Mol Cryst. Liq. Cryst., 1993, Vol. 237, pp. 39–46
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

A New Optical Method for the Measurement of Viscoelastic Coefficient in Nematic Liquid Crystals

L. MARRUCCI, G. ABBATE, S. FERRAIUOLO, P. MADDALENA and E. SANTAMATO

Dipartimento di Scienze Fisiche, Università Federico II, Mostra d'Oltremare, Padiglione 20, I-80125 Napoli, Italy

(Received October 27, 1992)

A new method to measure the ratio between orientational viscosity and the bend elastic constant of nematic liquid crystals is presented. The method is based on the observation of the molecular rotation induced in the sample by the angular momentum transfer from a circularly polarized laser beam Backflow is also considered. PACS numbers: 61 30.Gd, 42 65 -k, 64.70.Md

Keywords: non-linear optics, nematic liquid crystal, viscoelastic coefficient

It is well known that a continuous wave laser beam impinging on a Nematic Liquid Crystal (NLC) sample can induce a change in the orientation of the molecular director $\hat{n}$.[1,2]

In the case of homeotropic alignment and for normal incidence, no reorientation can be induced until the laser beam intensity reaches a characteristic threshold value. This phenomenon is known as Optical Fréedericksz Transition (OFT).[3]

When the beam polarization is circular, the director $\hat{n}$ does not even reach a final steady configuration after the transition. Rather, it goes through a continuous rotation at a fixed tilt angle around the propagation axis $\hat{z}$.[4] Emerging light is then found elliptically polarized with the ellipse major axis rotating at the same speed as $\hat{n}$.

This phenomenon has been explained in terms of a continuous angular momentum transfer from light to NLC molecules. In a quantum picture, each photon, travelling through the film, may suffer a scattering on the NLC film from its initial elicity state to the opposite one, thus imparting to the molecules an angular momentum of $2\hbar$ directed along $\hat{z}$. In view of the analogy with the Brillouin Stimulated Light Scattering, this dynamical phenomenon has been called Self-Induced Stimulated Light Scattering (SISLS).[5] A first quantitative theory for SISLS in nematics has been developed very recently.[6]

In this paper we show how to obtain informations on the viscoelastic coefficient of the NLC, by comparing the period of the laser induced molecular rotation with the theoretical predictions. Backflow effects may affect the results and therefore, unlike in Reference 6, we take them into account.

Let us consider a layer of NLC of thickness L. We use a Cartesian coordinate system having the z axis normal to the plane walls. The director $\hat{\mathbf{n}}$ is specified by its Cartesian components n_i or by the polar angles ϑ and φ. The impinging light beam is assimilated to a monochromatic plane wave, having wavelength λ. We may then drop all spatial derivatives with respect to x and y, and denote $\partial/\partial z$ with a prime.

The dynamics of the director field $\hat{\mathbf{n}}$ is described by the equation

$$\frac{\partial R}{\partial \dot{n}_i} = \frac{\partial}{\partial z}\frac{\partial F}{\partial n_i'} - \frac{\partial F}{\partial n_i} + \beta n_i, \tag{1}$$

where

$$F = \frac{1}{2}K_{11}(\text{div }\hat{\mathbf{n}})^2 + \frac{1}{2}K_{22}(\hat{\mathbf{n}}\cdot\text{rot }\hat{\mathbf{n}})^2 + \frac{1}{2}K_{33}(\hat{\mathbf{n}}\times\text{rot }\hat{\mathbf{n}})^2 - \frac{\varepsilon_a}{16\pi}|\hat{\mathbf{n}}\cdot E|^2 \tag{2}$$

is the director free energy per unit volume in the presence of the optical electric field E (K_{11}, K_{22}, K_{33} are Frank's elastic constants, and $\varepsilon_a = n_e^2 - n_o^2$ is the dielectric anisotropy, n_e and n_o being the NLC extraordinary and ordinary indices, respectively). Moreover,

$$R = \frac{1}{2}\gamma_1(\dot{n}_i - w_{ij}n_j)(\dot{n}_i - w_{ih}n_h) + \gamma_2(\dot{n}_i - w_{ih}n_h)v_{ij}n_j \tag{3}$$

