Elongational perturbations on nematic liquid crystal polymers under a weak shear

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The two-dimensional Smoluchowski equation is employed to study the effect of elongational perturbations on nematic liquid crystal polymers under a weak shear. We use the multiscale asymptotic analysis to show that (1) when the elongational perturbation is small relative to the weak shear, the orientational probability density function (pdf) tumbles periodically only in an intermediate range of polymer concentration; outside this intermediate range (i.e., for very small and very large polymer concentration) the orientational pdf converges to a steady state and there is no tumbling. (2) When the elongational perturbation is about 20% of the shear rate or larger, the intermediate range of tumbling disappears and the orientational pdf always converges to a steady state regardless of the polymer concentration. Our theoretical predictions are consistent with various earlier results based on the Leslie–Ericksen theory [C. V. Chaubal and L. G. Leal, J. Non-Newtonian Fluid Mech. **82**, 22 (1999)] or analogous 3D numerical simulations [M. G. Forest, R. Zhou, and Q. Wang, Phys. Rev. Lett. **93**, 088301 (2004); M. G. Forest, Q. Wang, R. Zhou, and E. Choate, J. Non-Newtonian Fluid Mech. **118**, 17 (2004)]. © *2007 American Institute of Physics.*
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I. INTRODUCTION

Nematic liquid crystal polymers (LCP) are technologically important materials.[1,2] For example, nematic LCPs have been spun into high strength fibers which were used as airbags that cushioned the landing of NASAs highly successful missions to Mars.[3] The dynamics of nematic LCPs has been widely modeled by the kinetic Doi–Hess theory.[4,5] The basic idea behind the Doi–Hess theory is to treat the LCP as a suspension of rigid rodlike nematogenic molecules and describe the ensemble with an orientational probability density function (pdf). The orientational pdf evolves according to a nonlinear Smoluchowski (Fokker–Planck) equation where the forces acting on the rodlike molecules include hydrodynamic, Brownian, and intermolecular forces. The Smoluchowski equation provides a rich variety of mathematical problems and it has been investigated theoretically for pure nematic equilibria,[6–12] extensional flow-induced equilibria,[13] equilibria of dipolar ensembles,[14–16] effect of high[17] and weak shear,[18] and effect of coplanar magnetic field.[19] Numerical simulations of the Smoluchowski equations and the Doi tensor model have been carried out extensively, for example, see Refs. 20–32.

The material properties of the LCPs are highly affected by the processing flows. It is practically important to understand flow effects. Marrucci and Maffettone gave the first elegant two-dimensional analysis on the effect of shear.[33–35] Later Larson[30] carried out three-dimensional numerical simulations using spherical harmonic expansions. Larson's results confirmed the theoretical predictions of Marrucci and Maffettone. In 1999 Chaubal and Leal[20] applied a Lagrangian numerical method called the smoothed particle hydrodynamics technique to explore the effect of flow perturbations on LCPs under a simple shear. They considered elongational perturbations to a simple shear to make it slightly more extensional than a pure shear and found that slight perturbations to the flow field can arrest periodic tumbling behavior. This phenomenon has been observed for both low and high shear rates even though the mechanism is different. In Ref. 20, numerical simulations were carried out in the case of finite Péclet number where the Péclet number denotes the shear rate nondimensionalized with respect to the rotational diffusivity. In the asymptotic limit of small Péclet number, the Leslie–Ericksen theory was used to explore deviations from simple shear. By introducing an effective aspect ratio and plotting the critical value of perturbation (α) as a function of the polymer concentration (U), it was revealed that for $\alpha > 0.04$, the orientational director flow-aligns for all U; for $\alpha < 0.04$, there is an interval (U_{min}, U_{max}) within which the orientational director displays periodic tumbling behavior; outside of the interval (U_{min}, U_{max}), the director flow-aligns again. For U less than the isotropic-nematic transition 4.49, the director flow-aligns for all $\alpha \geq 0$.

Recently Forest's group did extensive research on various solutions of the 3D Smoluchowski equations under different kinds of conditions.[19,22–28,36,37] In particular, in Ref. 36 they formulated a monodomain correspondence principle of kinetic and mesoscopic theory for arbitrary aspect ratio nematic polymers in general linear planar flow. From the principle, the monodomain response of nematic polymers for all linear flows in the plane of shear is identical to the mon-

odomain response of another nematic polymer liquid with a renormalized molecular aspect ratio parameter in pure simple shear flow. Later they extended the study in Ref. 36 to track the boundaries of the chaotic region of kinetic theory due to finite molecule aspect ratio and the addition of straining flow in the plane of shear.[25] It was predicted that sheared chaotic response persists up to a threshold straining flow strength and minimum aspect ratio, beyond which chaotic behavior is arrested. Both[25,36] gave predictions on how steady and unsteady attractors are modified and eventually lost or persist in the presence of a variable extensional flow contribution.

In the 2D case, it is well-known that in the absence of flow the isotropic-nematic phase transition occurs at $U=2$, where U is the normalized polymer concentration. In the presence of an imposed weak shear there is a threshold ($U_0 \approx 2.41144646$) for U: When $U < U_0$, steady state solution exists; otherwise there is no steady state and the orientational pdf is temporally periodic ("tumbling") and can never reach a steady state. If one perturbs the weak shear by a small elongation, this scenario will change. In fact, a shear flow can be viewed as the superposition of a rigid body rotation and an elongational flow. So the imposed elongational flow will either increase or decrease the elongational component of the shear flow depending on how their directions are aligned with respect to each other. We will show that if the elongational component is increased by a small fraction, then there is no tumbling at large polymer concentration. This leads to the situation that the orientational pdf tumbles periodically only in an intermediate range of polymer concentrations; outside this intermediate range (below the lower end of the range or above the upper end of the range) the orientational pdf converges to a steady state and there is no tumbling. If the elongational component is increased further (by about 20% of the original elongational component of the shear flow), the intermediate range of tumbling disappears; that is, for all polymer concentrations, the orientational pdf converges to a steady state. Our theoretical results are consistent with Refs. 20, 25, and 36.

