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Research Article

On the Equilibria of the Extended Nematic Polymers under Elongational Flow

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We classify the equilibrium solutions of the Smoluchowski equation for dipolar (extended) rigid nematic polymers under imposed elongational flow. The Smoluchowski equation couples the Maier-Saupe short-range interaction, dipole-dipole interaction, and an external elongational flow. We show that all stable equilibria of rigid, dipolar rod dispersions under imposed uniaxial elongational flow field are axisymmetric. This finding of axisymmetry significantly simplifies any procedure of obtaining experimentally observable equilibria.

1. Introduction

Nematic liquid crystal polymers are viscoelastic anisotropic materials that have many important applications [1]. The dynamic behavior of rigid rod nematic liquid crystal polymers is modeled by the Smoluchowski equation [2–4]. Analytical results on pure nematic equilibria have been obtained in a series of papers [5–11]. In [12] a 2D Smoluchowski equation under weak shear is analyzed. In [13] a coplanar magnetic field is coupled in the Smoluchowski equation to investigate the monodomain dynamics for rigid rod and platelet suspensions. However, dipole-dipole interaction is not included in [13]. Recently, the authors [14, 15] studied kinetic equilibria of rigid, dipolar rod ensembles for coupled dipole-dipole and Maier-Saupe short-range potentials. This work is a natural extension of [14, 15] to include an external elongational flow. For completeness and for reader's convenience, we include all the lemmas and theorems necessary for reaching the conclusions even though some of the lemmas and theorems have appeared in previous works.

This paper is organized as follows. We first briefly give the mathematical formulation of the Smoluchowski equation for extended (polar) nematics in Section 2. Our main theoretical results of the equilibrium solutions of the Smoluchowski equation are presented in Sections 3, 4, and 5. More specifically, we first show that the first moment of an equilibrium solution must be aligned with one of the principal axes of the second moment. Then in Section 4 we exploit free energy to show that an equilibrium solution whose first moment is not parallel to the imposed external elongational flow field is unstable. Finally in Section 5 we prove the most important result of this paper: all stable equilibrium solutions are axisymmetric. We give concluding remarks in Section 6.

2. The Smoluchowski equation for extended (polar) nematics

In this paper, we study equilibrium solutions of the Smoluchowski equation for rigid extended (polar) nematics under elongational flow. Here the nematic molecules are magnetically polar. For these extended nematics, the molecular interaction includes both the dipole-dipole interaction and the short-range Maier-Saupe interaction. We assume the system has an imposed elongational flow field. The potential due to the external elongational field is given by [16],

$$V_e = -\frac{\alpha_0}{2} kT\mathbf{EE} : \mathbf{mm}, \tag{2.1}$$

where $\mathbf{E}$ is the direction of the elongation, $\alpha_0 > 0$ corresponds to the stretching in the $\mathbf{E}$ direction (*uniaxial elongation*) and $\alpha_0 < 0$ corresponds to compressing, k is the Boltzmann constant, and T is the absolute temperature.

Now let $\rho(\mathbf{m}, t)$ denote the probability density function (pdf) for dipolar rod-like nematic molecules in unit direction $\mathbf{m}$ at time t. The dynamic evolution of the orientational pdf for the ensembles of rigid rods with inherent dipoles is governed by the Smoluchowski equation [3]:

$$\frac{\partial \rho}{\partial t} = D \frac{\partial}{\partial \mathbf{m}} \cdot \left(\frac{1}{kT} \frac{\partial U}{\partial \mathbf{m}} \rho + \frac{\partial \rho}{\partial \mathbf{m}} \right). \tag{2.2}$$

Here $\partial/\partial\mathbf{m}$ represents the orientational gradient operator [17], and the total potential is given by

$$U(\mathbf{m}) = -\alpha kT \langle \mathbf{m} \rangle \cdot \mathbf{m} - bkT \langle \mathbf{mm} \rangle : \mathbf{mm} - \frac{\alpha_0}{2} kT\mathbf{EE} : \mathbf{mm}, \tag{2.3}$$

where α is the strength of the dipole-dipole interaction, b denotes the strength of the Maier-Saupe short-range interaction, and

$$\langle (\bullet) \rangle = \int_{\|\mathbf{m}\|=1} (\bullet) \rho \, d\mathbf{m} \tag{2.4}$$

is the ensemble average with respect to the pdf ρ, which is a solution of the Smoluchowski equation (2.2). For simplicity, from now on we assume $kT = 1$, or equivalently we assume that all energies are normalized by kT.