(sum over repeated indices is implied) is the dissipation function in the presence of a nonzero fluid velocity field $\mathbf{v}$ (v_{ij} and w_{ij} are the symmetric and antisymmetric part of the tensor $\partial v_i/\partial x_j$ respectively, γ_1 and γ_2 are the orientational viscosity constants). In Equation (1) β is a Lagrange's multiplier to be chosen in such a way that $\hat{\mathbf{n}}^2(t) = 1$.[7,8] Projecting Equation (1) on the plane normal to $\hat{\mathbf{n}}$, the term βn_i cancels out and we are left with the evolution equation

$$\gamma_1(\dot{n}_i - w_{ik}n_k - n_i n_h n_k w_{hk}) + \gamma_2(n_j v_{ji} - n_i n_j n_h v_{jh}) = G_i, \tag{4}$$

where G_i are the functions of $\hat{\mathbf{n}}$, of its z-derivatives $\hat{\mathbf{n}}'$, $\hat{\mathbf{n}}''$, and of the electromagnetic fields, given by

$$G_i = (\delta_{ij} - n_i n_h)\left(\frac{\partial}{\partial z}\frac{\partial F}{\partial n_h'} - \frac{\partial F}{\partial n_h}\right). \tag{5}$$

Equation (4) is to be solved together with the lights propagation equations, which, in the slow envelope approximation, can be shown[9,10] to reduce to a precession equation for Stokes' parameter vector $\hat{\mathbf{s}} = (s_1, s_2, s_3)$, viz.

$$\hat{\mathbf{s}}' = \frac{2\pi}{\lambda}\Delta n[\vartheta(z)](\cos 2\varphi(z), \sin 2\varphi(z), 0) \times \hat{\mathbf{s}}, \tag{6}$$

where

$$\Delta n(\vartheta) = \frac{n_o n_e}{(n_o^2 \sin^2\vartheta + n_e^2 \cos^2\vartheta)^{1/2}} - n_o. \tag{7}$$

For small angle $\vartheta(z)$, we retain only terms linear in the small quantities n_x, n_y, and $\mathbf{v}'$ in Equation (4), and quadratic in Equation (6). Equation (4) is then reduced to

$$\gamma_1 \dot{n}_i - \frac{\gamma_1 - \gamma_2}{2} v_i' = G_i \qquad (i = x, y) \tag{8}$$

(n_z equation is not necessary, due to the normalization condition). In the same approximation, Navier-Stokes' equation for the fluid velocity $\mathbf{v}$ can be shown to reduce (in the non-inertial limit) to[8]

$$\frac{\partial}{\partial z}\left[\mu_2 \dot{n}_i + \frac{1}{2}(\mu_4 + \mu_2) v_i'\right] = 0 \qquad (i = x, y), \tag{9}$$

where μ_i with $i = 1 \div 6$ are Leslie's viscosity coefficients, lied to the γ_i by the relations

$$\begin{aligned} \gamma_1 &= \mu_3 - \mu_2, \\ \gamma_2 &= \mu_3 + \mu_2. \end{aligned} \tag{10}$$

Integrating Equations (9), and using the conditions $\mathbf{v} = 0$ at boundaries, we can solve for v_i', and find

$$v_i' = -\frac{2\mu_2}{\mu_4 + \mu_5 - \mu_2}\left(\dot{n}_i - \frac{1}{L}\int_0^L \dot{n}_i \, dz\right) \qquad (i = x, y). \tag{11}$$

Substitution in Equation (8) then yields

$$\gamma_1 \dot{n}_i - \frac{2\mu_2^2}{\mu_4 + \mu_5 - \mu_2}\left(\dot{n}_i - \frac{1}{L}\int_0^L \dot{n}_i \, dz\right) = G_i \qquad (i = x, y). \tag{12}$$