Our asymptotic analysis invokes a multiscale analysis[38] with the assumption that the solution is a quasisteady state. From the physical point of view, this assumption is reasonable. When the flow field is weak, it takes a long time for the flow field to change the orientation distribution significantly. In other words, the time scale of the flow field affecting the orientation distribution is large. On the other hand, in the absence of flow field, the time scale of the intermolecular force (Maier–Saupe interaction) relaxing the orientation distribution to the Boltzmann form is fixed. So in the presence of a weak flow field, it is reasonable to expect that the intermolecular interaction will be able to drive the orientation distribution approximately to the Boltzmann form before the flow field can take a significant effect on the orientation distribution. This is because the time scale of relaxing to the Boltzmann form is small in comparison to the time scale of the weak flow field. Thus, it is reasonable to assume mathematically that to the leading order the orientation distribution is of the Boltzmann form. Of course, the director of the orientation distribution may change in time driven by the weak flow field. Although this assumption is reasonable on

physical grounds, it is desirable to have a rigorous mathematical justification based on the solvability condition[27] or other arguments. Currently such a rigorous mathematical justification remains an open problem. We will pursue this issue in our future work. In recent years, multiscale asymptotic analysis has become a powerful mathematical tool in many applications. In Ref. 39, Choate and Forest used it to investigate the viscoelastic response of nematic polymers to small amplitude oscillatory shear using the Doi–Hess mesoscopic orientation tensor model; Vicente Alonso et al.[40] applied it to study the nonlinear dynamics of a nematic liquid crystal in the presence of a plane Couette flow using a Landau-de Gennes model; Chillingworth et al.[41] employed it to describe the geometry and dynamics of a nematic liquid crystal in a uniform shear flow.

It is worthwhile to point out that other perturbation schemes have been used to study both the kinetic and tensor models for nematic liquid crystal polymers. For example, Kuzuu and Doi[42,43] and Semenov[44] developed a weak-flow asymptotic analysis of the molecular kinetic equation for nematic liquid crystals, which provides a molecular basis for all continuum theory parameters. Doi and Larson[45] analyzed the effect of steady flow fields on the isotropic-nematic phase transition of rigid rodlike polymers by a singular perturbation method; Forest, Zhou, and Wang[27] expanded the orientational distribution function in the Péclet number to find approximate steady solutions in weak shear. The works of Refs. 42, 43, 45, and 27 all solved the kinetic equation and derived rigorous "solvability conditions." The advantage of this kind of approach is that unlike multiscale asymptotic analysis, it does not posit a form of the solution. However, these earlier works focused on the persistence of steady states and did not perform constructions of asymptotic periodic solutions. Previous results on periodic solutions have been given by Lee, Forest, and Zhou[46] using a mesoscopic 2D tensor model. By invoking the Poincaré–Bendixson theorem which describes the long term behavior of orbits of continuous dynamical systems on the plane, they gave an elegantly simple proof that limit cycles ("tumbling orbits") must arise beyond the parameter boundary for the steady-unsteady transition.

The purpose of this paper is to revisit the well-studied elongation-perturbed shear problem from a different theoretical point of view. Our study will provide a detailed analysis on how perturbations of flow conditions affect the system dynamics at small flow rates. We will develop a multiscale analysis directly on the Smoluchowski equation of the kinetic theory. We thus extend results of Chillingworth et al.,[41] Vicente Alfonso et al.,[40] Lee et al.,[46] and Choate et al.[39] from a tensor model to the kinetic model. Our work differs from Refs. 25 and 36 in three parts: (1) Refs. 25 and 36 are based on numerical simulations of the 3D Smoluchowski equation, whereas ours is on the asymptotic analysis of the 2D Smoluchowski equation; (2) Refs. 25 and 36 include the effect of arbitrary aspect ratio, whereas our study is limited to the infinite aspect ratio; and (3) Refs. 25 and 36 concentrated on the persistence of steady states, whereas our focus is on both steady and unsteady solutions.

In this paper we limit our attention to the two-

dimensional case with a modified weak shear flow. More precisely, the Péclet number from shear flow (ε) is small and the Péclet number from elongation ($q \cdot \varepsilon$) is of the same order or smaller than that of the shear flow. Our restriction of the orientational probability density from a function on the sphere to the circle is mainly due to its mathematical simplicity, its possible relevance to experimental results on monolayer films of nematic polymers,[47,48] and its potential guidance to the full 3D orientational distributions. This kind of approach has been widely adopted by others.[33,34,46–53]

This paper is organized as follows: We give a brief description on the mathematical formulation of the Smoluchowski equation coupled with modified shear flow for nematic liquid crystalline polymers in Sec. II. We show the effect of elongational perturbations on the weak shear behavior in Sec. III. Finally, in Sec. IV we summarize our results.

II. MATHEMATICAL FORMULATION FOR THE TWO-DIMENSIONAL CASE

We briefly review the two-dimensional mathematical formulation of the Doi–Hess kinetic theory for homogeneous flows of rigid rodlike nematogenic molecules immersed in a viscous solvent subject to an imposed flow field.[4,5] First, we denote the orientation of each polymer rod by an angle θ. Then the orientational direction of a polymer rod is given by $\mathbf{u} = (\cos\theta, \sin\theta)$. To model the nematic tendency of the rodlike molecules to align with each other, it is common to use the short range Maier–Saupe potential,

$$
\begin{aligned}
V_{\mathrm{MS}} &= -2k_B T U \mathbf{u}\mathbf{u} : \langle \mathbf{u}\mathbf{u} \rangle \\
&= -k_B T U [\langle \cos 2\theta \rangle \cos 2\theta + \langle \sin 2\theta \rangle \sin 2\theta],
\end{aligned} \tag{1}
$$

where $\mathbf{u}\mathbf{u}$ and $\mathbf{A}:\mathbf{B}$ are defined as

$$
\mathbf{u}\mathbf{u} \equiv \begin{pmatrix} u_1 u_1 & u_1 u_2 \\ u_2 u_1 & u_2 u_2 \end{pmatrix},
$$

$$
\mathbf{A}:\mathbf{B} \equiv \sum_{i,j} a_{ij} b_{ij}. \tag{2}
$$

In Eq. (1) U is the normalized polymer concentration which describes the strength of intermolecular interactions, k_B is the Boltzmann constant, T is the absolute temperature, and $\langle \mathbf{u}\mathbf{u} \rangle$ is the second moment of the orientation distribution,

$$
\langle \mathbf{u}\mathbf{u} \rangle \equiv \int_{\|\mathbf{u}\|=1} \mathbf{u}\mathbf{u} \rho(\mathbf{u},\mathbf{x},t) d\mathbf{u}, \tag{3}
$$

where $\rho(\mathbf{u},\mathbf{x},t)$ is the orientational probability density function of the ensemble, i.e., the probability density that a polymer rod has direction $\mathbf{u}$ at location $\mathbf{x}$ and time t.