The total potential (2.3) can be split into the sum of the internal potential representing the interaction among polymer rods with themselves and the external potential:

$$U(\mathbf{m}) = U_{\text{int}}(\mathbf{m}) + U_{\text{ext}}(\mathbf{m}), \tag{2.5}$$

where the internal and external potentials are given by

$$\begin{aligned} U_{\text{ext}}(\mathbf{m}) &= -\frac{\alpha_0}{2}\mathbf{EE}:\mathbf{mm}, \\ U_{\text{int}}(\mathbf{m}) &= U_1(\mathbf{m}) + U_2(\mathbf{m}), \\ U_1(\mathbf{m}) &= -\alpha\langle\mathbf{m}\rangle\cdot\mathbf{m}, \\ U_2(\mathbf{m}) &= -b\langle\mathbf{mm}\rangle:\mathbf{mm}. \end{aligned} \tag{2.6}$$

In this paper, we restrict our study to the case of $\alpha > 0$, $\alpha_0 > 0$, and $b > 0$. In particular, $\alpha_0 > 0$ (uniaxial elongation) will help us eliminate many unstable equilibrium solutions.

3. Equilibrium solutions for extended nematics

An equilibrium solution of (2.2) is given by the Boltzmann distribution [3]

$$\rho_{\text{eq}}(\mathbf{m}) = \frac{1}{Z}\exp[-U(\mathbf{m})], \qquad Z = \int_S \exp[-U(\mathbf{m})]d\mathbf{m}. \tag{3.1}$$

The nonlinear integral equation for the first moment vector $\langle\mathbf{m}\rangle$ is

$$\langle\mathbf{m}\rangle = \int_S \mathbf{m}\rho_{\text{eq}}(\mathbf{m})d\mathbf{m}, \tag{3.2}$$

whereas the nonlinear integral equation for the second moment tensor $\langle\mathbf{mm}\rangle$ is

$$\langle\mathbf{mm}\rangle = \int_S \mathbf{mm}\rho_{\text{eq}}(\mathbf{m})d\mathbf{m}. \tag{3.3}$$

We establish the coordinate system as follows. We select the uniaxial elongation field $\mathbf{E}$ as the z-axis. We select the x-axis and the y-axis to be perpendicular to the z-axis but otherwise arbitrary. In this Cartesian coordinate system, we have

$$\begin{gathered} \mathbf{m} = (m_1, m_2, m_3), \qquad \mathbf{E} = (0,0,1), \qquad \langle\mathbf{m}\rangle = (q_1, q_2, q_3), \\ U(\mathbf{m}) = -\alpha(q_1m_1 + q_2m_2 + q_3m_3) - b\langle\mathbf{mm}\rangle:\mathbf{mm} - \frac{\alpha_0}{2}m_3^2. \end{gathered} \tag{3.4}$$

The nonlinear equations for the first and second moments become

$$\begin{aligned} q_i = \langle m_i\rangle &= \int_S m_i\rho_{\text{eq}}(\mathbf{m})d\mathbf{m}, \\ \langle\mathbf{mm}\rangle_{ij} = \langle m_im_j\rangle &= \int_S m_im_j\rho_{\text{eq}}(\mathbf{m})d\mathbf{m}. \end{aligned} \tag{3.5}$$

We select the spherical coordinate system (ϕ,θ) using the y-axis as the pole. In this spherical coordinate system, we have

$$(m_1, m_2, m_3) = (\sin\phi\sin\theta, \cos\phi, \sin\phi\cos\theta). \tag{3.6}$$

Theorem 3.1. *For an equilibrium probability density, U_{Ext} satisfies*

$$\left\langle \frac{\partial}{\partial\theta} U_{\text{Ext}}(\phi,\theta) \right\rangle = 0. \tag{3.7}$$

Proof. We first prove that

$$\left\langle \frac{\partial}{\partial\theta} U(\phi,\theta) \right\rangle = 0. \tag{3.8}$$

This is not surprising. Physically, this quantity is the (negative) total torque on the system about the y-axis. Since the system is in equilibrium, the total torque should be zero. Mathematically, we have

$$\begin{aligned} \left\langle \frac{\partial}{\partial\theta} U(\phi,\theta) \right\rangle &= \frac{1}{Z}\int_0^{\pi}\int_0^{2\pi} \frac{\partial}{\partial\theta} U(\phi,\theta)\exp\left[-U(\phi,\theta)\right] d\theta \sin\phi\, d\phi \\ &\quad - \frac{1}{Z}\int_0^{\pi}\int_0^{2\pi} \frac{\partial}{\partial\theta}\exp\left[-U(\phi,\theta)\right] d\theta \sin\phi\, d\phi = 0. \end{aligned} \tag{3.9}$$

The second part is to show that

$$\left\langle \frac{\partial}{\partial\theta} U_1(\phi,\theta) + \frac{\partial}{\partial\theta} U_2(\phi,\theta) \right\rangle = 0. \tag{3.10}$$

