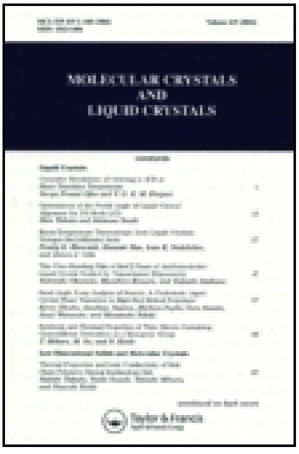

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Circular Shearing Flow of Homeotropic Nematic Liquid Crystals

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Circular Shearing Flow of Homeotropic Nematic Liquid Crystals

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The equation of motion of director in a homeotropic liquid crystal cell with circular plates rotating against each other is derived with cylindrical coordinates. The optical pattern observed under crossed polarizers with monochromatic light is discussed. Numerical integrations for the case of MBBA have been carried out and the result is compared with the experimental observation reported by Wahl and Fischer in 1973, Mol. Cryst. Liq. Cryst., *22*, 359. Instead of the linear relationship between $(m/l)/(lv)^2$ and $(lv)^2$ given by Wahl and Fischer, we suggest a parabolic relationship between $(lv)^2/(m/l)$ and (m/l), where m is the order of the dark ring, l is the thickness of the cell, and v is the velocity of flow of the liquid crystal at a distance r from the center of the cell.

1. INTRODUCTION

Wahl and Fischer[1,2] investigated the optical pattern in a homeotropic nematic liquid crystal cell of p-methoxy benzilidene p-*n*-butylaniline (MBBA) with circular plates rotating against each other. They analysed their experimental results by using the formulae given by Leslie[3] for simple shear flow. He, Shu and Lin[4] analysed the same problem by using the equation of motion of director for nematic liquid crystals flowing in the azimuthal direction with axial symmetry derived from the Erickson-Leslie theory.[3] However, in Equation (2.3) of Reference 4, the curvature force f_i is derived from the Frank free energy density expression[5] with the term $k_{33}(n \cdot \nabla n)^2$ while the curvature stress tensor Π_{ij} is derived from the expression with the term $k_{33}(n \times \nabla \times n)^2$. This inconsistency makes the equation of motion of director used by He, Shu and Lin in error. In this paper we use cylindrical coordinates

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(r, ϕ, x_3) in deriving the equation of conservation of momentum and the equation of motion of director from the general expression of the Frank free energy density g in the form

$$g = \frac{1}{2}[k_{11}(\nabla \cdot n)^2 + (k_{22} - k_{33})(n \cdot \nabla \times n)^2 + k_{33}(\nabla \times n)^2]$$

$$= \frac{1}{2}[k_{11}n^i{}_{,j}\delta^j_i\, n_p{}^{,q}\delta^p_q + (k_{22} - k_{33})(\epsilon_{pqs}n^p n^{s,q})(\epsilon^{lji}n_l n_{i,j}) \qquad (1)$$

$$+ k_{33}\epsilon^{lji}\epsilon_{lpq}n^{q,p}n_{i,j}],$$

where the contravariant components of the fundamental metric tensor g^{ij} and the permutation tensor ϵ^{ijk} are given by

$$g^{rr} = g^{33} = 1 \qquad g^{\phi\phi} = 1/r^2, \qquad (2)$$

and

$$\epsilon^{r\phi 3} = \epsilon^{\phi 3r} = \epsilon^{3r\phi} = 1/r, \qquad \epsilon^{r3\phi} = \epsilon^{\phi r3} = \epsilon^{3\phi r} = -1/r, \qquad (2)$$

respectively.

As a first approximation, the equation of conservation of momentum leads to the result that the torsional shear is uniform, an assumption used in Reference 4. Under the assumption that the angle θ between the director n and the x_3-axis varies slowly with r, using computer, we made a numerical calculation of the functional relationship between θ_M and lv for MBBA where θ_M is the maximum angle of deformation of the director on the central layer of the liquid crystal cell, l is the thickness of the cell and $v = \omega_o r$ is the velocity of flow of the liquid crystal at r, ω_o is the relative angular velocity of the two plates of the cell. The relationship between θ_M and m/l, where m is the order of the dark ring beginning with $m = 0$ at the center, is also calculated numerically. From the above relations we obtain the m/l versus lv curve and the $(m/l)/(lv)^2$ versus $(lv)^2$ curve. Approximate expressions for small θ_M are also given. Our calculations are compared with the results of Wahl and Fischer.[1,2]