A shear flow with velocity

$$
\mathbf{v} = \gamma(0,-x) \tag{4}
$$

can be decomposed into the sum of a rotational flow $\mathbf{v}_R$ and an elongational flow $\mathbf{v}_E$,

$$
\mathbf{v} = \frac{\gamma}{2}(y,-x) + \frac{\gamma}{2}(-y,-x) \equiv \mathbf{v}_R + \mathbf{v}_E, \tag{5}
$$

where γ is the shear rate. For the elongational flow $\mathbf{v}_E = \gamma/2(-y,-x)$, we have

$$
\begin{aligned}
\nabla \mathbf{v}_E &= \frac{\gamma}{2} \begin{bmatrix} 0 & -1 \\ -1 & 0 \end{bmatrix} \\
&= \frac{\gamma}{2} \begin{bmatrix} \dfrac{\sqrt{2}}{2} & \dfrac{\sqrt{2}}{2} \\ -\dfrac{\sqrt{2}}{2} & \dfrac{\sqrt{2}}{2} \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \begin{bmatrix} \dfrac{\sqrt{2}}{2} & -\dfrac{\sqrt{2}}{2} \\ \dfrac{\sqrt{2}}{2} & \dfrac{\sqrt{2}}{2} \end{bmatrix}.
\end{aligned} \tag{6}
$$

So $\nabla \mathbf{v}_E$ has eigenvector $[\sqrt{2}/2, -\sqrt{2}/2]^T$ associated with the eigenvalue 1 and eigenvector $[\sqrt{2}/2, \sqrt{2}/2]^T$ associated with the eigenvalue -1.

The torque induced by the rotational flow $\mathbf{v}_R$ on a polymer rod is

$$
\mathbf{F}_R = \frac{k_B T}{Dr} \frac{\gamma}{2} = k_B T \frac{\varepsilon}{2}, \tag{7}
$$

where Dr is the rotational diffusivity of the polymer rod (assumed constant here), and $\varepsilon \equiv \gamma/Dr$ is the Péclet number of the shear flow. Similarly, the torque induced by the elongational flow $\mathbf{v}_E$ is

$$
\mathbf{F}_E = -\frac{k_B T}{Dr} \frac{\gamma}{2} \cos 2\theta = -k_B T \frac{\varepsilon}{2} \cos 2\theta. \tag{8}
$$

If we increase the elongation by a fraction of q, then the total elongation is

$$
\mathbf{v}_{E_T} = \frac{\gamma}{2}(1+q)(-y,-x) \tag{9}
$$

and the torque induced by the total elongation is

$$
\mathbf{F}_{E_T} = -k_B T \frac{\varepsilon}{2}(1+q)\cos 2\theta. \tag{10}
$$

Figure 1 depicts velocity fields of a shear flow and an elongation-perturbed shear flow, respectively.

If we assume the polymer orientation distribution is homogeneous in space, then the orientational pdf under an imposed perturbed shear evolves according to the Smoluchowski equation,[4]

$$
\frac{\partial \rho(\theta,t)}{\partial t} = D_r \frac{\partial}{\partial \theta} \left\{ [V'_{\mathrm{PS}}(\theta) + V'_{\mathrm{MS}}(\theta)]\rho + \frac{\partial \rho}{\partial \theta} \right\}, \tag{11}
$$

where $V_{\mathrm{MS}}(\theta)$ is the Maier–Saupe potential (1) and $V_{\mathrm{PS}}(\theta)$ is the potential due to the external field (i.e., perturbed shear flow)

$$
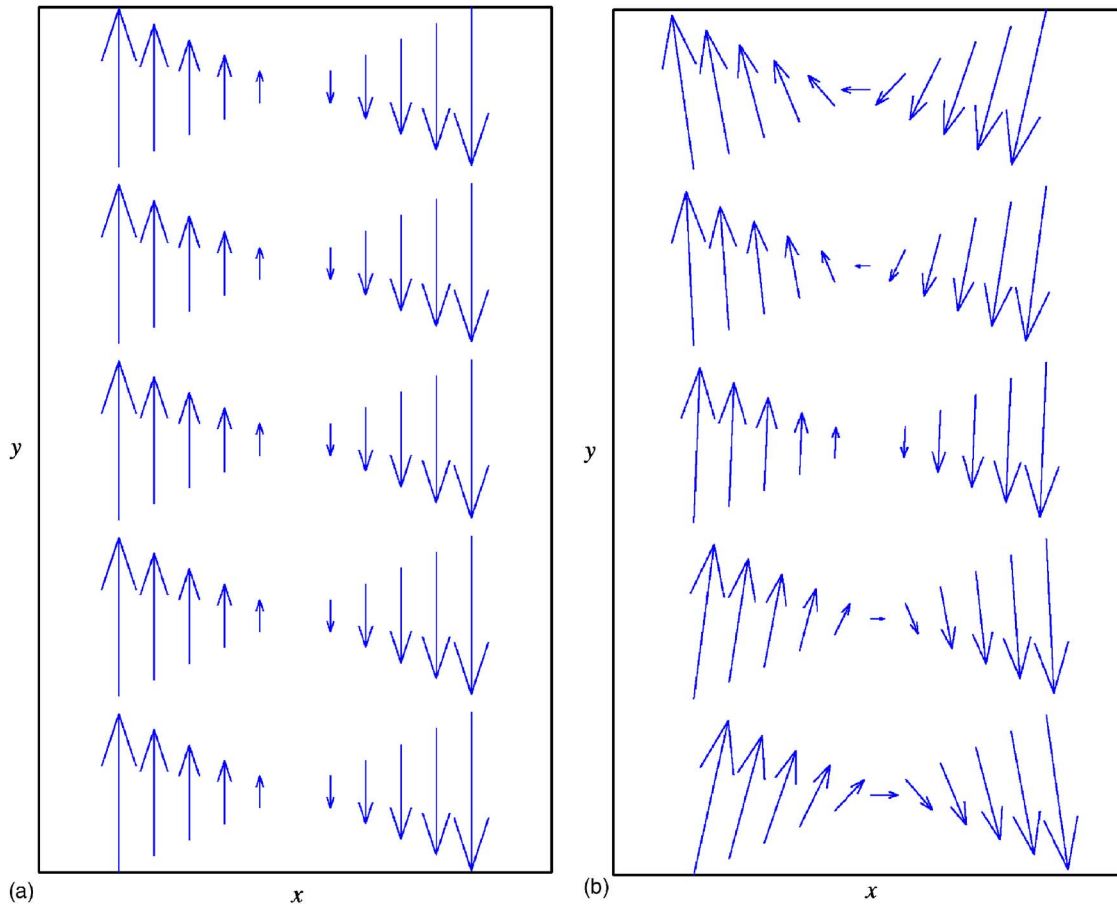V_{\mathrm{PS}}(\theta) = -\frac{\varepsilon}{2}\theta + \frac{\varepsilon}{4}(1+q)\sin 2\theta. \tag{12}
$$

