will be reported.

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New Method for Measuring the Twist Elastic Constant K_{22}/χ_a and the Shear Viscosity γ_1/χ_a for Nematics

P. E. Cladis

Physique des Solides, Université Paris-Sud, 91-Orsay, France*

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By monitoring the change in the interference figure for an oriented nematic (optic axis perpendicular to the incident light cone) as twist deformation is induced by a magnetic field (Freedericksz transition), one is led to a simple and direct method for measuring K_{22}/χ_a and γ_1/χ_a. An analytic expression is obtained for the effect of twist on the interference figure and is verified experimentally.

The interference figure[1] for an oriented nematic (held between rubbed[2] parallel glass plates), with the director (optic axis) in a plane perpendicular to the axis of the incident light cone, is composed of four more or less equilateral hyperbolas. In 1911, Mauguin[3] demonstrated that twisting the top plate with respect to the bottom by an angle α resulted in a simple rotation of the hyperbolas (about an axis perpendicular to the figure) through an angle $\delta = \frac{1}{2}\alpha$. More recently, de Gennes[4] has remarked (see Appendix) that for any twisted nematic the interference figure would, to a first approximation, rotate by an amount δ, given by

$$\tan 2\delta = \langle \sin 2\theta \rangle / \langle \cos 2\theta \rangle, \quad (1)$$

where $\theta(z)$ is the twist angle of the director and $z = \pm \frac{1}{2}d$ defines the glass-nematic interfaces (inset, Fig. 1). $\sin 2\theta$ and $\cos 2\theta$ are to be averaged over the sample thickness d. Thus, if twist is induced by means of a magnetic field applied in the plane of the plates, but perpendicular to the direction of rubbing (Freedericksz transition) so that $\theta(\pm \frac{1}{2}d) = 0$, but $\theta(z = 0) = \theta_m \neq 0$, δ will not be zero and such a deformation would be observable even for relatively small θ_m. It is evident that, because of the boundary conditions and the adiabatic theorm, such a deformation may not be observed by microscopy. Nor can it be observed by monitoring the dielectric constant (or thermal conductivity or any other anisotropic property of a uniaxial nematic), since the director remains in the same plane as the glass plates in both the twisted and untwisted configurations. Consequently, Eq. (1) represents a simple and new method for quantitatively observing twist deformations in uniaxial liquid crystals.

The purpose of this note is to verify Eq. (1) for the Freedericksz transition and to show that the existence of this effect leads to a new and direct method for detecting this transition and hence for measuring K_{22}/χ_a and γ_1/χ_a, where γ_1 is the shear viscosity, K_{22} the Frank elastic constant of twist, and $\chi_a = \chi_\parallel - \chi_\perp$ is the diamagnetic anisotropy. It is also shown that the average twist deformation may be quantitatively measured as a function of applied magnetic field.

The critical field H_c for the Freedericksz transition for twist is given by[5]

$$H_c = (\pi/d)(K_{22}/\chi_a)^{1/2}. \quad (2)$$

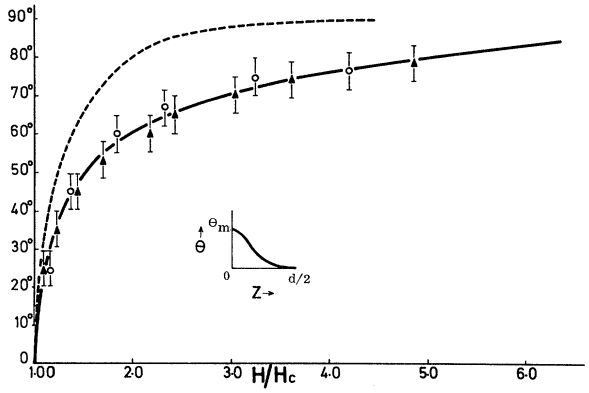

FIG. 1. θ_m, the maximum twist angle for the director, shown as a function of reduced field H/H_c (dashed curve). Solid curve, rotation angle of the interference figure [Eq. (4)]. Experimental points are for MBBA at room temperature.