Again, this is not surprising at all. Physically, this quantity is the torque on the system from the mutual interaction. If there is a torque on polymer rod A from other polymer rods, then by Newton's third law, rod A exerts a torque of the opposite sign on other rods. Thus, the sum of the torques on all rods from the mutual interaction is zero. Mathematically, we prove it as follows. We notice that

$$\frac{\partial m_1}{\partial\theta} = m_3, \qquad \frac{\partial m_2}{\partial\theta} = 0, \qquad \frac{\partial m_3}{\partial\theta} = -m_1. \tag{3.11}$$

Using this fact, we immediately obtain

$$\begin{aligned} \left\langle \frac{\partial}{\partial\theta} U_1(\phi,\theta) \right\rangle &= \frac{1}{Z}\int_0^{\pi}\int_0^{2\pi} \frac{\partial}{\partial\theta} U_1(\phi,\theta)\exp\left[-U(\phi,\theta)\right] d\theta \sin\phi\, d\phi \\ &= \frac{1}{Z}\int_S -\alpha\left(q_1 m_3 - q_3 m_1\right)\exp\left[-U(\mathbf{m})\right] d\mathbf{m} \\ &= -\alpha\left\langle q_1 m_3 - q_3 m_1 \right\rangle = -\alpha\left(q_1 q_3 - q_3 q_1\right) = 0. \end{aligned} \tag{3.12}$$

Here we have adopted the more concise and more convenient way of writing the spherical integrals in terms of $\mathbf{m}$ instead of (ϕ,θ). For the second part of the internal potential, we have

$$\frac{-1}{b}\left\langle\frac{\partial}{\partial\theta}U_2(\phi,\theta)\right\rangle = \frac{1}{bZ}\int_S b\langle\mathbf{mm}\rangle : \frac{\partial}{\partial\theta}(\mathbf{mm})\exp\left[-U(\mathbf{m})\right]d\mathbf{m} = \langle\mathbf{mm}\rangle : \left\langle\frac{\partial}{\partial\theta}(\mathbf{mm})\right\rangle$$
$$= \left\langle\begin{matrix} m_1^2 & m_1m_2 & m_1m_3 \\ m_1m_2 & m_2^2 & m_2m_3 \\ m_1m_3 & m_2m_3 & m_3^2 \end{matrix}\right\rangle : \left\langle\begin{matrix} 2m_1m_3 & m_2m_3 & m_3^2-m_1^2 \\ m_2m_3 & 0 & -m_1m_2 \\ m_3^2-m_1^2 & -m_1m_2 & -2m_1m_3 \end{matrix}\right\rangle = 0. \tag{3.13}$$

Combining (3.8) and (3.10) leads immediately to (3.7). □

Theorem 3.2. *For an equilibrium probability density, the z-axis is an eigenvector of the second moment tensor $\langle\mathbf{mm}\rangle$.*

Proof. Differentiating the external potential, we have

$$\frac{\partial}{\partial\theta}U_{\text{ext}}(\phi,\theta) = \frac{\partial}{\partial\theta}\left(\frac{\alpha_0}{2}m_3^2\right) = \alpha_0 m_1 m_3. \tag{3.14}$$

Applying Theorem 3.1, we obtain

$$\langle m_1m_3\rangle = 0. \tag{3.15}$$

Since the x-axis and the y-axis are selected arbitrarily, exchanging the roles of the x-axis and the y-axis yields immediately

$$\langle m_2m_3\rangle = 0. \tag{3.16}$$

Thus, the second moment tensor $\langle\mathbf{mm}\rangle$ has the form

$$\langle\mathbf{mm}\rangle = \begin{pmatrix} \langle m_1^2\rangle & \langle m_1m_2\rangle & 0 \\ \langle m_1m_2\rangle & \langle m_2^2\rangle & 0 \\ 0 & 0 & \langle m_3^2\rangle \end{pmatrix}. \tag{3.17}$$

It is obvious that the z-axis is an eigenvector of the the second moment tensor. □

To facilitate the analysis, let us select the x-axis and the y-axis properly in the xy-subspace to diagonalize the matrix $\langle\mathbf{mm}\rangle$. If $\langle\mathbf{m}\rangle$ is not parallel to the z-axis, then we can select the positive directions of x-axis and y-axis such that $q_1 > 0$ and $q_2 \geq 0$. In the new Cartesian coordinate system, we have

$$U(\mathbf{m}) = -\alpha(q_1m_1+q_2m_2+q_3m_3) - b(s_1m_1^2+s_2m_2^2+s_3m_3^2) - \frac{\alpha_0}{2}m_3^2, \tag{3.18}$$

where

$$s_j = \langle m_j^2\rangle. \tag{3.19}$$

Our next move is to prove two theorems which establish the relationship between the first moment vector and the second moment tensor.