2. EQUATIONS OF MOTION

We take the center of the liquid crystal cell as the origin of our coordinate system (Figure 1). The director n is assumed to be in the $(\hat{\phi}, \hat{x}_3)$ plane with contravariant components

$$n^r = 0, \qquad n^\phi = \sin\theta/r, \qquad n^3 = \cos\theta, \qquad (4)$$

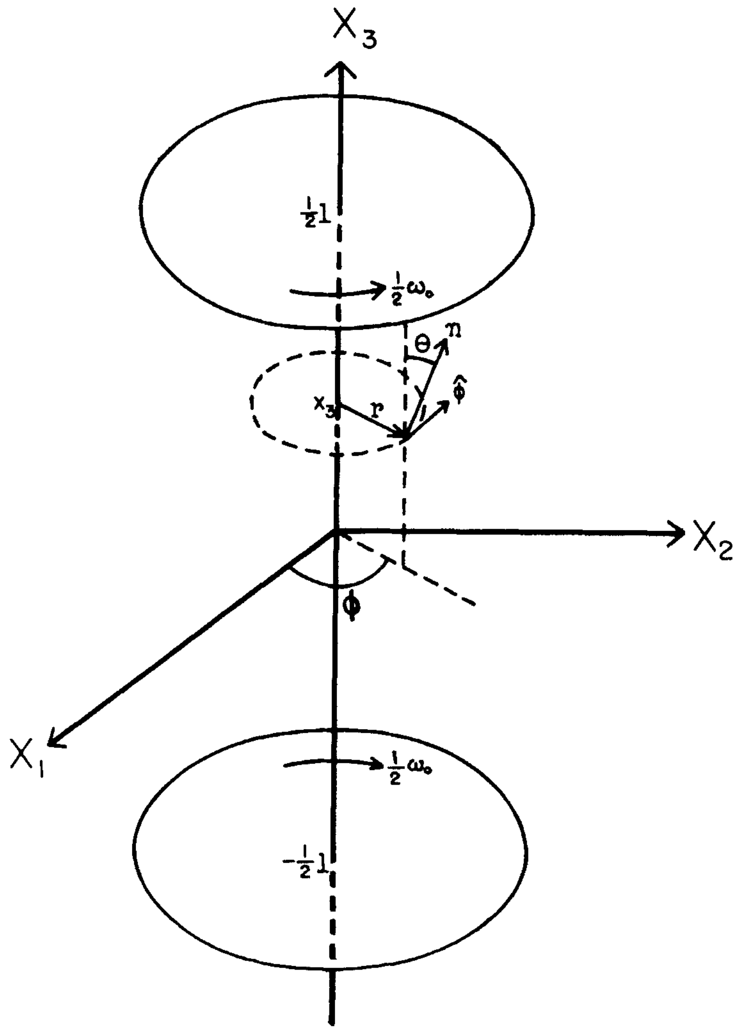

FIGURE 1 Geometry of the system.

where $\theta = \theta(r, x_3)$. The contravariant components of the velocity vector v are given by

$$v^r = \frac{dr}{dt} = 0, \qquad v^\phi = \frac{d\phi}{dt} = \omega(x_3), \qquad v_3 = \frac{dx_3}{dt} = 0, \tag{5}$$

where d/dt is the material derivative operator. The equation of conservation of momentum and the equation of motion of director are

$$\rho\frac{\delta}{\delta t}v^i + p^{,i} - (\Pi^{kj}n_k^{;i})_{,j} + t^{ij}_{,j} = 0, \tag{6}$$

and

$$I\frac{\delta}{\delta t}\left(\frac{\delta n^i}{\delta t}\right) + \Pi^{ij}_{,j} + f^i + f'^i = \gamma n^i \tag{7}$$

respectively, where ρ is the density, I is the moment of inertia per unit volume, p is the pressure, γ is the Lagrangian unknown multiplier, $\delta/\delta t$ is the intrinsic derivative operator and

$$\Pi^{ij} = -\frac{\partial g}{\partial n_{i,j}} = -k_{11}n^l_{,l}g^{ij} - (k_{22} - k_{33})(\epsilon_{pqs}n^p n^{s,q})\,\epsilon^{lji}n_l$$

$$- k_{33}\epsilon^{lji}\epsilon_{lpq}n^{q,p},$$

$$f^i = \frac{\partial g}{\partial n_i} = (k_{22} - k_{33})(\epsilon_{pqs}n^p n^{s,q})(\epsilon^{ijk}n_{k,j}),$$

$$t^{ij} = -\alpha_1(n_k n_p d^{kp})n^i n^j - \alpha_2 N^i n^j - \alpha_3 n^i N^j - \alpha_4 d^{ij}$$

$$- \alpha_5 d^{ik}n_k n^j - \alpha_6 n^i n_k d^{kj}, \tag{8}$$

$$f'^i = \gamma_1 N^i + \gamma_2 d^{ij}n_j,$$

$$N^i = \frac{\delta n^i}{\delta t} - \omega^{ij}n_j,$$

$$d^{ij} = \frac{1}{2}(v^{i,j} + v^{j,i}),$$

$$\omega^{ij} = \frac{1}{2}(v^{i,j} - v^{j,i}),$$

γ_1, γ_2 and the α's are the various viscosity coefficients.