When $H \leq H_c$, the nematic remains undeformed, i.e., $\theta(z) = 0$. For $H \geq H_c$, it is assumed that the nematic is strongly anchored to the glass plates so that $\theta(\pm \frac{1}{2}d) = 0$, but the interior of the sample may deform in a twist mode so that the maximum twist angle θ_m occurs in the middle of the sample ($z = 0$) and is given implicitly by[5]

$$F(\tfrac{1}{2}\pi, \sin\theta_m) = \tfrac{1}{2}\pi H/H_c, \tag{3}$$

where $F(\frac{1}{2}\pi, \sin\theta_m) = K(\theta_m)$ is the complete elliptic integral of the first kind. Equation (3) is shown as the dashed curve in Fig. 1. To compute Eq. (1) then for this case, it is assumed that (a) the deformation is given by the Frank-Zocher-Oseen theory,[5] and (b) strong anchoring prevails for all values of the field. Under these conditions

$$\tan 2\delta = \frac{2\sin\theta_m}{2E(\frac{1}{2}\pi, \sin\theta_m) - F(\frac{1}{2}\pi, \sin\theta_m)}, \tag{4}$$

where $E(\frac{1}{2}\pi, \sin\theta_m)$ is the complete elliptic integral of the second kind. Using Eq. (3), we can plot δ as a function of reduced coordinates H/H_c, shown in Fig. 1 as the solid curve.

The experimental points in Fig. 1 are for *p-n* methoxybenzilidene butylaniline (MBBA) at room temperature and are seen to agree quite well with the theory. Since the theoretical plot can be with reduced coordinates H/H_c, merely setting the field H at some value greater than H_c will result in a δ which will uniquely define H_c—as precisely as one can determine H and δ. This turns out to be useful since for $H \sim H_c$, δ approaches equilibrium very slowly. In fact for the points for which $H/H_c \cong 1.1$, the system required about 15 min to stabilize, whereas for $H \gg H_c$, δ reached equilibrium relatively quickly.

The sample thickness d was determined by means of Mylar spacers. Corrections to the Mylar-spacer thickness were made by the technique of focusing a microscope on the top and bottom inner glass surfaces (before filling) and reading directly the difference (in micrometers) between the two settings on the calibrated focusing device of the microscope.

Knowing H_c and d, K_{22}/χ_a was found [from Eq. (2)] to be 2.88 ± 0.03 (cgs). This value agrees well with that found by unwinding the pitch of cholesterized MBBA by means of a magnetic field.[6] However, this new method does not require the nematic to be doped.

If for time $t < 0$, $H > H_c$, but $\sin\delta \sim \delta$ (small deformation); at $t = 0$, the field is suddenly released, and $\delta(t)$ is observed to relax to zero with a single characteristic relaxation time τ. When $H \gg H_c$ initially so that one is no longer in the small deformation regime, a unique relaxation time is not observed, i.e., a plot of $\ln\delta$ versus t is not linear. For small deformations, $\theta(z) = \theta_m \cos(\pi z/d)$, the first Fourier component for a general expansion of $\theta(z)$. Consequently, the relaxation time for δ decaying to zero is given by[7]

$$\tau = (\gamma_1/\chi_a) H_c^{-2}. \tag{5}$$

Plotting τ versus H_c^{-2} for various thickness results in a straight line whose slope is given by γ_1/χ_a.

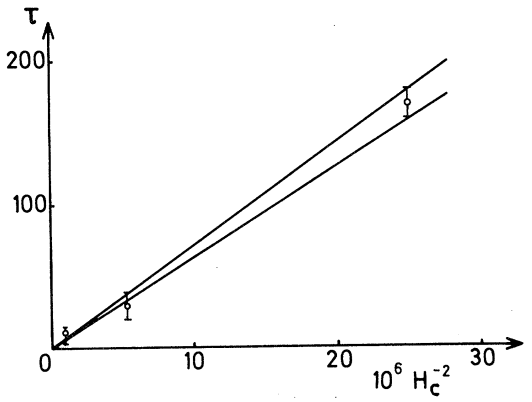

FIG. 2. The relaxation time τ (in seconds) versus H_c^{-2} (in G^{-2}) for MBBA. According to Eq. (5), the slope is given by γ_1/χ_a. $\delta(t = 0) = \frac{1}{4}\pi$.