FIG. 1. (Color online) Velocity field of (a) a shear flow ($q=0$) and (b) a shear flow plus a 20% elongational perturbation ($q=0.2$).

Here, for simplicity, all potentials have been normalized by $k_B T$. In Eq. (11), we can set $D_r=1$ by introducing the nondimensionalized time $t_{new}=tD_r$. Below we are going to do multiscale asymptotic analysis on (11) with $D_r=1$.

III. MULTISCALE ASYMPTOTIC ANALYSIS

We consider Eq. (11) for small ε which allows us to carry out a multiscale asymptotic analysis. First, we simplify the expression of the Maier–Saupe potential (1) by selecting an angle α so that $\langle \sin 2(\theta-\alpha)\rangle=0$. Then the Maier–Saupe potential can be written in a compact form,

$$V_{MS}(\theta) = -U\langle\cos 2(\theta-\alpha)\rangle\cos 2(\theta-\alpha). \quad (13)$$

We restrict our application of the multiple scale method to the introduction of two time scales, $T_0=t$ and $T_1=\varepsilon\cdot t$. Assume that ρ possesses an expansion of the form

$$\rho(\theta,t) = \rho^{(0)}(\theta,T_0,T_1) + \varepsilon\rho^{(1)}(\theta,T_0,T_1) + O(\varepsilon^2) \quad (14)$$

and substitute it into Eq. (11) to obtain the leading order equation

$$\frac{\partial\rho^{(0)}}{\partial T_0} = \frac{\partial}{\partial\theta}\left[V_{MS}^{(0)'}(\theta)\rho^{(0)} + \frac{\partial}{\partial\theta}\rho^{(0)}\right]. \quad (15)$$

Here $V_{MS}^{(0)}(\theta)=-U\langle\cos 2(\theta-\alpha)\rangle^{(0)}\cos 2(\theta-\alpha)$ corresponds to the leading order term of the Maier–Saupe potential and the average in the expression is taken with respect to the pdf

$\rho^{(0)}(\theta)$. Since Eq. (15) does not contain an external field effect, we have a pure nematic equation at the leading order. Also notice that Eq. (15) does not contain time scale T_1. As a result, the orientation distribution will converge to a steady state with respect to time scale T_0 (it may not be a steady state with respect to time scale T_1). The steady state pdf of Eq. (15) is given by the Boltzmann distribution,

$$\rho^{(0)}(\theta,T_0,T_1) = \frac{\exp[r\cos 2(\theta-\alpha)]}{\int_0^{2\pi}\exp[r\cos 2(\theta-\alpha)]d\theta}, \quad (16)$$

where r is defined as

$$r = U\langle\cos 2(\theta-\alpha)\rangle^{(0)}. \quad (17)$$

Equation (17) is also the equation for determining r for a given value of normalized polymer concentration U in the absence of flow. In the case of weak shear plus weak elongational perturbation, the leading term of r satisfies Eq. (17). We consider the simple situation where the polymer orientation starts with the steady state pdf (16). Then Eq. (15) implies that the pdf remains unchanged with respect to T_0. Mathematically, this means

$$\frac{\partial\rho^{(0)}(\theta,T_0,T_1)}{\partial T_0} = 0. \quad (18)$$

So $\rho^{(0)}(\theta,T_0,T_1)$ is independent of T_0 and thereby "quasi-steady." As a result, the phase angle α is also independent of

T_0. But α may depend on T_1 and this corresponds to the slow time evolution invoked by the imposed flow field. To find $\alpha(T_1)$, consider the expansion of $\partial\rho(\theta,t)/\partial t$,

$$\frac{\partial\rho(\theta,t)}{\partial t} = \frac{\partial\rho^{(0)}(\theta,T_0,T_1)}{\partial T_0} + \varepsilon\left[\frac{\partial\rho^{(0)}(\theta,T_0,T_1)}{\partial T_1}\right.$$
$$\left. + \frac{\partial\rho^{(1)}(\theta,T_0,T_1)}{\partial T_0}\right] + \cdots. \qquad (19)$$

Both our extensive numerical simulations and the physical intuitions suggest that $\rho^{(1)}(\theta,T_0,T_1)$ is independent of time scale T_0. Although a rigorous mathematical proof is still an open problem, here in our multiscale expansion, we adopt the assumption that $\rho^{(1)}(\theta,T_0,T_1)$ is quasisteady. With this assumption, Eq. (19) becomes

$$\frac{\partial\rho(\theta,t)}{\partial t} = \varepsilon\frac{\partial\rho^{(0)}(\theta,T_0,T_1)}{\partial T_1} + O(\varepsilon^2)$$
$$= -\varepsilon\frac{\partial\rho^{(0)}(\theta,T_0,T_1)}{\partial\theta}\cdot\frac{d\alpha(T_1)}{dT_1} + O(\varepsilon^2). \qquad (20)$$

On substituting this into Eq. (11) and rearranging terms we obtain

$$\frac{\partial}{\partial\theta}\left[\left(-\frac{\varepsilon}{2} + \frac{\varepsilon}{2}(1+q)\cos 2\theta + V'_{MS}(\theta)\right)\rho(\theta) + \frac{\partial\rho(\theta)}{\partial\theta}\right.$$
$$\left. + \varepsilon\rho^{(0)}(\theta)\frac{d\alpha(T_1)}{dT_1} + O(\varepsilon^2)\right] = 0. \qquad (21)$$