Firstly, let us consider the equation of conservation of momentum. The r component of Equation (6) simply determines the pressure $p = p(r)$ in the liquid crystal cell. The elastic constants k_{11}, k_{22}, k_{33} are of the order of 10^{-6} dynes and the viscosity coefficients $\alpha_1, \ldots, \alpha_6$ are of the order of one poise. Therefore we may neglect the terms involving the elastic constants. In case where θ varies slowly with r we may also neglect the $\partial\theta/\partial r$ term. For steady state and small θ, as first approximation, the φ component and the x_3 component of Equation (6) lead to

$$d\omega/dx_3 = \omega_o/l, \tag{9}$$

where ω_o is the relative angular velocity of the two plates and l is the thickness of the cell. Equation (9) is simply the assumption used by He, Shu and Lin.[4]

Next, let us consider the equation of motion of director, Equation (7). As usual we neglect the I term. For steady state, the three components of Equation (7) are given by

$$k_{11}\frac{\partial}{\partial r}\left(\sin\theta\frac{\partial\theta}{\partial x_3}\right) - k_{22}\frac{\partial}{\partial x_3}\left[\sin\theta\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right)\right] + \frac{k_{33}}{r}\frac{\partial}{\partial x_3}(\sin^2\theta\cos\theta)$$

$$- (k_{22} - k_{33})\cos\theta\frac{\partial\theta}{\partial x_3}\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right) - \gamma_1\omega\sin\theta = 0,$$

$$-\frac{k_{22}}{r}\frac{\partial}{\partial r}\left[\cos\theta\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right)\right] - \frac{k_{33}}{r}\left[\frac{\partial}{\partial x_3}\left(\cos\theta\frac{\partial\theta}{\partial x_3}\right) + \frac{\partial}{\partial r}\left(\frac{\sin^3\theta}{r}\right)\right]$$

$$+ (k_{22} - k_{33})\frac{\sin\theta}{r}\frac{\partial\theta}{\partial r}\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right) - \frac{1}{2}(\gamma_1 - \gamma_2)\cos\theta\frac{d\omega}{dx_3} = \gamma\frac{\sin\theta}{r},$$

$$k_{11}\frac{\partial}{\partial x_3}\left(\sin\theta\frac{\partial\theta}{\partial x_3}\right) + \frac{k_{22}}{r}\frac{\partial}{\partial r}\left[r\sin\theta\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right)\right]$$

$$- \frac{k_{33}}{r}\frac{\partial}{\partial r}(\sin^2\theta\cos\theta) + (k_{22} - k_{33})\cdot$$

$$\left(\cos\theta\frac{\partial\theta}{\partial r} + \frac{\sin\theta}{r}\right)\cdot\left(\frac{\partial\theta}{\partial r} + \frac{\sin\theta\cos\theta}{r}\right) - \frac{1}{2}(\gamma_1 + \gamma_2)r\sin\theta\frac{d\omega}{dx_3} = \gamma\cos\theta,$$

respectively. Figure 1 shows that a change of the direction of ω means a change of sign of the angle θ. Thus the first equation is invariant under the change of direction of ω. The second and the third equation, upon eliminating the unknown multiplier γ, lead to the equation of motion of director

$$(k_{11}\sin^2\theta + k_{33}\cos^2\theta)\frac{\partial^2\theta}{\partial x_3^2} + (k_{11} - k_{33})\sin\theta\cos\theta\left(\frac{\partial\theta}{\partial x_3}\right)^2 + k_{22}\frac{\partial^2\theta}{\partial r^2}$$

$$+ \frac{k_{22}}{r}\frac{\partial\theta}{\partial r} - (k_{22}\cos^2\theta + k_{33}\sin^2\theta)\frac{\sin\theta\cos\theta}{r^2} + (k_{22} - k_{33})\frac{\sin^3\theta\cos\theta}{r^2} \tag{10}$$

$$+ \frac{1}{2}(\gamma_1 - \gamma_2\cos^2\theta)r\frac{d\omega}{dx_3} = 0,$$

where Equation (9) gives that $d\omega/dx_3 = \omega_o/l$. Instead of Equation (2.3) of Reference 4, Equation (10) is the correct form of the equation of motion of director. Obviously, the stationary state solution $\theta = 0$ is a solution of both the first equation and Equation (10).