For small angle $\vartheta(z)$, and for negligible twist $\varphi'(z)$, the distortion shape is approximately given by the single mode expression

$$n_i(z, t) = A_i(t) \sin\frac{\pi z}{L} \qquad (i = x, y). \tag{13}$$

Substitution of Equation (13) into Equation (12), and "projection" over the first mode (i.e., multiplication by $\sin(\pi z/L)$ and integration over z going from 0 to L) gives

$$\gamma_1^* \dot{A}_i \qquad \frac{2}{L}\int_0^L G_i \sin\frac{\pi z}{L}\,dz \qquad (i \quad x,\ y), \tag{14}$$

with

$$\gamma_1^* = \gamma_1 - \frac{2\mu_2^2}{\mu_4 + \mu_5 - \mu_2}\left(1 - \frac{8}{\pi^2}\right). \tag{15}$$

We notice that the backflow correction to γ_1 given by Equation (15) turns out to be independent of the director motion (rotation or relaxation), provided $\vartheta(z)$ and $\varphi'(z)$ are small.

We can now rewrite Equation (4) with the substitutions $\mathbf{v} \to 0$, $\gamma_1 \to \gamma_1^*$, obtaining

$$\gamma_1^* \dot{n}_i = G_i \qquad (i = x,\ y). \tag{16}$$

As shown in Reference [6], Equations (16) and (6) can be combined to obtain an expression for the rotation angular velocity

$$\Omega = \pi^2 \tilde{I}\left[\frac{s_3(L) - s_3(0)}{\alpha}\right]\frac{K_{33}}{\gamma_1^* L^2}, \tag{17}$$

and for the output light ellipticity

$$s_3(L) = s_3(0)\cos\alpha. \tag{18}$$

In these equations we introduced the phase shift due to birefringence

$$\alpha = \frac{2\pi}{\lambda}\int_0^L \Delta n[\vartheta(z)]\,dz \tag{19}$$

and the dimensionless intensity $\tilde{I} = I/I_{th}$, where

$$I_{th} = \frac{2\pi^2 c n_e^2 K_{33}}{(n_e^2 - n_o^2) n_o L^2} \tag{20}$$

is the threshold intensity value for circularly polarized light.[3]

As shown in Reference 6, α can be expressed as a function of $\tilde{I}$ by inverting the relationship

$$\tilde{I}(\alpha) = \frac{1 - [1 - 2H(\alpha)]^{1/2}}{H(\alpha)}, \tag{21}$$

where $H(\alpha)$ is defined as

$$H(\alpha) = 2\pi^2 \int_0^1 \frac{\sin[\alpha u(\zeta)][1 - u(\zeta)][1 - \cos \alpha u(\zeta) - (1 - \cos \alpha)u(\zeta)]}{\alpha \sin^2 \pi\zeta} d\zeta$$

$$+ \pi^2 \int_0^1 \left\{ \frac{1 - \cos[\alpha u(\zeta)] - (1 - \cos \alpha)u(\zeta)}{\alpha \sin \pi\zeta} \right\}^2 d\zeta, \qquad (22)$$

with

$$u(\zeta) = \zeta - \frac{1}{2\pi} \sin(2\pi\zeta). \qquad (23)$$

By substituting for $\alpha(\tilde{I})$ in Equations (18) and (17), we obtain the final expression for the rotation period $T = |2\pi/\Omega|$

$$T(\tilde{I}) = \frac{2\alpha(\tilde{I})}{\pi\tilde{I}[1 - \cos \alpha(\tilde{I})]} \left(\frac{\gamma_1^* L^2}{K_{33}} \right) = f(\tilde{I}) \left(\frac{\gamma_1^* L^2}{K_{33}} \right). \qquad (24)$$

We notice that the numerical factor $f(\tilde{I})$ in Equation (24) is independent of material constants, film thickness, etc., provided the laser intensity is normalized to the threshold value. $f(\tilde{I})$ is always of the order of one, as shown in Figure 1.

The measure of the rotation period T provides therefore a very simple way to have good estimates of the viscoelastic ratio γ_1^*/K_{33}. The sensitivity of the method is only limited by the precision by which we know the thickness L of the film, and by the correctness of the approximations we made. Moreover, the intensity ratio $\tilde{I} = I/I_{th}$ may be replaced by the laser total power ratio P/P_{th}, which can be determined without knowing the beam spot-size.