This suggests

$$\left[-\frac{\varepsilon}{2} + \frac{\varepsilon}{2}(1+q)\cos 2\theta + V'_{MS}(\theta)\right]\rho(\theta) + \frac{\partial\rho(\theta)}{\partial\theta} + \varepsilon\rho^{(0)}$$
$$\times(\theta)\frac{d\alpha(T_1)}{dT_1} + O(\varepsilon^2) = -\varepsilon J_1, \qquad (22)$$

where εJ_1 is the integration constant of the order $O(\varepsilon)$. In Eq. (22) both $d\alpha(T_1)/dT_1$ and J_1 are unknown so we need two linear equations to determine them. To do so, we first integrate Eq. (22) from 0 to 2π to obtain

$$-\frac{\varepsilon}{2} + \frac{\varepsilon}{2}(1+q)\int_0^{2\pi}\cos 2\theta\rho(\theta)d\theta + \int_0^{2\pi}V'_{MS}(\theta)\rho(\theta)d\theta$$
$$+ \varepsilon\frac{d\alpha(T_1)}{dT_1} + O(\varepsilon^2) = -2\pi\varepsilon J_1. \qquad (23)$$

The second term on the left of Eq. (23) has the expression

$$\int_0^{2\pi}\cos 2\theta\rho(\theta)d\theta = \langle\cos 2\theta\rangle$$
$$= \langle\cos 2(\theta-\alpha)\cos 2\alpha$$
$$\quad - \sin 2(\theta-\alpha)\sin 2\alpha\rangle$$
$$= \langle\cos 2(\theta-\alpha)\cos 2\alpha\rangle$$
$$= \langle\cos 2(\theta-\alpha)\rangle^{(0)}\cos 2\alpha + O(\varepsilon)$$
$$= \frac{r}{U}\cos 2\alpha + O(\varepsilon), \qquad (24)$$

where $r/U \equiv \langle\cos 2(\theta-\alpha)\rangle^{(0)}$ is the order parameter. The third term on the left of Eq. (23) has the expression

$$\int_0^{2\pi}V'_{MS}(\theta)\rho(\theta)d\theta = \int_0^{2\pi}\frac{d}{d\theta}[-r\cos 2(\theta-\alpha)]\rho(\theta)d\theta$$
$$= \int_0^{2\pi}2r\sin 2(\theta-\alpha)\rho(\theta)d\theta$$
$$= 2r\langle\sin 2(\theta-\alpha)\rangle = 0, \qquad (25)$$

where we have used the fact that the angle α is selected to make $\langle\sin 2(\theta-\alpha)\rangle = 0$. With these expressions, Eq. (23) becomes

$$-\frac{1}{2} + \frac{1+q}{2}\frac{r}{U}\cos[2\alpha(T_1)] + \frac{d\alpha(T_1)}{dT_1} + O(\varepsilon) = -2\pi J_1. \qquad (26)$$

To find a second equation for $d\alpha(T_1)/dT_1$ and J_1, we manipulate Eq. (22) by dividing it by $\varepsilon\rho(\theta)$, approximating $\rho(\theta)$ by $\rho^{(0)}(\theta)$, and integrating from 0 to 2π to obtain

$$\int_0^{2\pi}\left(-\frac{1}{2}\right)d\theta + \int_0^{2\pi}\frac{1}{2}(1+q)\cos 2\theta d\theta + \frac{1}{\varepsilon}\int_0^{2\pi}V'_{MS}d\theta$$
$$+ \frac{1}{\varepsilon}\int_0^{2\pi}\frac{1}{\rho^{(0)}(\theta)}\frac{\partial\rho^{(0)}(\theta)}{\partial\theta}d\theta + \int_0^{2\pi}\frac{d\alpha(T_1)}{dT_1}d\theta + O(\varepsilon)$$
$$= -\int_0^{2\pi}\frac{J_1}{\rho^{(0)}(\theta)}d\theta. \qquad (27)$$

Note that the integrands $\cos 2\theta$, V'_{MS}, and $1/\rho^{(0)}(\theta)\partial\rho^{(0)}$ $\times(\theta)/\partial\theta = \partial\ln[\rho^{(0)}(\theta)]/\partial\theta$ are all periodic on $[0,2\pi]$, so the second, third, and fourth definite integrals in Eq. (27) vanish. Using Eq. (16) and carrying out the details, we get

$$-\pi + 2\pi\frac{d\alpha(T_1)}{dT_1} + O(\varepsilon) = -(2\pi)^2 g(r)J_1, \qquad (28)$$

where the function $g(r)$ is defined as

$$g(r) \equiv \left[\frac{1}{2\pi}\int_0^{2\pi}\exp(r\cos 2\theta)d\theta\right]^2. \qquad (29)$$

Multiplying Eq. (26) by a factor $2\pi g(r)$ and subtracting it from Eq. (28) leads to

$$-\pi + 2\pi\frac{d\alpha(T_1)}{dT_1} = 2\pi g(r)\left[-\frac{1}{2} + \frac{1+q}{2}\frac{r}{U}\cos[2\alpha(T_1)]\right.$$
$$\left. + \frac{d\alpha(T_1)}{dT_1}\right]. \qquad (30)$$

Solving for $d\alpha(T_1)/dT_1$ yields

$$\frac{d\alpha(T_1)}{dT_1} = \frac{1}{2[g(r)-1]}\left[-1 + g(r)\left(1 - \frac{(1+q)r}{U}\right.\right.$$
$$\left.\left.\times\cos[2\alpha(T_1)]\right)\right]. \qquad (31)$$

Applying the double angle formula $\cos[2\alpha(T_1)] = 1 - 2\sin^2[\alpha(T_1)]$, we obtain after some manipulations

$$\frac{d\alpha(T_1)}{dT_1} = c_1 \cdot \{\sin^2[\alpha(T_1)] + c_2\}, \tag{32}$$

where c_1 and c_2 are

$$c_1 = \frac{g(r)}{g(r)-1} \cdot \frac{r}{U} \cdot (1+q),$$

$$c_2 = \frac{-1}{2(1+q)}[p(r)+q]. \tag{33}$$

Here the function $p(r)$ is defined as

$$p(r) \equiv \frac{U}{r}\left[\frac{1}{g(r)} - 1\right] + 1. \tag{34}$$

Note that in the order parameter $r/U \equiv \langle \cos 2(\theta - \alpha)\rangle^{(0)}$, probability density (16) is used in calculating the average. As a function of r, r/U has the expression

$$\frac{r}{U} = \frac{\int_0^{2\pi} \cos 2\theta \exp(r\cos 2\theta)d\theta}{\int_0^{2\pi} \exp(r\cos 2\theta)d\theta}. \tag{35}$$

