$

$$\cdot\left[\frac{k_0^2 u_1 \epsilon_a \pi\sin(2\alpha_E)}{2d\left(\beta_{q_0+q_1}\cos^2\alpha_E - \frac{1}{2}(q_0+q_1)\sin 2\alpha_E\right)}\right]^2 \frac{\cos^2\frac{d}{2}\xi}{[\xi^2-p^2]^2} \ . \tag{21}$$

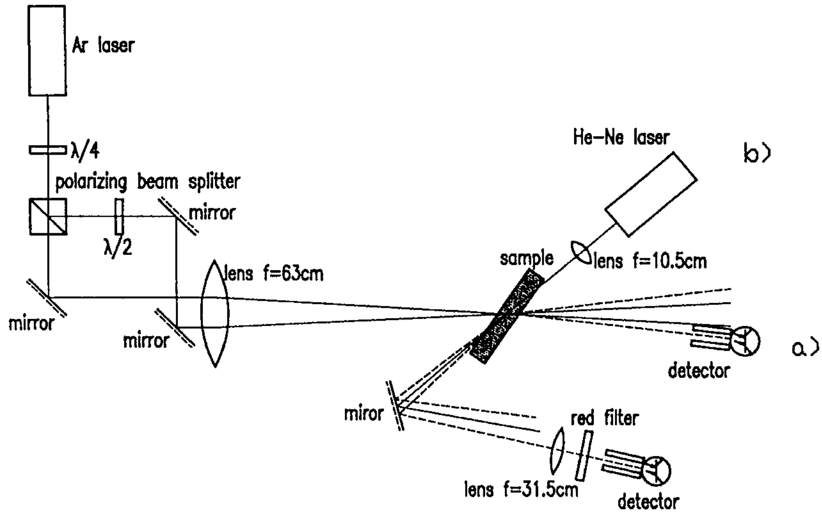

Figure 2: *Experimental arrangement.* a)Self-diffraction. b)The diffraction of the probe beam.

The Gaussian forms of the beams cause two corrections. The first one is the factor (20) and the second is hidden in u_1 in factor (21). As said before, the correction to u_1 due to the finite beam sizes of the pump beams is negligible, when w_p is greater than the cell thickness or the grating period. This is true for most of the usual experimental conditions. In that case the factor (21) corresponds to the diffraction efficiency of a diffraction grating formed by two plane waves with intensities $|E_1|^2$ and $|E_2|^2$. The inclusion of the finite beam sizes leads to the additional factor (20) in the diffraction efficiency. In the case of self-diffraction this factor is close to 1/3. In the case of the diffraction of the probe beam, however, this factor goes to 1, when the beam waist of the probe beam is much smaller than the beam waist of the pump beams. When $w_p < w$, the diffraction efficiency is significantly reduced.

Numerical calculations were also done to obtain the diffraction efficiency from the expression (11) without any further approximation. For usual experimental conditions the results agree with expression (20) - (21) to better than 1 %, so that the approximations leading to (20) - (21) are very reasonable.

EXPERIMENT

The experimental arrangement for the self-diffraction measurements is shown on fig. 2a) (the part without a He-Ne laser). A circularly polarized laser beam from an Ar-laser ($\lambda = 514.5\,\mathrm{nm}$) is split by a polarizing beam splitter. The polarization of one of the beams is then rotated for $90\,^\circ$. The resulting two parallelly polarized beams are focused on the homogeneously aligned nematic cell. The intensity of transmitted and diffracted beams is measured by a detector that consists of a spatial filter and a fotodiode.

Measurements were done on a $50\,\mu$m and a $75\,\mu$m thick cells of the commercial nematic mixture $ZLI1738$ of the Merck Company. The cells were homogeneously aligned. The dependence of the diffraction efficiency on the magnitude of the grating vector **q** was measured. Precautions were taken for the pump power to be low enough, so that no beam self-focusing occurred. The pump beam intensity was 24 mW at the $75\,\mu$m thick cell and 50 mW at the $50\,\mu$m cell.