In the experiments of Wahl and Fischer,[1,2] ω_o is of the order of 10^{-4} sec^{-1}, l is of the order of 10^{-2} cm, and the radius of the first dark ring is of the order of 10^{-1} cm. Even for the first dark ring, $\gamma_{1(2)}r\omega_o/l$ is of the order of 10^{-3}, while $k_{22(33)}/r^2$ is of the order of 10^{-4}. Thus if we assume that θ varies slowly within r, we may neglect the $\partial^2\theta/\partial r^2$, $\partial\theta/\partial r$ and the $1/r^2$ terms in Equation (10) and obtain

$$(k_{11}\sin^2\theta + k_{33}\cos^2\theta)\frac{\partial^2\theta}{\partial x_3^2} + (k_{11} - k_{33})\sin\theta\cos\theta\left(\frac{\partial\theta}{\partial x_3}\right)^2 + (1/2l)(\gamma_1 - \gamma_2\cos^2\theta)r\omega_o = 0. \quad (11)$$

With the boundary conditions

$$\theta = 0 \qquad \text{at } x_3 = \pm 1/2,$$

$$\theta = 0 \qquad \text{at } r = o,$$

$$\theta = \theta_M, \ \frac{\partial\theta}{\partial x_3} = 0 \qquad \text{at } x_3 = 0,$$

the first integral of Equation (11) is

$$(k_{11}\sin^2\theta + k_{33}\cos^2\theta)\left(\frac{\partial\theta}{\partial x_3}\right)^2 = (r\omega_o/l)\left[\gamma_1(\theta_M - \theta) - \frac{1}{2}\gamma_2(\sin 2\theta_M - \sin 2\theta)\right]. \quad (12)$$

Since θ is a function of r and x_3, in order to find a solution of Equation (12) we have to make further approximations. For θ varies slowly with r, let us neglect the variation of r with respect to θ in Equation (12). The integration of Equation (12) then determines the value of θ_M at r:

$$\left(\frac{\gamma_1 l r\omega_o}{4k_{33}}\right)^{1/2} = \int_o^{\theta_M}\left[\frac{1 - P\sin^2\theta}{\theta_M - \theta - (\gamma_2/2\gamma_1)(\sin 2\theta_M - \sin 2\theta)}\right]^{1/2} d\theta, \quad (13)$$

where

$$P = (k_{33} - k_{11})/k_{33}. \quad (14)$$

Since the integrand of Equation (13) must be real, it is necessary that

$$\theta^M - \theta - (\gamma_2/2\gamma_1)(\sin 2\theta_M - \sin 2\theta) \geq 0. \quad (15)$$

For $\frac{1}{2}\pi \geq \theta_M - \theta \geq 0$, Equation (15) shows that there is a limiting value θ_{M_o} for θ_M determined by

$$\theta_{M_o} = \frac{1}{2}(\pi - \cos^{-1}|\gamma_1/\gamma_2|). \tag{16}$$

Therefore θ_M varies continuously and asymptotically from zero at the center of the cell to θ_{M_o} at very large distances from the center.

For small values of θ_M, if we expand the integrand of Equation (13) in the form of $(\theta_M - \theta)^{-1/2}$ times a power series of θ, we find that

$$\begin{aligned}\gamma_1\frac{lr\omega_0}{4k_{33}} &= \frac{\gamma_1}{4k_{33}}lv \\ &= \frac{4}{M}\theta_M\Bigg[1 + \left(\frac{11}{5}N - \frac{8}{15}P\right)\theta_M^2 - \left(\frac{193}{315}N - \frac{128}{945}P - \frac{2704}{525}N^2\right. \\ &\left. + \frac{16}{525}P^2 + \frac{2024}{1575}PN\right)\theta_M^4 + \left(\frac{3238}{45045}N - \frac{2048}{135135}P - \frac{665552}{225225}N^2\right. \\ &+ \frac{41984}{2027025}P^2 + \frac{159736}{225225}PN + \frac{189776}{15015}N^3 - \frac{10496}{675675}P^3 \\ &\left. - \frac{43984}{675675}P^2N - \frac{16136}{5005}PN^2\right)\theta_M^6\Bigg],\end{aligned} \tag{17}$$

where

$$M = (\gamma_1 - \gamma_2)/\gamma_1, \qquad N = -2\gamma_2/3(\gamma_1 - \gamma_2). \tag{18}$$

