Static External Field Effects on the Hydrodynamic Instability of Liquid Crystals Subject to Simple Shear Flow

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ABSTRACT: External field effects on the hydrodynamic instability of a nematic liquid crystal (LC) monodomain are analyzed using the two-dimensional Ericksen−Leslie continuum theory neglecting the Frank elasticity. It is found that in a static magnetic field, the so-called aligning−type LC with the third Leslie coefficient $\alpha_3 < 0$ can, for an appropriate field strength and direction, exhibit tumbling under a simple shear flow, whereas the original tumbling−type LC with $\alpha_3 > 0$ can show aligning in a strong magnetic field. The aligning−tumbling and tumbling−aligning transition points are calculated. Corresponding state diagrams for director tumbling and shear aligning are presented, and the tumbling loop is found. The aligning angles in the stable state and the tumbling periods in the unstable state are described analytically, along with the necessary and sufficient conditions for director tumbling. It is especially noteworthy that the direction of the external field plays a crucial role in controlling the hydrodynamic instability of LCs. As a consequence, this paper affords an interesting and feasible model system of a complex anisotropic fluid and establishes a framework for later theoretical and experimental studies.

Introduction

Liquid crystals (LCs) including liquid crystalline polymers (LCPs) constitute an important class of complex fluids.[1,2] The main focus of both academic interest and practical application is their orientation properties. The orientation of LCs is strongly affected by the flow field, external electric or magnetic fields, etc.

The rotation of the director of an LC can be discussed in terms of the Ericksen−Leslie (E−L) continuum theory.[3−5] The static and dynamic behavior of a nematic LC in a flow field can be described by the six Leslie coefficients, α_1−α_6 along with the Frank elastic constants.[4] With respect to the influence of the flow field on LC orientation, nematic LCs may be roughly divided into two classifications according to the sign of the third Leslie coefficient.[6] If $\alpha_3 < 0$, a steady alignment of the LC can be achieved at a characteristic angle relative to the flow direction known as the Leslie angle. In contrast to this case, a positive α_3 leads to an indefinite and periodic rotation of the average molecular orientation, namely, the LC director, about the vorticity axis under simple shear flow. The former situation is called shear aligning, while the latter is referred to as director tumbling. Most small molecular nematics are known to be flow aligning with only a few exceptions.[7,8] However, a large number of LCPs, especially the main-chain ones, show director tumbling.[9−17] The concept of director tumbling, a kind of hydrodynamic instability, has been successfully employed to explain many unusual rheological properties of LCPs, such as the negative first normal stress difference at moderate shear rates and pronounced oscillations of stress and dichroism with time.[9−29]

Static external field effects on the orientation of LCs with or without shear flow have been practically applied to measure the fundamental parameters of LCs such as Frank elastic constants and viscosity coefficients.[1] A nematic LC in a continuously rotating magnetic field has also been studied experimentally, and novel nonlinear dynamic spatial structures have been found.[30,31] In contrast, reports concerning the effects of static external fields on director tumbling instead of shear aligning are rather scarce. Carlson and Skarp found that a transverse electric field suppressed tumbling during the shear flow of small molecular nematics with $\alpha_3 > 0$.[32,33] Their findings were further confirmed by Yang and Shine in an LCP.[34] Nevertheless, these works are limited to transverse electric fields and their stabilizing effect on the orientation state of LCs. Several important questions are thus open: for instance, how the direction of an external field affects the stability of an LC and whether a static external field can induce flow instability in an LC. Very recently, Brownian dynamics simulations and the Lebwohl−Lasher nematogen model were combined by the authors to investigate external magnetic field effects on director dynamics and flow instability in LCs subject to simple shear flow.[35,36] The most important finding is that a static magnetic field can, depending on the field strength and orientation, enhance or inhibit the flow instability. That is very interesting, and further studies in theory and experiment are absolutely necessary.

