Director waves in nematic liquid crystals

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A two-dimensional field model was proposed to interpret the director waves in nematic liquid crystals on the basis of the Ericksen-Leslie theory. It is different from previous models and can explain most of the experimental phenomena. The theoretical results are compared in connection with the experimental data. An experimental method was suggested to test the theory.

I. INTRODUCTION

Although some liquid-crystal deformations such as domain walls and disclinations have been well known in both experiment and theory for many years, director waves, which were considered to be the propagation of the orientation of the molecules in liquid crystals, were not verified until this decade. Exciting a liquid-crystal film by manipulating mechanical device, Zhu *et al.* have observed director waves in the nematic liquid-crystal (NLC) material *p,p*-methoxybenzylidene *n*-butylaniline (MBBA). Through two crossed polarizers, they viewed some dark lines moving forward while a film in NLC was driven manually.[1-3] This discovery has attracted great interest, because it could promote developments for practical applications, and some similar phenomena in other systems were reported afterward.[4,5]

To understand the phenomena, Lin Lei *et al.* developed a interesting theory using a soliton model,[6-8] and Wang supposed the existence of solitary waves.[9-11] Both of them reached their conclusions by employing a one-dimensional approximation and explained some aspects of the experimental results. However, the investigation of the problem is not yet complete.

In this work, we pay special attention to the fact that the emergence of the dark lines relies on the anchoring condition presupposed. In the linear approximation of phenomenological equations proposed by Ericksen and Leslie,[12,13] the dark lines can be interpreted simply as director waves. Our results show for the first time that the dependence of the observational phenomena on anchoring conditions is subject to the intrinsic properties, such as the anisotropy of viscosity, of NLC. The movement solution of the dark lines is obtained analytically and is in good agreement with the experimental data.

II. THE EQUATION OF DIRECTOR'S DISTRIBUTION

Based on the Ericksen-Leslie theory, the equations for the director in NLC is (using a convention where repeated indices are to be summed over)

$$\frac{\partial}{\partial t}(I\dot{\mathbf{n}})_i+\frac{\partial}{\partial x_j}(I\dot{n}_i v_j)+\frac{\partial}{\partial x_j}\Pi_{ij}+f_i+f_i'+\lambda n_i=0 \ , \qquad (1)$$

where I is the moment of inertia of the director, λ is the Lagrangian multiplier determined by the relation $\mathbf{n}\cdot\mathbf{n}=1$, $\mathbf{v}$ is the velocity of the fluid, Π_{ij} the strain tensor, f_i the body force density, f_i' the part of the body force associated with the presence of velocity gradients, and n_i the ith component of the director. According to the equation, $\mathbf{n}$ must be $\mathbf{v}$ dependent and may be expanded as a power series in the fluid velocity $\mathbf{v}$,

$$\mathbf{n}=\mathbf{n}^{(0)}+\mathbf{n}^{(1)}+\mathbf{n}^{(2)}+\cdots \ , \qquad (2)$$

where $\mathbf{n}^{(j)}$ is proportional to the jth power of $\mathbf{v}$, and $\mathbf{n}^{(0)}$ is a fixed vector determined by the anchoring conditions. Substituting Eq. (2) and the details of f_i, f_i', Π_{ij} (Ref. 14) into Eq. (1), one can find the first-order equation of $\mathbf{n}$ to be [see Eq. (4.17) in Ref. 15]

$$I\frac{\partial^2 n_i^{(1)}}{\partial t^2}+(\alpha_3-\alpha_2)\frac{\partial n_i^{(1)}}{\partial t}-k_{11}[\nabla_i(\nabla\cdot\mathbf{n}^{(1)})-\mathbf{n}^{(0)}\cdot\nabla(\nabla\cdot\mathbf{n}^{(1)})n_i^{(0)}]-k_{22}[\mathbf{n}^{(0)}\times\nabla(\mathbf{n}^{(0)}\cdot\nabla\times\mathbf{n}^{(1)})]_i-k_{33}n_j^{(0)}n_k^{(0)}n_{i,jk}^{(1)}$$
$$-(\alpha_2+\alpha_3)(\mathbf{n}^{(0)}\cdot\nabla\mathbf{v}\cdot\mathbf{n}^{(0)})n_i^{(0)}+\alpha_2\mathbf{n}^{(0)}\cdot\nabla v_i+\alpha_3\nabla_i\mathbf{v}\cdot\mathbf{n}^{(0)}=0 \ , \qquad (3)$$