3. OPTICAL PATTERNS

Between crossed polarizers and with monochromatic light of wave length λ_o normal to the liquid crystal layer, one observes concentric dark and light rings.[1,2] The mth dark ring occurs at[4]

$$\begin{aligned} m/l &= (n_o/2\lambda_o)\int_{-1}^{1}[(1 - \alpha\sin^2\theta)^{-1/2} - 1]\,dz, \\ \alpha &= 1 - n_o^2/n_e^2, \\ z &= 2x_3/l,\end{aligned} \tag{19}$$

where n_o and n_e are the ordinary and extraordinary refractive indices of the liquid crystal respectively. Using Equation (12) we have that

$$m/l = (n_o\alpha/\lambda_o)(\gamma_1 lv/4k_{33})^{-1/2}\int_0^{\theta_M}[(1-\alpha\sin^2\theta)^{-1/2}-1]\cdot \tag{20}$$

$$(1-P\sin^2\theta)^{1/2}[\theta_M-\theta-(\gamma_2/2\gamma_1)(\sin 2\theta_M-\sin 2\theta)]^{-1/2}\,d\theta,$$

where again we have neglected the variation of r with respect to θ. Expansion of $[(1-\alpha\sin^2\theta)^{-1/2}-1]$ into power series of θ up to the θ^6 term gives that

$$[(1-\alpha\sin^2\theta)^{-1/2}-1]=\frac{1}{2}n_o\left[\theta^2-\left(\frac{1}{3}-\frac{3}{4}\alpha\right)\theta^4+\left(\frac{2}{45}-\frac{1}{2}\alpha-\frac{5}{8}\alpha^2\right)\theta^6\right]. \tag{21}$$

For small values of θ_M, with the same way of expansion in deriving Equation (17), and with Equation (17) we find that

$$\frac{m}{l}=\frac{4n_o\alpha}{15\lambda_o}\theta_M^2\left\{1-\left(\frac{16}{63}-\frac{4}{7}\alpha-\frac{22}{105}N+\frac{4}{35}P\right)\theta_M^2+\left[\left(\frac{256}{9009}-\frac{320}{1001}\alpha\right.\right.\right.$$
$$\left.+\frac{400}{1001}\alpha^2\right)-\left(\frac{2378}{15015}-\frac{112}{715}\alpha\right)N+\left(\frac{10496}{135135}-\frac{1312}{15015}\alpha\right)P+\frac{32547}{75075}N^2 \tag{22}$$
$$\left.\left.-\frac{13424}{225225}P^2+\frac{1648}{225225}PN\right]\theta_M^4\right\}.$$

4. COMPARISON WITH EXPERIMENTS AND DISCUSSIONS

For MBBA the refractive indices[6] for 22°C are $n_o = 1.555$, $n_e = 1.787$. The viscosity coefficients of MBBA measured by Kneppe, Schneider and Sharma[7] are $\gamma_1 = 1.526$ poises, $\gamma_2 = -1.542$ poises for 20°C and $\gamma_1 = 1.093$ poises, $\gamma_2 = -1.115$ poises for 25°C. Taking average, we take $\gamma_1 = 1.309$ poises and $\gamma_2 = -1.328$ poises for 22°C. With these values, Equation (16) gives the value of the limiting angle as

$$\theta_{M_o} = 1.471, \tag{23}$$

which agrees excellently with the experimental value 1.475 for 22°C observed by Gähwiller.[8] No accurate data on the elastic constants of MBBA are available. The approximate values[9] are $k_{11} = 6\times10^{-7}$ dynes and $k_{33} = 7.5\times10^{-7}$ dynes. The wave length of the monochromatic light used by Wahl and Fischer in their experiments is $\lambda_o = 5460\text{Å}$. With these constants, using computer and taking intervals of $\Delta\theta_M = 0.01$, we did numerical integrations of Equations (13) and (20). The results are plotted in Figure 2 and Figure 3 respectively. The relationship between