In this paper, the effect of a static magnetic field on the hydrodynamic instability of an LC is investigated

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by the E–L continuum theory. From our analysis, we find that the so-called aligning–type LC with $\alpha_3 < 0$ can exhibit tumbling for an appropriate strength and direction of a static external field, whereas the tumbling–type LCs can be aligned by an external field. The conditions of the aligning–tumbling transition induced by a static magnetic field are given analytically, and the state diagram for director tumbling and shear aligning in an external field is presented. Since the E–L theory is valid for both small molecular LCs and LCPs, the results obtained in this paper are, of course, not limited to LCPs, although LCPs exhibit more complicated hydrodynamic instabilities and are thus more interesting. The main conclusions are also applicable for an external electric field, because the effects of electric fields on an LC are similar to those of magnetic fields to some extent, unless charge effects are striking.

Theoretical Method

For simplicity, we adopt the linear E–L theory for an LC monodomain in two dimensions. The LC dynamics in three dimensions might be more complicated, due to the possible out-of-plane rotation of local directors during a tumbling orbit.[21] Nevertheless, the physical essence of the external field effects on the hydrodynamic instability of LCs undergoing shear flow can be captured in a two-dimensional analysis. The coordinate frame is presented in Figure 1. A constant shear rate $\dot{\gamma}$ is imposed in a simple shear flow. Both the orientations of the LC director and the magnetic field can be varied, which are described by θ and θ_h, measured anticlockwise with the zero angle along the x-axis, the flow direction. In order to simplify the theoretical description while the essential feature is retained, several further approximations are made. We neglect the Frank elastic contribution and the surface anchoring effect, which implies that the system is spatially uniform. Fluctuations of the order parameter are also not present in the classic theory. Then, the director rotation of an LC monodomain undergoing a simple shear flow and in a static magnetic field can be described by[3,4]

$$-\gamma_1 \frac{d\theta}{dt} = (-\alpha_2 \sin^2 \theta + \alpha_3 \cos^2 \theta)\dot{\gamma} + \epsilon_h \sin[2(\theta - \theta_h)] \quad (1)$$

Here α_2 and α_3 are the second and third Leslie coefficients, which usually satisfy the relationships $\alpha_2 < 0$ and $|\alpha_3/\alpha_2| << 1$, and thus the other two important parameters with the physical meaning of the LC viscosity, $\gamma_1 = \alpha_3 - \alpha_2$ and $\gamma_2 = \alpha_2 + \alpha_3$, must have the signs $\gamma_1 > 0$ and $\gamma_2 < 0$. The term ϵ_h refers to the maximum magnetic energy for a given magnetic field strength, H, with $\epsilon_h = 0.5\chi_a H^2$, where χ_a is the anisotropy of the magnetic susceptibility and is, without loss of generality, assumed to be positive. On the basis of eq 1, we can obtain the state diagram using a linear stability analysis.

Results and Discussion

Tumbling Condition in Static Magnetic Fields. Both sides of eq 1 can be divided by $\epsilon_h \cos^2 \theta$ if we tentatively assume $\theta \neq (k + 1/2)\pi$, where k is an integer. Then, eq 1 is rewritten as

$$\frac{-\gamma_1}{\epsilon_h \cos^2 \theta} \frac{d\theta}{dt} = (-\alpha_2 \,\mathrm{tg}^2\theta + \alpha_3)\frac{\dot{\gamma}}{\epsilon_h} + 2 \cos 2\theta_h \mathrm{tg}\theta - \sin 2\theta_h(1 - \mathrm{tg}^2 \theta) \quad (2)$$

Let

$$x = \mathrm{tg}\, \theta, \quad y = \frac{-\gamma_1}{\epsilon_h \cos^2 \theta} \frac{d\theta}{dt} \quad (3)$$

we have

$$y = ax^2 + bx + c \quad (4)$$

where

$$a = \sin 2\theta_h - \frac{\alpha_2\dot{\gamma}}{\epsilon_h} \quad (5a)$$

$$b = 2 \cos 2\theta_h \quad (5b)$$

$$c = -\sin 2\theta_h + \frac{\alpha_3\dot{\gamma}}{\epsilon_h} \quad (5c)$$

This is a parabolic equation if $a \neq 0$. Linear stability analysis reveals that if $\Delta = b^2 - 4ac > 0$, one of the two solutions of $y = 0$ corresponds to a stable state, which results in shear aligning. On the other hand, if $\Delta < 0$, no real solution of the equation $y = 0$ exists, which leads to director tumbling with a definite rotation direction ($d\theta/dt < 0$). In fact, all of the tentative assumptions such as $\theta \neq (k+1/2)\pi$ and $a \neq 0$ can be relaxed, and detailed derivations are omitted for brevity here. Finally, we obtain the necessary and sufficient condition for director tumbling in an LC monodomain, which reads