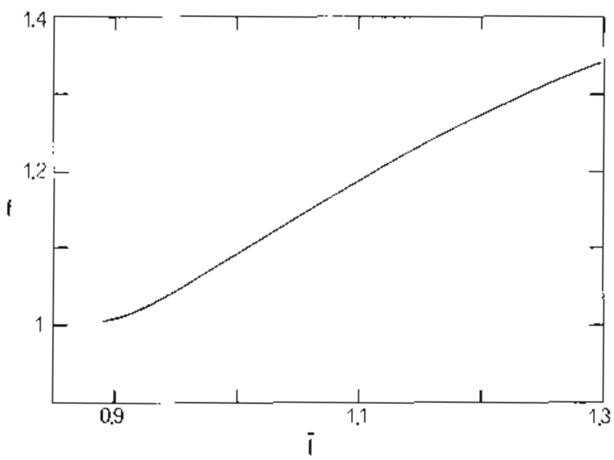

FIGURE 1 The universal function $f(\tilde{I})$. The function extends to the region $\tilde{I} < 1$ because of the hysteresis of the rotational motion.

In our experiment we used a thin film of E7, a NLC mixture from BDH. The film thickness was measured under a microscope to be 120 ± 5 μm (this was achieved by observing the sample from the side, and measuring the distance between the internal edges of the containing glasses by means of a rule in the microscope ocular). The containing glasses were coated with HTAB for homeotropic alignment. The sample was put into a oven having temperature control within 0.1°C. The molecular rotation was induced by circularly polarized Ar^+ laser beam, with $\lambda = 514.5$ nm, focused at normal incidence onto the sample in a Gaussian profile spot having a diameter of about 160 μm. The experimental apparatus is shown in Figure 2.

We checked that the heating due to laser absorption in our sample was negligible, by measuring the ordinary refractive index change in the presence of the pump laser, below the OFT threshold. The index temperature dependence was measured by an interferometric technique.[11,12] The sample worked as a Fabry-Perot, leading to rapid oscillations in the transmitted light intensity as the oven temperature was increased at constant rate. The further heating due to laser absorption should produce a shift in the transmission versus temperature curve. Within the sensitivity of the method (±0.1°C), no shift was actually observed.

For each temperature we measured also the threshold power P_{th}, which was found strongly dependent on temperature. The rotation periods for different values of ratio $\bar{I} = P/P_{th}$ were then measured, by looking at the rotation of the light polarization ellipse beyond the sample. By a complete ellipsometric technique based on an heterodine Mach-Zender interferometer,[13] we checked that ellipticity was almost constant during the rotation. A fixed polarizer has then been put beyond the sample, to collect only a linear component of the emerging light on the photodetector. As expected, the signal during the rotation showed a uniform oscillation at a frequency 2Ω, whose amplitude depended on the light ellipticity (a little difference between two consecutive oscillations was actually observed, which we ascribed to small amount of nutaton superimposed to the rotation of $\hat{n}$).

The observed periods were fitted with the theoretical curve given by Equation (24), using γ_1^*/K_{33} as a fitting parameter. The best fits in the temperature range 22°C ÷ 50°C are shown in Figure 3.

Since the elastic constants are proportional to S^2, S being the order parameter, the predicted dependence of γ_1^*/K_{33} on temperature T in E7 is approximately given by[14]

$$\frac{\gamma_1^*}{K_{33}} \sim \frac{e^{E/kT}}{S}, \tag{25}$$

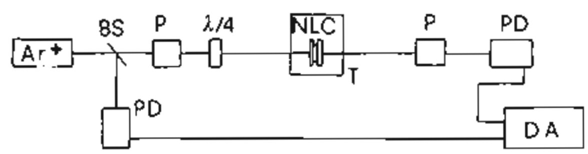

FIGURE 2 Experimental apparatus. NLC–sample; T–thermostatic oven; Ar^+–laser, BS–beam splitter; P–polarizer; λ/4–birefringent plate; PD–photodiode; DA–data acquisition system.