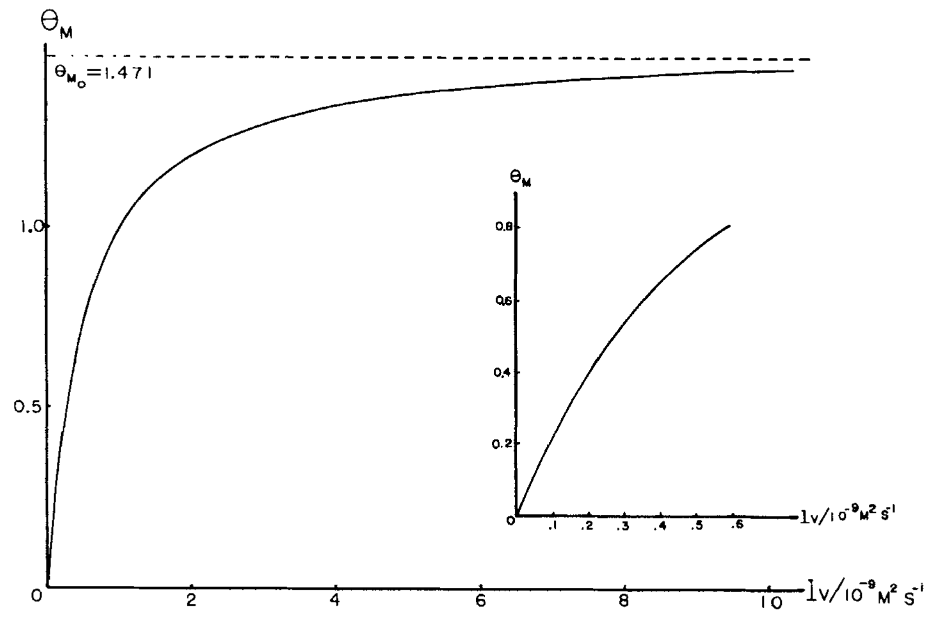

FIGURE 2 The relationship between θ_M and lv.

m/l and lv is shown in Figure 4. For each curve, the part which is near the origin is shown in an enlarged scale on the right side of the corresponding curve.

Equations (17) and (22) now take the form

$$lv = 4.5506 \times 10^{-6}\theta_M(1 + 0.63200\theta_M^2 + 0.31440\theta_M^4 + 0.14110\theta_M^6)\ \mathrm{cm^2\ s^{-1}}, \quad (24)$$

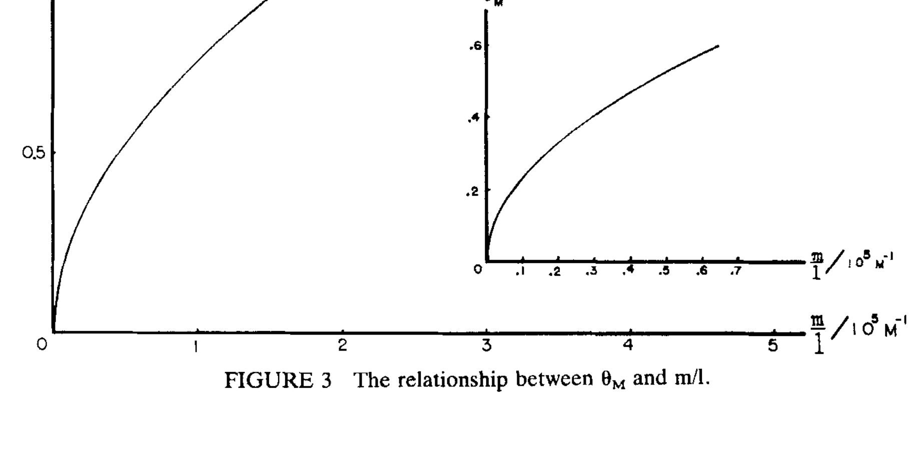

FIGURE 3 The relationship between θ_M and m/l.

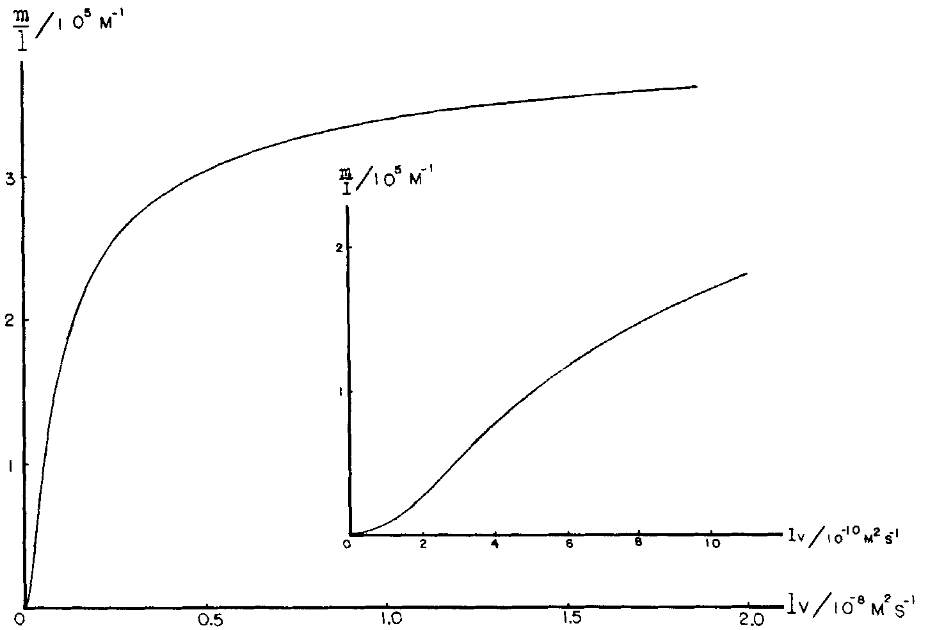

FIGURE 4 The relationship between m/l and lv.