$$g(S_h) = S_h^{\,2} - (\alpha_2 + \alpha_3)(\sin 2\theta_h)S_h + \alpha_2\alpha_3 < 0 \quad (6)$$

where $S_h = \epsilon_h/\dot{\gamma} = \chi_a H^2/(2\dot{\gamma})$, indicating the effective field strength relative to the shear rate. Equation 6 demonstrates that whether an LC system exhibits tumbling or aligning subject to a simple shear flow is determined not only by the sign of the third Leslie coefficient, α_3, but also by the strength and direction of the external field if it is also exerted on the LC. For convenience in the following discussion, we will denote the field direction I as referring to the first or third quadrant and the field direction II as referring to the second or fourth quadrant of the coordinate frame defined by Figure 1.

Aligning–Tumbling Transition and the State Diagram. On the basis of eq 6, we can readily obtain the state diagram for director tumbling and shear aligning in a static magnetic field. The state diagram associated with the so-called tumbling–type LC with $\alpha_3 > 0$ is presented in Figure 2a. The occurrence of director tumbling is retained for the original tumbling–type LC, if the external field is very weak ($\epsilon_h < \alpha_3\dot{\gamma}$). On the other hand, the tumbling is completely suppressed by a strong field with $\epsilon_h > -\alpha_2\dot{\gamma}$. At a moderate field strength ($\alpha_3\dot{\gamma} \leq \epsilon_h \leq -\alpha_2\dot{\gamma}$), whether an LC microdomain undergoes director tumbling or flow aligning depends strongly upon the field direction as well as upon the field strength and shear rate according to the relation found in eq 6. The tumbling–aligning transition point, $S_{h,o}$, can be determined by solving the

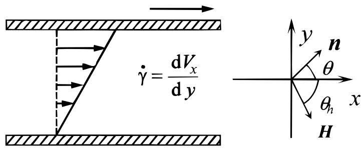

Figure 1. Coordinate frame for a two-dimensional nematic monodomain subject to a simple shear flow and in a static magnetic field. $\dot{\gamma}$ is the constant shear rate along the velocity gradient direction (y-axis). θ and θ_h denote the orientation angle of the LC director $\boldsymbol{n}$ and that of the external magnetic field $\boldsymbol{H}$, respectively, with the zero angle defined along the flow direction (x-axis) and the positive angle defined along the anticlockwise direction.

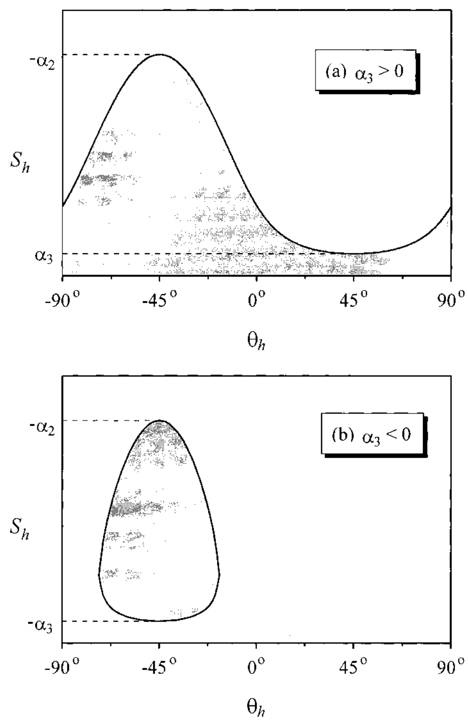

Figure 2. State diagrams of an original tumbling–type (a) and aligning–type (b) nematic monodomains subject to a simple shear flow and in a static magnetic field. $S_h = \epsilon_h/\dot{\gamma}$ refers to the relative field strength. Director tumbling takes place in the shaded regimes, while flow aligning occurs outside the shaded regimes.