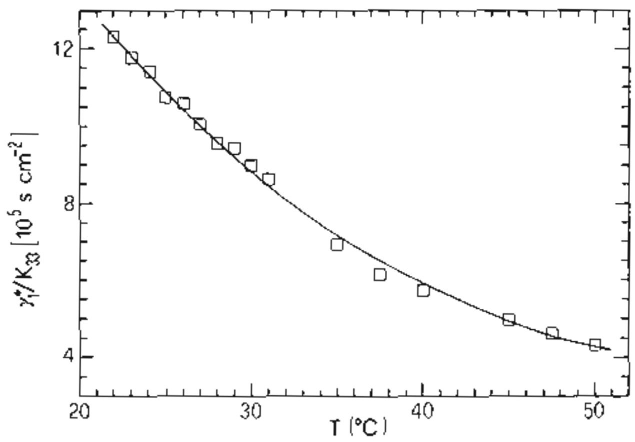

FIGURE 3 Ratio γ_1^*/K_{33} versus temperature in E7. Points are from best fit with theory. Solid line is a polynomial fit on the points.

where E is an activation energy and k is the Boltzmann constant. We may assume $S = (1 - T/T_{ni})^{\beta}$ where $T_{ni} \simeq 65°C$ is an effective temperature for the nematic-isotropic phase transition and β is an exponent which typically ranges from 0.1 to 0.3. Equation (25) then predicts a monotonic almost linear decrease of γ_1^*/K_{33}, up to a temperature close to T_{ni} where it saturates or eventually starts increasing. This qualitative behaviour is evident in Figure 3.

A quantitative comparison with the experimental data on γ_1/K_{11} in E7 obtained by optical response time measurement,[15] after correction by the factor $K_{11}/K_{33} = 0.6$,[16] shows an agreement better than 10%.

Acknowledgment

This work was supported by Ministero dell'Università e della Ricerca Scientifica e Tecnologica, and by Consiglio Nazionale delle Ricerche, Italy.

References

1. A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, N. I. Sobolev and L. Csilag, *Pis'ma Zh. Eksp. Teor. Fiz.*, **32**, 170 (1980), (*JETP Lett.*, **32**, 158).
2. S. D. Durbin, S. M. Arakelian and Y. R. Shen, *Phys. Rev. Lett.*, **47**, 1411 (1981).
3. B. Ya. Zel'dovich, N. V. Tabiryan and Yu. S. Chilingaryan, *Zh. Eksp. Teor. Fiz.*, **81**, 72 (1981), (*Sov. Phy. JETP*, **54**(1), 32); B. Ya. Zel'dovich and N. V. Tabiryan, *Zh. Eksp. Teor. Fiz.*, **82**, 1126 (1982), (*Sov. Phys. JETP*, **55**(4), 656).
4. E. Santamato, B. Daino, M. Romagnoli, M. Settembre and Y. R. Shen, *Phys. Rev. Lett.*, **57**, 2423 (1986).
5. E. Santamato, M. Romagnoli, M. Settembre, B. Daino and Y. R. Shen, *Phys. Rev. Lett.*, **61**, 113 (1988).
6. L. Marrucci, G. Abbate, S. Ferraiuolo, P. Maddalena and E. Santamato, *Phys. Rev. A*, **46**, 4859 (1992).
7. P. G. de Gennes, *The Physics of Liquid Crystals*, Oxford Univ. Press, Oxford 1974.
8. S. Chandrasekhar, *Liquid Crystals*, Cambridge Univ. Press, 1977.
9. H. Kubo, R. Nagata, *J. Opt. Soc. Am.*, **73**, 1719 (1983).

10. E. Santamato and Y. R. Shen, *J. Opt. Soc. Am. A*, **4**, 356 (1987).
11. G. Abbate, A. Attanasio, U. Bernini, E. Ragozzino and F. Somma, *J. Phys. D: Appl. Phys.*, **9**, 1945 (1976).
12. G. Abbate, U. Bernini, G. Brescia and E. Ragozzino, *Optics and Laser Technology*, 97 (April 1981).
13. R. Calvani, R. Caponi and F. Cisternino, *Opt. Commun.*, **54**, 63 (1985).
14. V. V. Belyaev, S. A. Ivanov and M. F. Gorebenkin, *Sov. Phys. Cristallogr.*, **30**, 674 (1985)
15. Shin-Tson Wu, A. M. Lackner and U. Efron, *App. Opt.*, **26**, 3441 (1987).
16. H. L. Ong, *Phys. Rev. A*, **28**, 2393 (1983).