$$n_{i,jk}^{(1)}=\partial^2 n_i^{(1)}/\partial x_j\partial x_k \ ,$$

where the parameter λ has been replaced by the quantity gained from $\mathbf{n}\cdot\mathbf{n}=1$. The zeroth-order equation is meaningless, and the higher-order ones may be ignored in linear approximation.

Assuming the Fourier expansions of $\mathbf{n}^{(1)}(t,\mathbf{r})$ and $\mathbf{v}(t,\mathbf{r})$ are

$$\mathbf{n}^{(1)}(t,\mathbf{r})=\int_{-\infty}^{\infty}\mathbf{n}^{(1)}(\omega,\mathbf{q})e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}d\omega\, d\mathbf{q}\ , \tag{4}$$

$$\mathbf{v}(t,\mathbf{r})=\int_{-\infty}^{\infty}\mathbf{v}(\omega,\mathbf{q})e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}d\omega\, d\mathbf{q}\ , \tag{5}$$

respectively, one finds that

$$[I\omega^2+i\omega(\alpha_3-\alpha_2)-k_{33}(\mathbf{n}^{(0)}\cdot\mathbf{q})^2]\mathbf{n}^{(1)}(\omega,\mathbf{q})-k_{11}\{\mathbf{q}\cdot\mathbf{n}^{(1)}(\omega,\mathbf{q})\mathbf{q}-[\mathbf{q}\cdot\mathbf{n}^{(1)}(\omega,\mathbf{q})](\mathbf{q}\cdot\mathbf{n}^{(0)})\mathbf{n}^{(0)}\}$$
$$-k_{22}(\mathbf{n}^{(0)}\times\mathbf{q})[\mathbf{q}\times\mathbf{n}^{(1)}(\omega,\mathbf{q})\cdot\mathbf{n}^{(0)}]=i\alpha_2(\mathbf{n}^{(0)}\cdot\mathbf{q})\mathbf{v}(\omega,\mathbf{q})+i\alpha_3[\mathbf{v}(\omega,\mathbf{q})\cdot\mathbf{n}^{(0)}]\mathbf{q}-i(\alpha_2+\alpha_3)[\mathbf{n}^{(0)}\cdot\mathbf{v}(\omega,\mathbf{q})](\mathbf{q}\cdot\mathbf{n}^{(0)})\mathbf{n}^{(0)}\ . \tag{6}$$

Assuming $k_{11}=k_{22}=k_{33}=k$ as being accepted generally, one can obtain the solution for $\mathbf{n}^{(1)}(\omega,\mathbf{q})$:

$$\mathbf{n}^{(1)}(\omega,\mathbf{q})=i\{\alpha_2(\mathbf{n}^{(0)}\cdot\mathbf{q})\mathbf{v}(\omega,\mathbf{q})+\alpha_3[\mathbf{n}^{(0)}\cdot\mathbf{v}(\omega,\mathbf{q})]\mathbf{q}-(\alpha_2+\alpha_3)[\mathbf{n}^{(0)}\cdot\mathbf{v}(\omega,\mathbf{q})](\mathbf{n}^{(0)}\cdot\mathbf{q})\mathbf{n}^{(0)}\}/[I\omega^2+i\omega(\alpha_3-\alpha_2)-kq^2]\ . \tag{7}$$

This result is independent of the choice of frame and requires only that the directors in NLC are well aligned initially, or $\mathbf{n}^{(0)}$ is a fixed vector in the region being considered.

It is worthwhile to point out that this outcome is obtained with the linear approximation in the fluid velocity $\mathbf{v}$. According to the experimental data, the velocity of the fluid is very small (about 2 mm/sec) and may be fairly considered as a perturbation; thus the linear perturbation method can be employed effectively in the calculation.