As a matter of fact Eq. (35) is the equation for determining r for a given value of normalized polymer concentration U. Here for mathematical convenience, we use r (instead of U) as the independent variable. Note that, for $r > 0$, the mapping between r and U as given in Eq. (35) is monotonic (see Appendix A for a mathematical derivation of the monotonicity). As a consequence of the monotonicity, for $r > 0$, the mapping between r and U is one-to-one. Therefore, a result in terms of r can easily be translated to the corresponding result in terms of U. Below we will first discuss the dynamic behavior of $\alpha(T_1)$ governed by Eq. (32) for various regions of r. Then all conclusions will be restated in terms of U, the normalized polymer concentration.

Equation (32) describes the dynamics of the slow time evolution of the phase angle $\alpha(T_1)$. In Eq. (32), the factor c_1 is always positive for $r > 0$. If c_2 is negative (and $|c_2| < 1$), then the phase angle $\alpha(T_1)$ will stop at an angle α_0 satisfying $\sin^2 \alpha_0 + c_2 = 0$. In other words, the polymer orientation distribution will converge to a steady state. If c_2 is positive, then the phase angle $\alpha(T_1)$ will keep increasing and the polymer orientation distribution will rotate periodically in time (tumbling). Thus, positive c_2 causes tumbling. The sign of c_2 is the opposite of the sign of $p(r)+q$. It follows that negative value of $p(r)+q$ corresponds to tumbling. Figure 2 shows the graph of function $p(r)$ and its asymptotic approximation as $r \to +\infty$,

$$p(r) = \frac{-1}{2r}\left(1 + \frac{3}{4r}\right) + \cdots \quad \text{as } r \to +\infty. \tag{36}$$

The asymptotic approximation of $p(r)$ as $r \to +\infty$ is derived in Appendix B. As shown in Fig. 2, the function $p(r)$ starts at $r = 0$ with $p(0) = 1$. As r increases from 0 to $r_{\min}$, the function $p(r)$ decreases monotonically. In particular, the function $p(r)$ intersects with the x-axis at $r_0 = 1.3276$. At $r_{\min} = 2.6463$, the function $p(r)$ attains its minimum, and the minimum value is $\min_r p(r) = -0.18754$. For $r > r_{\min}$, function $p(r)$ increases monotonically. For $r > r_{\min}$, function $p(r)$ is well approxi-
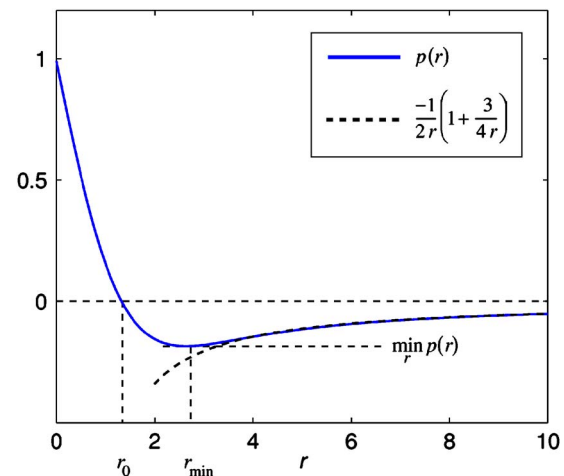

FIG. 2. (Color online) Graph of function $p(r)$ and its asymptotic approximation as $r \to +\infty$. The function $p(r)$ intersects with the x-axis at $r_0 = 1.3276$. At $r_{\min} = 2.6463$, the function $p(r)$ attains its minimum, and the minimum value is $\min_r p(r) = -0.18754$.

mated by its asymptotic expression at $r = +\infty$.

Figure 3(a) shows the tumbling region (shaded) in the (q, r) space. Here the tumbling region is the largest region where $p(r) + q$ is negative. Recall from our discussion above that negative value of $p(r) + q$ corresponds to tumbling. So if the value of (q, r) falls in the tumbling region, then the polymer orientation rotates periodically in time (tumbling). From the asymptotic expression of function $p(r)$ for large r, we see that for small q, the upper boundary of the tumbling region in the (q, r) space is approximated by

$$q = \frac{1}{2r}\left(1 + \frac{3}{4r}\right). \tag{37}$$

Solving r in terms of q, we obtain

$$r_{\text{upper}} = \frac{1 + \sqrt{1 + 6q}}{4q}. \tag{38}$$

This asymptotic expression is valid for small q.

Because of the one-to-one correspondence between r and U, a region in the (q, r) space can be uniquely mapped to a region in the (q, U) space. Figure 3(b) shows the corresponding tumbling region in the (q, U) space, where U is the normalized polymer concentration and q is the magnitude of the elongational perturbation relative to the original elongational component of the shear flow. Again, for small q (corresponding to large r and large U), the upper boundary of the tumbling region in the (q, U) space can be well approximated by using the asymptotic expression of function $p(U)$ for large U (which is derived and presented in Appendix B). For small q, the upper boundary in U satisfies approximately