equation $g(S_h) = 0$. The maximum and minimum values are $S_{h,o}(\max) = -\alpha_2$ for $\sin 2\theta_h = -1$ and $S_{h,o}(\min) = \alpha_3$ for $\sin 2\theta_h = 1$, which are marked in Figure 2a. The tumbling–aligning transition takes place with increasing field strength or decreasing shear rate (see Figure 2a). It has been reported in the literature that the application of a sufficiently strong transverse electric field to a tumbling nematic LC undergoing shear flow can suppress the occurrence of tumbling and cause the director to assume a flow aligned angle,[32–34] a case which is included in the state diagram given in Figure 2a. However, the cases previously studied consider only a transverse field in the fixed perpendicular field direction ($\theta_h = 90°$ according to our notation). More complicated phenomena involving the whole field direction regime including both field direction I ($0 \leq \theta_h \leq 90°$) and field direction II ($90° < \theta_h < 180°$) predicted by Figure 2a are not included in these experiments and theoretical analyses.

What is unexpected on the surface and thus more interesting is that the so-called aligning–type LC with $\alpha_3 < 0$ can also exhibit tumbling subject to a static

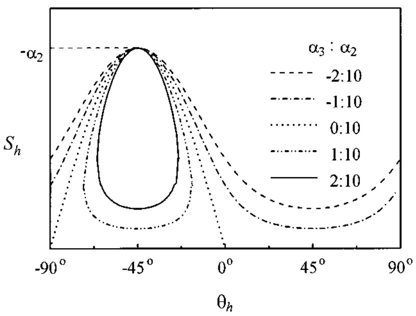

Figure 3. State diagrams for director tumbling and flow aligning of a nematic monodomain with different signs and absolute values of the third Leslie coefficient α_3 under a given second Leslie coefficient α_2 ($\alpha_2 < 0$, $|\alpha_2| >> |\alpha_3|$).

magnetic field with an appropriate field strength and direction. The corresponding state diagram is presented in Figure 2b. For a field direction I, for instance, with a transverse external field, the flow aligning is retained at any field strength and shear rate. On the contrary, two state transitions take place for a field direction II with increasing field strength or decreasing shear rate: the first is an aligning–tumbling transition; the second is a tumbling–aligning transition. In this field direction, the LCs exhibit different states in a low field ($\epsilon_h < -\alpha_3\dot{\gamma}$), in a moderate field ($-\alpha_3\dot{\gamma} \leq \epsilon_h \leq -\alpha_2\dot{\gamma}$) and in a high field ($\epsilon_h > -\alpha_3\dot{\gamma}$) (see Figure 2b). The state diagram shown in Figure 2b has a similar shape to that obtained from our corresponding Brownian dynamics simulation (Figure 1 in ref 35).

For the original aligning–type LCs, the aligning–tumbling or tumbling–aligning transition point can be analytically described. Both maximum and minimum values correspond to $\sin 2\theta_h = -1$ and read $S_{h,o}(\max) = -\alpha_2$, $S_{h,o}(\min) = -\alpha_3$, which are marked in Figure 2b. This figure indicates that the field-induced tumbling in the otherwise aligning–type nematics can only take place in field direction II. The transition points for an LC with $\alpha_3 < 0$ constitute a loop in this field direction regime (Figures 2b and 3). We term it a *tumbling loop*. Further analysis reveals that the tumbling loop shrinks with decreasing α_3 for a given α_2 ($\alpha_2 < 0$ and $|\alpha_2| >> |\alpha_3|$), which is clearly shown in Figure 3. Figures 2 and 3 also demonstrate that the tumbling regime is greatest when $\theta_h = -45°$, which implies that in order to achieve director tumbling or avoid this instability, it is better to adjust the field direction near or far away from the diagonal of the second or fourth quadrant.