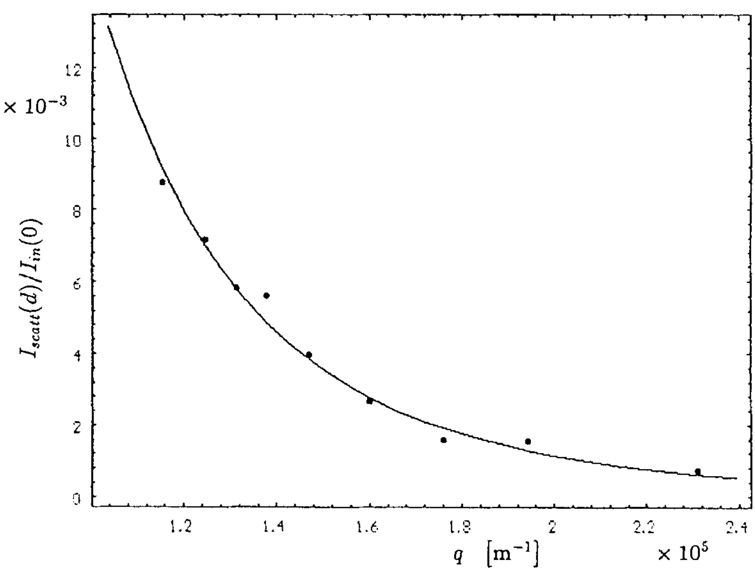

Figure 3: *Self-diffraction: the dependence of the diffraction efficiency on the grating period.* The continuous line is the best fit of the expression (20) - (21) to the experimental data; $d = 50\,\mu$m, $\gamma' = 66°$.

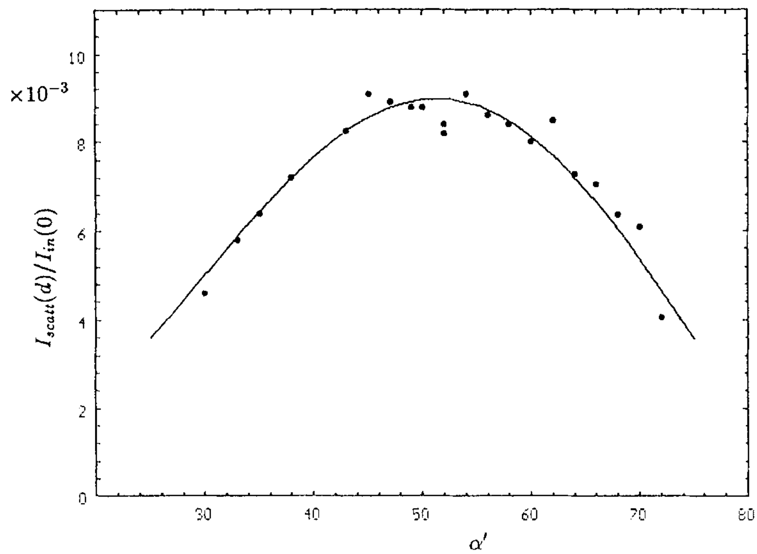

Figure 4: *The diffraction of the probe beam: the dependence of the diffraction efficiency on the angle of incidence of the probe beam.* $d = 50\,\mu$m, $\gamma' = 40°$, $\varphi' = 0.8°$; the perpendicular incidence to the grating is at $\alpha' = 40°$. The continuous line is the best fit of the expression (20) - (21) to the experimental data.

To measure the diffraction of the probe beam a He-Ne laser was used. It was mounted on a rotating arm and the beam was focused on the cell (fig. 2). In front of the detector a lens was put in order to focus the laser beam on the fotodiode. A strong background of the green light due to the Ar laser was cut off by a red filter.

The measurements with the probe beam were done only on the $50\,\mu$m thick cell. The power of the pump beams at the cell was 35 mW and the power of the He-Ne laser was 4 mW. The dependence of the diffraction efficiency on the angle of incidence of the probe beam was measured at several grating periods.

RESULTS AND DISCUSSION

Fig. 3 shows the dependence of the diffraction efficiency on the grating vector for the self-diffraction measurements on the $50\,\mu$m thick cell. The values for K_1 and K_3 calculated from the experimental data obtained on the $50\,\mu$m and $75\,\mu$m thick cells are: $K_1 = 8.3 \times 10^{-12}\,\mathrm{N}\,(1 \pm 0.5)$, $K_3 = 1.2 \times 10^{-11}\,\mathrm{N}(1 \pm 0.3)$. The correction to u_1 due to the finite beam sizes is of the order of 0.1% and is therefore negligible. So the only important correction to the plane wave approximation is the factor (20).

The same is true for the diffraction of the probe beam. The most important correction is again the factor (20). Without this correction the theory predicts the maximum diffraction efficiency at $\alpha' \approx 62°$. The experimental results (fig. 4) show that the maximum is already at $\alpha' \approx 52°$. The theoretical diffraction efficiency corrected by the factor (20) shows good agreement with the experimental results.

From the measurements at a fixed grating period only the factor

$$K_1 \left(\sin^2\gamma + (p/q)^2\right)^2 + K_3 \cos^2\gamma$$

can be determined. From the measurements at different grating periods the values of K_1 and K_3 were determined to be $K_1 = 1.1\times10^{-11}\,\mathrm{N}\,(1\pm0.5)$, $K_3 = 1.2\times10^{-11}\,\mathrm{N}\,(1\pm 0.3)$, which agrees with the results of self-diffraction within the experimental error.

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