$$q = \frac{1}{2U}\left(1 + \frac{5}{4U}\right). \tag{39}$$
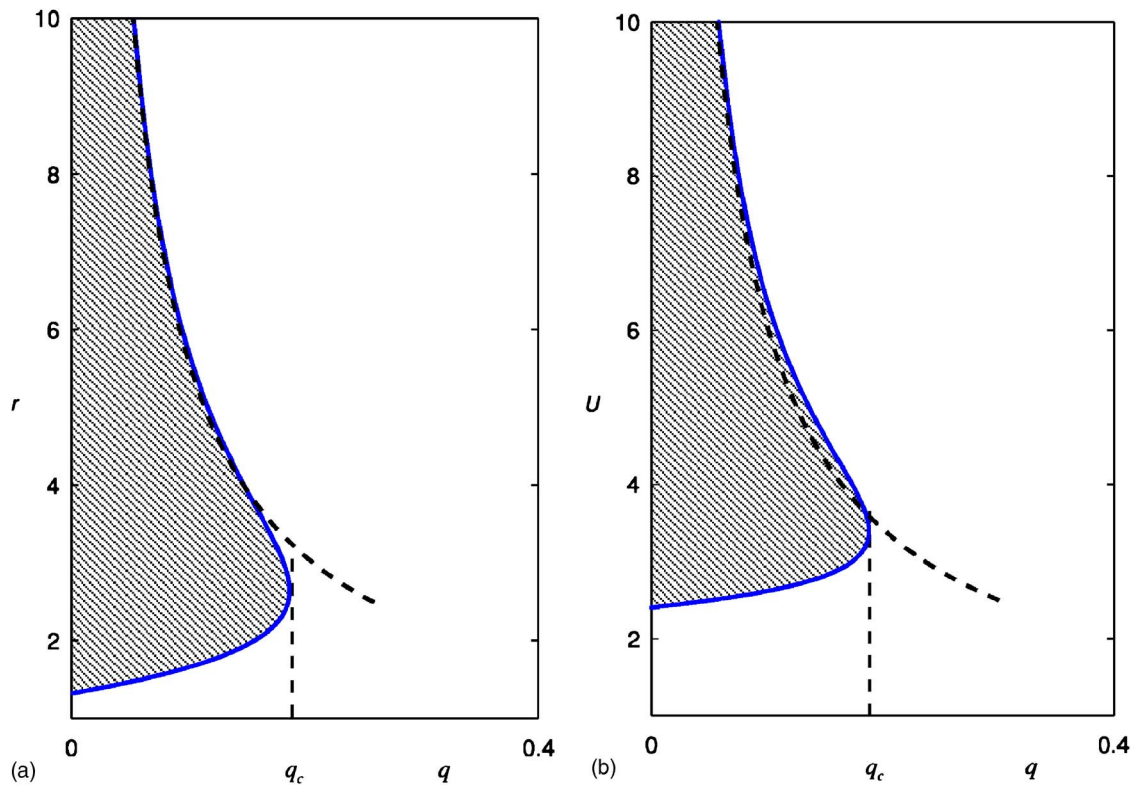
Solving U in terms of q, we obtain

FIG. 3. (Color online) Tumbling regions are shown as shaded. (a) Tumbling region in the (q,r) space. (b) Tumbling region in the (q,U) space. The critical value for q is $q_c=0.18754$.

$$U_{\text{upper}} = \frac{1+\sqrt{1+10q}}{4q}. \tag{40}$$

This asymptotic expression is valid for small q. In Fig. 3, the asymptotic approximation of the upper bound of the tumbling region for small q is shown as a dashed line and it agrees well with the exact solution.

Let $q_c \equiv -\min_r p(r) = 0.18154$. q_c is the critical value of q, when the $q>q_c$ function $p(r)+q$ is positive for all values of $r>0$; when $0<q<q_c$ the function $p(r)+q$ may be negative for certain values of r. As shown in Fig. 3, when $0<q<q_c$ tumbling occurs only for r (U) in a finite interval. As q increases toward the critical value q_c, this tumbling interval gets smaller and smaller. When $q>q_c$, the tumbling interval disappears and there is no tumbling. That is, with an elongational perturbation of less than 20% ($q_c=0.18154$) of the original elongational component of the shear flow, the tumbling disappears for all values of U.

IV. CONCLUSIONS

We have applied the multiscale asymptotic analysis to study the effect of elongational perturbations on the weak shear behavior of nematic liquid crystal polymers. It is found that when the elongational perturbation is small, the tumbling behavior occurs only in an intermediate range of polymer concentrations; when the elongational perturbation is above about 20% of the shear rate, tumbling disappears for all polymer concentrations. These findings suggest a mechanism on the control of the structure of nematic liquid crystal polymers.

APPENDIX A: MONOTONICITY OF $U(r)$

We first apply integration by parts to the numerator on the right-hand side of Eq. (35),

$$\int_0^{2\pi} \cos 2\theta \exp(r\cos 2\theta)d\theta$$

$$= \frac{1}{2}\int_0^{2\pi} \exp(r\cos 2\theta)d(\sin 2\theta)$$

$$= r\int_0^{2\pi} \sin^2 2\theta \exp(r\cos 2\theta)d\theta$$

$$= r\int_0^{2\pi} (1-\cos^2 2\theta)\exp(r\cos 2\theta)d\theta. \tag{A1}$$

Substituting this result into Eq. (35) and writing U as a function of r, we have

$$\frac{1}{U(r)} = 1 - \frac{\int_0^{2\pi} \cos^2 2\theta \exp(r \cos 2\theta) d\theta}{\int_0^{2\pi} \exp(r \cos \theta) d\theta}. \tag{A2}$$

To show that $U(r)$ is monotonically increasing, we only need to show

$$w(r) \equiv \frac{\int_0^{2\pi} \cos^2 2\theta \exp(r \cos 2\theta) d\theta}{\int_0^{2\pi} \exp(r \cos 2\theta) d\theta} \tag{A3}$$

is monotonically increasing. Function $w(r)$ can be viewed as an average, $w(r) = \langle \cos^2 2\theta \rangle$ with probability density

$$\rho(\theta, r) = \frac{\exp(r \cos 2\theta)}{\int_0^{2\pi} \exp(r \cos 2\theta) d\theta}. \tag{A4}$$

The derivative of the probability density with respect to r is

$$\frac{d\rho(\theta, r)}{dr} = (\cos 2\theta - \langle \cos 2\theta \rangle)\rho(\theta, r). \tag{A5}$$

The first derivative of $w(r)$ is

$$w'(r) = \langle \cos^2 2\theta (\cos 2\theta - \langle \cos 2\theta \rangle) \rangle. \tag{A6}$$

The second derivative of $w(r)$ is

$$\begin{aligned} w''(r) &= \langle \cos^2 2\theta (\cos 2\theta - \langle \cos 2\theta \rangle)^2 \rangle \\ &\quad - \langle \cos^2 2\theta \rangle \cdot (\langle \cos^2 2\theta \rangle - \langle \cos 2\theta \rangle^2) \\ &= -2\langle \cos 2\theta \rangle w'(r) + \mathrm{var}(\cos^2 2\theta). \end{aligned} \tag{A7}$$

Thus, function $w(r)$ satisfies the special property that whenever $w'(r) = 0$ we have $w''(r) > 0$. At $r = 0$, we have $w(0) = 0.5$ and $w'(0) = 0$. The special property of $w(r)$ guarantees that $w'(r) > 0$ for $r > 0$. This leads immediately to the conclusion that $U(r)$ is monotonically increasing.