Preliminary results for MBBA are shown in Fig. 2. γ_1/χ_a was found to be $\sim 7 \times 10^6$ (cgs) so that using $\chi_a = 1.16 \times 10^{-7}$ (cgs),[8] $\gamma_1 \sim 0.8$ sec^{-1}. Although this method is direct, its accuracy is limited by the precision with which $\delta(t)$ can be determined for small angles.

In conclusion, with the assumption of a strong anchoring condition [$\theta(\pm \frac{1}{2}d) = 0$, $H > H_c$] and the Frank-Zocher-Oseen theory,[5] Eq. (1) has been verified for MBBA at room temperature. It thus leads to a new and simple method for measuring K_{22}/χ_a and γ_1/χ_a.

I would like to thank Professor P. G. de Gennes for the material in the Appendix and for his comments on the manuscript.

Appendix: Derivation of Eq. (1).—(a) For a uniaxial monocrystal, define in the plane of the glass plates a local coordinate system (ξ, η), such that ξ lies along the optical axis and is perpendicular to it. Let the direction normal to the glass plates be the z direction. Then, for an incident wave of frequency ω, the component of the ordinary wave vector in the z direction, k_{oz}, is

given by

$$k_{oz}^2 = (\omega n_o/c)^2 - k_\xi^2 - k_\eta^2. \tag{A1}$$

For the other wave, we have

$$k_{ez}^2 = (\omega n_e/c)^2 - k_\eta^2 - k_\xi^2(n_e/n_o)^2. \tag{A2}$$

The total change in phase for a sample of thickness d is

$$\varphi = d(k_{ez} - k_{oz}) = \varphi_0\left[1 + \left(\frac{c}{\omega}\right)^2 \frac{1}{2n_o}\left(\frac{k_\eta^2}{n_e} - \frac{k_\xi^2}{n_o}\right)\right], \tag{A3}$$

assuming k_ξ and $k_\eta \ll (\omega/c)\varphi_0 = (n_e - n_o)d$ is the phase change in the center of the field. Equation (A3) shows that lines of constant phase in k space are hyperbolas.

(b) For a twisted nematic, the electromagnetic equations will have solutions of the form $\exp(-i\omega t) \times \exp(k_x x + k_y y) f(z)$, where x is the direction of polishing of the glass plates. We may still define

$$k_\xi = k_x \cos\theta + k_y \sin\theta, \quad k_\eta = -k_x \sin\theta + k_y \cos\theta,$$

where $\theta(z)$ is the twist angle. We assume that $\theta(z)$ varies slowly in an optical wavelength. Then, we may apply an adiabatic approximation similar to that defined by Mauguin[3] and say that $f(z) = \exp(ik_z z)$, where k_z is defined locally by Eqs. (A1) or (A2).

The change in phase, $\Delta\varphi$, from a layer Δz thick is found from Eq. (A3):

$$\Delta\varphi = \left(\frac{\omega}{c}\right)(n_e - n_o)\Delta z \times\left[1 + \left(\frac{c}{\omega}\right)^2 \frac{1}{4n_o^2 n_e}\{(n_e - n_o)(k_x^2 + k_y^2) - (n_e + n_o)[(k_x^2 - k_y^2)\cos 2\theta + 2k_x k_y \sin 2\theta]\}\right]. \tag{A4}$$

Integrating Eq. (A4) over the sample thickness,

$$\varphi = \varphi_0\left[1 + \left(\frac{c}{\omega}\right)^2 \frac{1}{4n_o^2 n_e}\{(n_o - n_e)(k_x^2 + k_y^2) - (n_e + n_o)[(k_x^2 - k_y^2)\langle\cos 2\theta\rangle + 2k_x k_y \langle\sin 2\theta\rangle]\}\right], \tag{A5}$$

where the angular brackets mean the averages of $\cos 2\theta$ and $\sin 2\theta$. Thus Eq. (A5) predicts lines of constant phase in k space to be hyperbolas rotated by an amount δ given by Eq. (1). This formula is of interest mainly when the averages ($\langle\cos 2\theta\rangle$ or $\langle\sin 2\theta\rangle$) are not too small. Should they become small (e.g., in a sample twisted several turns), Eq. (A5) shows that the pattern of hyperbolas expands and the effect may not be observed.

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*Laboratory associated with the Centre National de la Recherche Scientifique.

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