III. RELATIONSHIP WITH EXPERIMENT

While driving the exciter manually in the early experiment,[1] Zhu had observed that there were some dark lines moving when the directors $\mathbf{n}^{(0)}$ were forced to be initially perpendicular to the surface of the cell, and that no dark lines emerged when the directors $\mathbf{n}^{(0)}$ were forced to be initially parallel to the surface of the cell. In a word, the dark lines can be generated only if the directors were initially well aligned to the orientation that was perpendicular to the axis on which the Mylar film,as an exciter, can move and cause a perturbation. Zhu *et al.* had also observed that the directors where the dark lines emerged were perpendicular to the surface of the cell. This was verified soon after.[2,3,16] All of these facts show that one can foresee the dark lines by determining the distribution $\mathbf{n}(t,\mathbf{r})$ of the directors.

In our theory, the distribution of the directors is determined by Eq. (7). To discuss it in detail, one may employ a two-dimensional velocity field as one does in considering shear flow, or one may suppose that the directors are motionless along the X_3 axis. Then $v_3=0$ and q_3 vanishes. In addition, the conservation law must be maintained because of the incompressibility of fluid. That is $\nabla\cdot\mathbf{v}=0$, or

$$q_1v_1+q_2v_2=0\ . \tag{8}$$

Moreover, one can rewrite the Eq. (7) in components as

$$n_1^{(1)}=0\ , \tag{9}$$

$$n_2^{(1)}=i(\alpha_2q_1v_2+\alpha_3q_2v_1)/H\ , \tag{10}$$

$$n_3^{(1)}=0\ , \tag{11}$$

$$H=I\omega^2+i\omega(\alpha_3-\alpha_2)-kq^2\ , \tag{12}$$

for the parallel case in which the directors $\mathbf{n}^{(0)}$ are well aligned to be parallel to the X_1-axis initially, and

$$n_1^{(1)}=i(\alpha_2q_2v_1+\alpha_3q_1v_2)/H\ , \tag{13}$$

$$n_2^{(1)}=0\ , \tag{14}$$

$$n_3^{(1)}=0\ , \tag{15}$$

for the perpendicular case, $\mathbf{n}^{(0)}\|\mathbf{X}_2$ (see Fig. 1). These

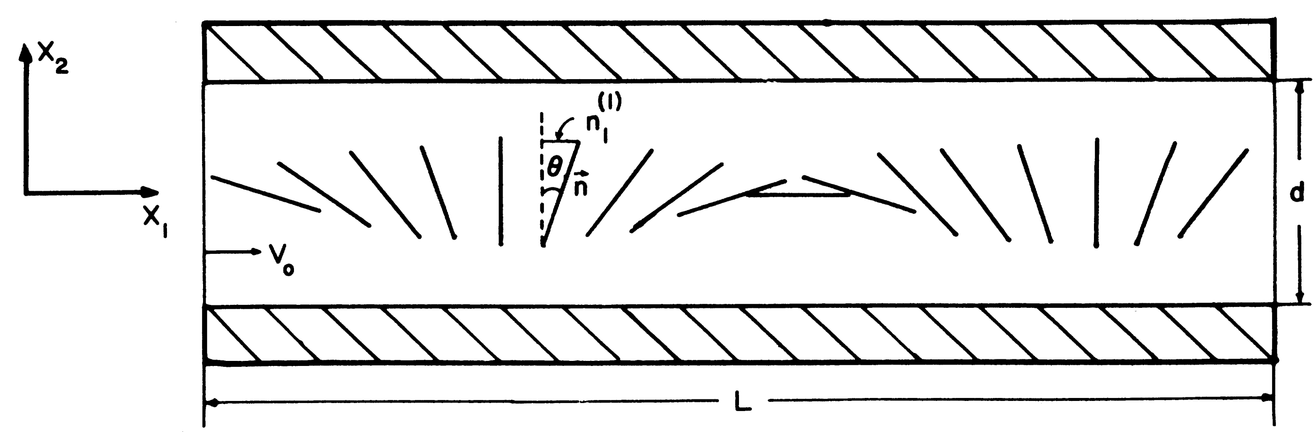

FIG. 1. Orientation of the director for sheared NLC is formed into waves. The sketch shows an intersection of the cell with thickness d and length L. θ is the angle between the director and X_2 axis. The coordinate system used here is the same as that in Ref. 3.

equations show that the director field is two dimensional also, as expected.