We would like to give a preliminary explanation for the static magnetic field effects on LCs undergoing shear flow, especially for the aligning–tumbling transition in otherwise aligning–type nematics at the moderate field strength. The rheological behavior of LCs in a low field and high field can be understood easily. In a low field, the magnetic torque exerted on the director is too weak to overcome the hydrodynamic force. The LC monodomain will retain its original aligning or tumbling characteristics (Figures 2a and 2b). In the other extreme case, a very strong external field must lock the nematic director near the field direction and suppress the flow instability completely. Essentially, the director dynamics and the corresponding dissipative structures in these systems are determined by a subtle balance between the viscous, elastic, and magnetic effects. For director tumbling, a torque is needed to drive the director past a bottleneck clockwise in the shear plane according to the coordinate frame defined in Figure 1. The so-called bottleneck is, in zero magnetic field, along the flow direction ($\theta = 0$), where the hydrodynamic torque is zero or nearly zero (see eq 1 considering $|\alpha_3| << |\alpha_2|$). However, this bottleneck may be altered or even eliminated when a static external magnetic field is imposed on the LC. A magnetic field with a direction in the second or fourth quadrant forces the LC director with an orientation near the flow direction to rotate clockwise. Therefore, such a static field may enhance director tumbling at an appropriate field strength. It will be proven below that the fastest tumbling occurs when $\theta_h = -45°$, since the field direction is then in a suitable regime, in which the magnetic torque exerted on the LC director with $\theta = 0$ is maximal for a given field strength (see eq 1). Due to a similar reason, a static magnetic field with a direction in the first or third quadrant must inhibit tumbling and enhance aligning. This explains why a static external field should be in direction II and the field strength should be moderate in order to induce the aligning−tumbling transition.

Aligning Angles and Tumbling Periods in External Fields. If the tumbling condition (eq 6) is not satisfied, the nematics, irrespective of being the tumbling type or aligning type, are aligned in the stable state. However, the aligning angle of the LC director in an external field must be different from the Leslie angle in zero field. The value can be calculated according to the formula given by the solution of $y = 0$ (eq 4) corresponding to the stable state,

$$\mathrm{tg}\theta_b = \frac{-(\epsilon_h/\dot{\gamma})\cos 2\theta_h + \sqrt{(\epsilon_h/\dot{\gamma})^2 + \alpha_2\alpha_3 - (\alpha_2 + \alpha_3)(\epsilon_h/\dot{\gamma})\sin 2\theta_h}}{(\epsilon_h/\dot{\gamma})\sin 2\theta_h - \alpha_2} \tag{7}$$

This expression is valid for both $\alpha_3 > 0$ and $\alpha_3 \le 0$. In an extreme case with $\epsilon_h/\dot{\gamma} \to \infty$, eq 7 leads to $\theta_b \to \theta_h$. Hence, the LC director is locked along the field direction if the external field is sufficiently strong.

The results obtained from the continuum theory are consistent with those obtained from our Brownian dynamics simulation.[36] The theoretical calculation of aligning angles agrees with the experimental measurement by Grabowski and Schmidt very well.[37] Unfortunately, director tumbling was not observed by them, because the magnetic field for NMR is, according to our analysis, overly strong and the field direction of a transverse field is beyond the tumbling regime.

In the tumbling regime, the tumbling period is one of the key physical quantities in describing nonlinear dynamics of LCs. In zero field, tumbling periods in terms of shear time t_p or shear strain γ_p follow a well-known scaling relation: $\gamma_p = \dot{\gamma} t_p =$ constant, which has been repeatedly observed experimentally[12−17] and reproduced theoretically.[20−29] We find out, however, that this simple scaling relation is never satisfied when an external magnetic field is exerted on the LC.

After rewriting eq 1 followed by integral calculus, we successfully obtained the analytical expression of the tumbling period of an LC monodomain undergoing shear flow with a static magnetic field imposed on it,

$$\gamma_p = \dot{\gamma} t_p = \frac{\gamma_1 \pi}{\sqrt{-\alpha_2\alpha_3 + \gamma_2(\sin 2\theta_h)(\epsilon_h/\dot{\gamma}) - (\epsilon_h/\dot{\gamma})^2}} \tag{8}$$