$$m/l = 1.8440 \times 10^3\theta_M^2(1 - 6.774 \times 10^{-2}\theta_M^2 - 7.784 \times 10^{-3}\theta_M^4)\ \mathrm{cm}^{-1} \quad (25)$$

respectively. The values of m/l calculated from Equation (25) agrees very well with that calculated directly from Equation (20). Even up to $\theta_M = 1.10$ they differ by less than 1%. Equation (24) is not a very good approximation of Equation (13) because θ_M should approach θ_{M_0} asymptotically as lv increases. However, for $\theta_M < 0.8$ they still differ by less than 1%.

A quantitative comparison of the present calculation with the experimental results of Wahl and Fischer is not meaningful. On one hand, the liquid crystal used by Wahl and Fischer[2] was impure and furthermore no detailed data are given in References 1 and 2; on the other hand, the elastic constants used in the present calculation may have high error. However, our m/l versus lv curve and the experimental curve obtained by Wahl and Fischer do have the same behavior. This indicates that the present calculation is reasonably correct. For small lv (i.e., for small θ_M), Wahl and Fischer made some approximations[2] to get a linear relationship between $(m/l)/(lv)^2$ and $(lv)^2$. With our calculated numerical values the $(m/l)/(lv)^2$ versus $(lv)^2$ curve is plotted in Figure 5. The part of the curve near the center of the liquid crystal cell is plotted in Figure 6 in an enlarged scale. Both Figure 5 and Figure 6 are not straight lines. This is understandable, because in general the θ_M^3 term in Equation (24) is not negligible. Besides, experimental points shown in Figure 6 of Reference 2 actually are somewhat scattered.

For very small values of θ_M, if we keep only the linear term in Equation (17) and substitute it into Equation (22) we find that

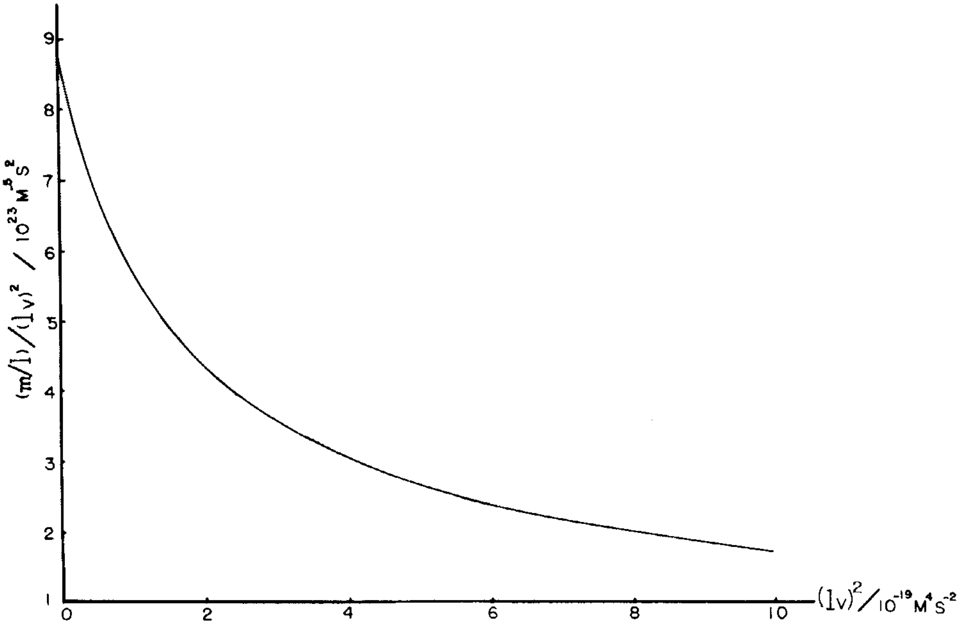

FIGURE 5 Theoretically calculated relationship between $(m/l)/(lv)^2$ and $(lv)^2$.