It is obvious that the variation of directors in parallel case is different from that in perpendicular case when one applies identical velocity field to the system. The difference can be determined from Eqs. (8), (10), and (13) by the ratio

$$\frac{n_{2,\parallel}^{(1)}}{n_{1,\perp}^{(1)}}=\frac{\alpha_2 q_1 v_2+\alpha_3 q_2 v_1}{\alpha_2 q_2 v_1+\alpha_3 q_1 v_2}=\frac{\alpha_3 q_2^2-\alpha_2 q_1^2}{\alpha_2 q_2^2-\alpha_3 q_1^2} . \tag{16}$$

According to the experimental data, one can estimate the values of the wave vectors[3] (Fig. 1) to be

$$q_1=2\pi/L\sim 10^{-2}\ \mathrm{cm}^{-1},\quad q_2=2\pi/d\sim 10^{2}\ \mathrm{cm}^{-1} ,$$

and acquire the values of the viscosity coefficient[14] (MBBA),

$$\alpha_2=-77.5 \text{ centipoise},\quad \alpha_3=-1 \text{ centipoise} .$$

Therefore, the ratio is roughly

$$n_{2,\parallel}^{(1)}/n_{1,\perp}^{(1)}=\alpha_3/\alpha_2=77.5^{-1} . \tag{17}$$

This implies that the influence of the perturbation is unusually smaller in the parallel case than that in the perpendicular case. In other words, while one attempts to view such a pattern in the parallel case as viewed in the perpendicular case, he must use an exciter which is 77.5 times as powerful as that used in the perpendicular case. This concerns the intrinsic properties of NLC: The molecule in NLC is elongated and has a fairly rigid backbone so that one may consider it as a rigid string. The perturbation due to the velocity field produces a torque which is imposed on the molecule and rotates it to a certain degree. The greater the torque, the larger the rotation degrees. The torque is proportional to the body force and thus depends on the velocity field applied and the viscosity coefficients of the materials used. Though the velocity is the same in the parallel case as that in the perpendicular case, the consequences are different because of the anisotropy of NLC, which is why no dark lines were observed in the parallel case. This idea may be tested in future by using some NLC materials whose viscosity coefficient values, α_2 and α_3, are closer. On the other hand, it would be impossible to test it by employing a more powerful exciter. In doing so, the fluid would be so turbulent that the directors could no longer be regular, and the observable phenomena would be completely different and beyond the scope of this paper.

To compare these results with the experimental data in the perpendicular case, one may solve Eq. (13) in a coordinate system. The transformation yields

$$\left[I\frac{\partial^2}{\partial t^2}+(\alpha_3-\alpha_2)\frac{\partial}{\partial t}-k\left[\frac{\partial^2}{\partial X_1^2}+\frac{\partial^2}{\partial X_2^2}+\frac{\partial^2}{\partial X_3^2}\right]\right]n_1^{(1)} =-\alpha_3\frac{\partial v_2}{\partial X_1}-\alpha_2\frac{\partial v_1}{\partial X_2} . \tag{18}$$