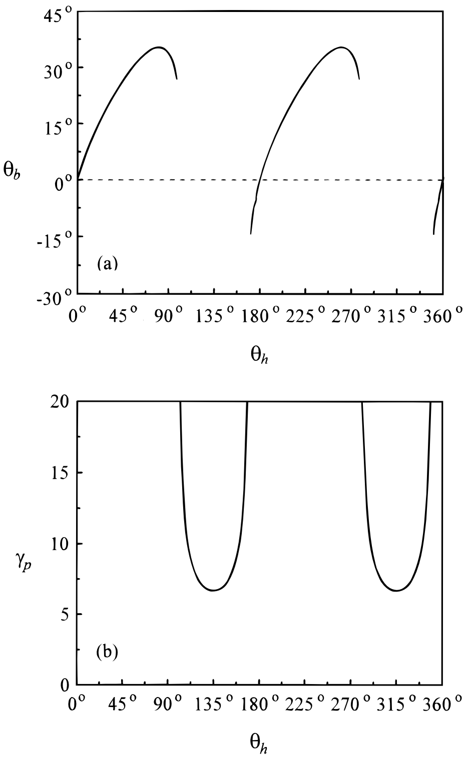

Figure 4. Aligning angle θ_b (a) and tumbling period γ_p (b) as a function of field direction θ_h showing the field direction effects on the rheological behavior of a nematic monodomain subject to a simple shear flow and in a static magnetic field. The field strength is relatively moderate with $\epsilon_h/\dot{\gamma} = -\alpha_2/3$, and the approximation $\alpha_3 = 0$ is employed for simplicity.

This expression is valid for both tumbling−type and aligning−type LCs. Equation 8 clearly illustrates that tumbling periods of an LC director in a static magnetic field are very sensitive to shear rates as well as to field strengths and directions.

Further analysis reveals that γ_p with $\epsilon_h \neq 0$ is always smaller than that in zero field. So, LC directors tumble faster in a static magnetic field than in zero field. However, the tumbling cannot proceed indefinitely fast. Especially when the field direction is located at the diagonal of the second or fourth quadrant ($\theta_h = -45°$) and the field strength is appropriate with $\epsilon_h = -0.5\,\gamma_2\dot{\gamma}$, there is a lower limit tumbling period with $\gamma_p(\min) = 2\pi$ in terms of shear strain. This case corresponds to a unique linear tumbling mode with a constant rotation velocity. These theoretical analyses are in accord with the corresponding Brownian dynamics simulation,[35,36] but unfortunately, we have not found any experimental evidence in the present literature as of yet to support this.

It seems worthwhile to emphasize that the field direction plays a crucial role in controlling the flow instability of LCs, especially at a moderate relative field strength. Some typical results are presented in Figure 4, where field directions are varied while the field strength and shear rate are given. Figure 4a shows some aligning angles in the aligning state associated with a magnetic field oriented mainly in field direction I, whereas Figure 4b describes some tumbling periods in the tumbling state associated with a magnetic field oriented in field direction II. Both aligning angles and tumbling periods depend strongly upon field directions.

Concluding Remarks

The two-dimensional linear E−L continuum theory has been employed in this paper to investigate external field effects on a nematic monodomain subject to a simple shear flow. The most important finding is that static magnetic fields can, depending on field strengths and directions as well as shear rates and LC viscosities, enhance and even induce director tumbling, rather than merely suppress tumbling. The necessary and sufficient conditions for director tumbling are given analytically, together with aligning angles in the aligning state and tumbling periods in the tumbling state. The tumbling loop in an otherwise aligning−type LC with $\alpha_3 < 0$ is found and analyzed. Corresponding state diagrams for director tumbling and flow aligning are presented graphically in both cases of $\alpha_3 > 0$ and $\alpha_3 \leq 0$. It is revealed that an appropriate magnetic direction is a key to director tumbling in external fields, which might be helpful for guiding later experiments.

It is noteworthy that LC samples involve, as usual, many defects and are thus multiple-domain systems instead of monodomain ones. In a multiple-domain system, director tumbling gives way to damped oscillation,[22,23] which might account for the final disappearance of the oscillations of material functions.[10−17,38] In fact, director tumbling has never been reported as lasting for a very long time according to experimental observations in the present literature. Nevertheless, we would like to promise that the lasting tumbling of nematics undergoing a simple shear flow may possibly be found in a static magnetic field due to the following reasons: first, to impose an external field on an LC sample is a conventional method to achieve an LC monodomain;[1,34] second, for a monodomain nematic, the tumbling in the shear plane is very striking and can last, in principle, for an indefinitely long time in an appropriate magnetic field, according to our two-dimensional analysis. In consequence, this paper affords a feasible LC system to investigate hydrodynamic instabilities and associated nonlinear rheological behavior of anisotropic fluids.

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