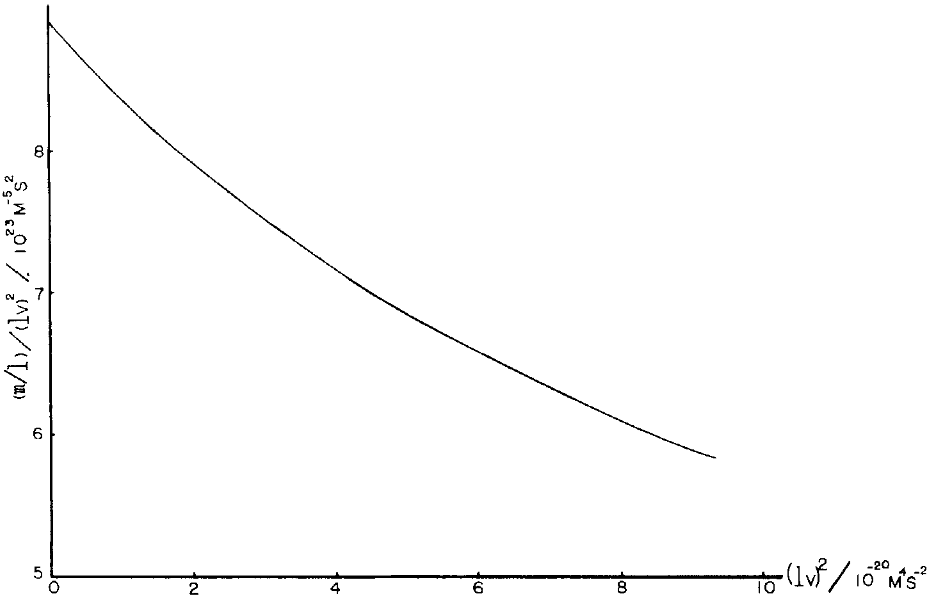

FIGURE 6 Enlargement of Figure 5 near the center of the cell.

$$(m/l)/(lv)^2 = b_1 + b_2(lv)^2 + \ldots, \tag{26}$$

where

$$b_2 = (n_o/960\lambda_o)(1 - n_o^2/n_e^2)[(\gamma_1 - \gamma_2)/k_{33}]^2,$$

$$b_4 = -\frac{15\lambda_o b_2^2}{4n_o(1 - n_o^2/n_e^2)}\left[\frac{4}{7}\frac{n_o^2}{n_e^2} - \frac{20}{63} + \frac{44}{315}\left(\frac{\gamma_2}{\gamma_1 - \gamma_2}\right) + \frac{4}{35}\left(1 - \frac{k_{11}}{k_{33}}\right)\right]. \tag{27}$$

The coefficient b_2 is exactly the coefficient a_2 given by Equation (12) of Reference 2, but b_4 is different from a_4 given in Reference 2. From the experimental value of $a_2 \equiv b_2$ it is possible to determine the value of $(\gamma_1 - \gamma_2)/k_{33}$, but the experimental value of a_4 is still incapable to determine the value of k_{11}/k_{33} from b_4.

Examination of Equations (24) and (25) indicates that it may be better to omit higher order terms in the expansion of m/l than taking lv linearly proportional to θ_M. When we neglect the higher order terms in Equation (22) and substitute it into Equation (17) we find that

$$\begin{aligned} b_2(lv)^2 = \left(\frac{m}{l}\right)\Bigg\{1 &- \frac{\lambda_o}{n_o(1 - n_o^2/n_e^2)}\left[11\left(\frac{\gamma_2}{\gamma_1 - \gamma_2}\right) + 4\left(1 - \frac{k_{11}}{k_{33}}\right)\right]\left(\frac{m}{l}\right) \\ &+ \frac{\lambda_o^2}{n_o^2(1 - n_o^2/n_e^2)^2}\left[\frac{80}{21}\left(1 - \frac{k_{11}}{k_{33}}\right) + \frac{965}{84}\left(\frac{\gamma_2}{\gamma_1 - \gamma_2}\right) + \frac{7949}{84}\left(\frac{\gamma_2}{\gamma_1 - \gamma_2}\right)^2 \right. \\ &\left. + \frac{22}{7}\left(1 - \frac{k_{11}}{k_{33}}\right)^2 + \frac{968}{21}\left(\frac{\gamma_2}{\gamma_1 - \gamma_2}\right)\left(1 - \frac{k_{11}}{k_{33}}\right)\right]\left(\frac{m}{l}\right)^2\Bigg\}. \end{aligned} \tag{28}$$

Thus a plot of $(lv)^2/(m/l)$ versus m/l should be a parabola. With known values of n_o, n_e, λ_o and reliable values of γ_1 and γ_2 from other experiments one should be able to determine k_{11} and k_{33} from the parabola.

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