APPENDIX B: ASYMPTOTIC APPROXIMATION OF $p(r)$

To derive an asymptotic approximation for $p(r)$ as $r \to +\infty$, we need to deal with integrals such as $\int_0^{2\pi} \exp(r \cos 2\theta) d\theta$. First, we use the symmetry of $\cos 2\theta$ to rewrite the integral as

$$\int_0^{2\pi} \exp(r \cos 2\theta) d\theta = 4 \int_0^{\pi/2} \exp(r \cos 2\theta) d\theta. \tag{B1}$$

To apply Watson's lemma[38] on the integral, we use a change of variable,

$$s = (1 - \cos 2\theta)/2,$$

$$ds = \sin 2\theta d\theta = 2 \sin \theta \cos \theta d\theta = 2\sqrt{s}\sqrt{1 - s}\, d\theta, \tag{B2}$$

$$d\theta = \frac{1}{2} \frac{1}{\sqrt{s}\sqrt{1 - s}} ds.$$

Applying the change of variables to the integral yields

$$\int_0^{2\pi} \exp(r \cos 2\theta) d\theta$$

$$= 2 \int_0^1 \exp[r(1 - 2s)] \frac{1}{\sqrt{s}\sqrt{1 - s}} ds$$

$$= 2 \exp(r) \int_0^1 \exp(-2rs) \frac{1}{\sqrt{s}} \left(1 + \frac{1}{2}s + \cdots\right) ds$$

$$= 2 \exp(r) \left(\frac{\Gamma(1/2)}{\sqrt{2r}} + \frac{1}{2} \cdot \frac{\Gamma(3/2)}{2r\sqrt{2r}} + \cdots\right)$$

$$= \frac{\sqrt{2\pi}}{\sqrt{r}} \exp(r) \left(1 + \frac{1}{8} \cdot \frac{1}{r} + \cdots\right). \tag{B3}$$

Substituting this result into function $g(r)$, we have

$$g(r) = \left(\frac{1}{2\pi} \int_0^{2\pi} \exp(r \cos 2\theta) d\theta\right)^2$$

$$= \frac{1}{2\pi} \cdot \frac{\exp(r)}{r} (1 + \cdots). \tag{B4}$$

That is, as $r \to +\infty$, function $g(r)$ is exponentially large and consequently $1/g(r)$ is exponentially small. In a similar way, we obtain

$$\int_0^{2\pi} (1 - \cos 2\theta) \exp(r \cos 2\theta) d\theta$$

$$= 4 \int_0^1 \exp[r(1 - 2s)] \frac{s}{\sqrt{s}\sqrt{1 - s}} ds$$

$$= 4 \exp(r) \int_0^1 \exp(-2rs) \sqrt{s} \left(1 + \frac{1}{2}s + \cdots\right) ds$$

$$= 4 \exp(r) \left(\frac{\Gamma(3/2)}{2r\sqrt{2r}} + \frac{1}{2} \cdot \frac{\Gamma(5/2)}{(2r)^2\sqrt{2r}} + \cdots\right)$$

$$= \frac{\sqrt{2\pi}}{\sqrt{r}} \exp(r) \left(\frac{1}{2} \cdot \frac{1}{r} + \frac{3}{16} \cdot \frac{1}{r^2} + \cdots\right). \tag{B5}$$

Combining Eqs. (B3) and (B5) gives us

$$\begin{aligned} 1 - \langle \cos 2\theta \rangle &= \frac{\int_0^{2\pi}(1 - \cos 2\theta)\exp(r\cos 2\theta)d\theta}{\int_0^{2\pi}\exp(r\cos 2\theta)d\theta} \\ &= \frac{\dfrac{1}{2}\cdot\dfrac{1}{r} + \dfrac{3}{16}\cdot\dfrac{1}{r^2} + \cdots}{1 + \dfrac{1}{8}\cdot\dfrac{1}{r} + \cdots} = \frac{1}{2r}\left(1 + \frac{1}{4r} + \cdots\right) \end{aligned} \tag{B6}$$

and

$$\langle \cos 2\theta \rangle = 1 - (1 - \langle \cos 2\theta \rangle) = 1 - \frac{1}{2r} + \cdots. \tag{B7}$$

To find an asymptotic approximation for function $p(r)$, we rewrite it as

$$p(r) = \frac{1}{\langle\cos 2\theta\rangle}\left(\frac{1}{g(r)} - 1\right) + 1$$

$$= \frac{1}{\langle\cos 2\theta\rangle} \cdot \frac{1}{g(r)} - \frac{1 - \langle\cos 2\theta\rangle}{\langle\cos 2\theta\rangle}. \quad \text{(B8)}$$

The first term on the right-hand side is exponentially small. The second term can be expanded using results (B6) and (B7) obtained above,

$$p(r) = -\frac{1 - \langle\cos 2\theta\rangle}{\langle\cos 2\theta\rangle} + \cdots = -\frac{\frac{1}{2r}\left(1 + \frac{1}{4r} + \cdots\right)}{1 - \frac{1}{2r} + \cdots} + \cdots$$

$$= \frac{-1}{2r}\left(1 + \frac{3}{4r} + \cdots\right). \quad \text{(B9)}$$

Now we derive the expansion of function $p(r)$ in terms of U. As $r \to +\infty$, U, as a function of r, has the expansion

$$U(r) = \frac{r}{\langle\cos 2\theta\rangle} = r\left(1 + \frac{1}{2r}\right) + \cdots. \quad \text{(B10)}$$

Expressing r in terms of U yields

$$r(U) = U\left(1 - \frac{1}{2U}\right) + \cdots. \quad \text{(B11)}$$

Substituting into Eq. (B7), we arrive at

$$p(U) = p(r(U)) = \frac{-1}{2r(U)}\left(1 + \frac{3}{4r(U)} + \cdots\right)$$

$$= \frac{-1}{2U}\left(1 + \frac{5}{4U} + \cdots\right). \quad \text{(B12)}$$

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