Although the rigorous solutions of this equation need to involve the momentum balance law and convenient boundary condition as shown in Ref. 17, it may be simplified at the moment. First, the boundary effect may be neglected because the anchoring force is weak and the disturbance is relatively strong, or the volume of the cell is so large in comparison with the dimension of the molecules in NLC that one can take it as infinite and ignore the influence of the boundary on the central region of the cell which primarily determines the observable phenomena. Second, one may substitute a suitable δ function for the right-hand side of the Eq. (18) because the perturbation is applied instantaneously at the origin[1-3] and the experimental data show that the velocity is high at the origin and nearly zero otherwise.[16] Furthermore, the inertia term can be omitted according to the point of view generally accepted in current literature, and the term $k\partial^2 n_1^{(1)}/\partial x_3^2$ is zero because the field is two dimensional. From Fig. 1 one becomes aware that $n_1^{(1)}$ is just equal to $\sin\theta$, where θ is the tilted angle caused by the perturbation. Therefore

$$n_1^{(1)}=\sin\theta=\theta-\tfrac{1}{3}\theta^3+\cdots . \tag{19}$$

In the linear approximation in θ, one can obtain the solution

$$\theta=v_0\frac{C}{4\pi a^2 t}\exp\left[-\frac{x_1^2+x_2^2}{4a^2 t}\right] , \tag{20}$$

where v_0 is the component of the fluid velocity in X_1 direction at the origin and is supposed to be the same as the velocity of the exciter, $a^2=k/(\alpha_3-\alpha_2)10^{-8}$ cm^2/sec, and C is a parameter which represents the strength of the coupling between director and the fluid velocity and needs a better fit with the experimental observation.

It is obvious that the variable x_2 in the Eq. (20) can be neglected since its changeable region (-25–25 μm) is extremely small in comparison with that of the variable x_1 (-25–25 cm) (Ref. 3) and can cause nothing meaningful

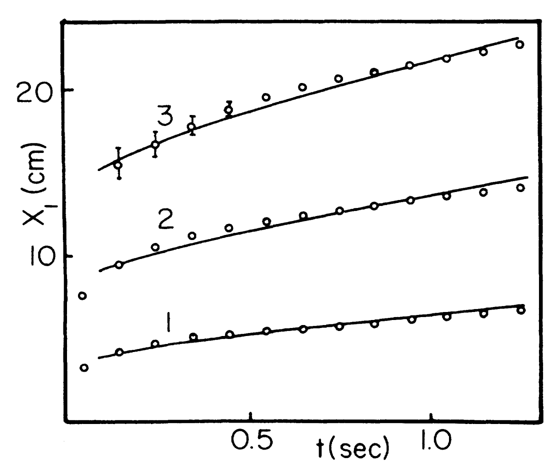

FIG. 2. Comparison of experimental data of dark lines (circles, adopted from Ref. 3) with the theoretical results (lines). See text for details.

to observe. That is, one can focus his attention primarily on the center line of the cell used in the experiment. In this way, one can now see that there must be some positions at which θ is a multiple of π, so that the directors at these positions are perpendicular to the X_1 axis and certain waves are formed with their peaks placed at these positions. These waves are called director waves and were observed through experiment as dark lines. Moreover, one can certainly see that θ is proportional to the velocity of the exciter; the greater the disturbing velocity, the larger the θ, or the larger the number of the dark lines. This expected consequence is in accordance with that reported earlier by Zhu in his experiment.[1]

Considering a certain dark line, one may be able to conclude the movement of the dark lines because θ is a constant at this moment. With the experimental data, the variations of x_1, which describes the positions of the dark lines at present, may be calculated from Eq. (20) and are sketched in Fig. 2. Here, the parameter C was suitably chosen to fit the experimental data.[3] Figure 2 shows that the theoretical results are in good agreement with the experimental data[3] for all of the three dark lines. Noting that there is only one adjustable parameter in the theory and that the experimental accuracy is rather low, such an agreement should be considered satisfactory. The discrepancy may be explained by the roughness of the model, not taking into account the details of the velocity field.

To summarize, it has been shown that the dark lines observed are director waves and can be explained by the Ericksen-Leslie theory. Because of the anisotropy of NLC, the dark lines can be generated only when the moving direction of the exciter is vertical to the orientation in which the NLC molecules are initially well aligned. In this sense, the present model is quite different from the previous theoretical consideration[6–11] on the same problem and is in good agreement with the most of the experimental results. The opinion presented here may be tried by testing some NLC materials whose viscosity coefficients, α_2 and α_3, are closer.

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