THEOREM 3.3. *If an equilibrium solution satisfies $q_1 > 0$, then $q_3 = 0$.*

Proof. We prove the theorem by contradiction. Suppose there is a solution satisfying $q_1 > 0$ and $q_3 \neq 0$. We are going to show that $q_1 > 0$ and $q_3 \neq 0$ lead to $(1/q_3)\langle m_1 m_3\rangle > 0$, which contradicts with the result of Theorem 3.2:

$$\begin{aligned}\frac{1}{q_3}\langle m_1 m_3\rangle &= \frac{1}{q_3 Z}\int_S m_1 m_3 \exp[-U(m_1,m_2,m_3)]d\mathbf{m}\\ &= \frac{1}{q_3 Z}\int_{m_1>0} m_1 m_3\{\exp[-U(m_1,m_2,m_3)]-\exp[-U(-m_1,m_2,m_3)]\}d\mathbf{m}.\end{aligned} \tag{3.20}$$

In the above, the second factor of the integrand is

$$\begin{aligned}g_1(m_1,m_2,m_3) &\equiv \exp[-U(m_1,m_2,m_3)]-\exp[-U(-m_1,m_2,m_3)]\\ &= 2\sinh(\alpha q_1 m_1)\exp(\alpha q_3 m_3)\exp[\alpha q_2 m_2 - U_{\text{ext}}(\mathbf{m}) - U_2(\mathbf{m})].\end{aligned} \tag{3.21}$$

Notice that both $U_{\text{ext}}(\mathbf{m})$ and $U_2(\mathbf{m})$ are even functions of m_1, m_2, and m_3. Substituting into (3.20), we get

$$\begin{aligned}\frac{1}{q_3}\langle m_1 m_3\rangle &= \frac{1}{q_3 Z}\int_{m1>0} m_1 m_3 g(m_1,m_2,m_3)d\mathbf{m}\\ &= \frac{1}{q_3 Z}\int_{m1>0,\,m_3>0} m_1 m_3\{g(m_1,m_2,m_3)-g(-m_1,m_2,m_3)\}d\mathbf{m}\\ &= \frac{4}{Z}\int_{m1>0,\,m_3>0} m_1 m_3 \sinh(\alpha q_1 m_1)\frac{\sinh(\alpha q_3 m_3)}{q_3}\\ &\qquad \times \exp[\alpha q_2 m_2 - U_{\text{ext}}(\mathbf{m}) - U_2(\mathbf{m})]d\mathbf{m}.\end{aligned} \tag{3.22}$$

Since the integrand satisfies

$$\begin{gathered} m_1 m_3 \sinh(\alpha q_1 m_1)\frac{\sinh(\alpha q_3 m_3)}{q_3}\exp[\alpha q_2 m_2 - U_{\text{ext}}(\mathbf{m}) - U_2(\mathbf{m})] > 0\\ \text{for } m_1 > 0,\ m_3 > 0,\ q_1 > 0,\ q_3 \neq 0\end{gathered} \tag{3.23}$$

we have $(1/q_3)\langle m_1 m_3\rangle > 0$, which contradicts with the result of Theorem 3.2. □

Remark 3.4. Theorem 3.3 tells us that if the first moment vector $\langle \mathbf{m}\rangle$ is not parallel to the z-axis, then it must be perpendicular to the z-axis.

THEOREM 3.5. *If an equilibrium solution satisfies $q_1 > 0$, then $q_2 = 0$.*

Proof. The proof of this theorem is very similar to the proof of Theorem 3.3. In the proof of Theorem 3.3, we did not use that the z-axis is the direction of $\mathbf{E}$. We only used the fact that both $U_{\text{ext}}(\mathbf{m})$ and $U_2(\mathbf{m})$ are even functions of m_1, m_2, and m_3. So by exchanging the roles of m_2 and m_3, we can extend the proof of Theorem 3.3 to Theorem 3.5. □

Remark 3.6. Theorem 3.5 indicates that if the first moment $\langle \mathbf{m} \rangle$ is not parallel to the z-axis, then it must be parallel to either the x-axis or y-axis.

Combining the results of Theorems 3.3 and 3.5, we conclude that the first moment $\langle \mathbf{m} \rangle$ of the equilibrium solution of (2.2) must be aligned with one of the major axes of the second moment tensor $\langle \mathbf{mm} \rangle$.

4. Free energy and stability

In this section we consider the stability of an equilibrium solution. The study of stability allows us to narrow down the possible solutions that can be observed experimentally. To investigate the stability of an equilibrium solution, it is useful to exploit the free energy of the system.

To do so, consider an arbitrary probability density $\rho(\mathbf{m})$, which is not necessarily an equilibrium probability density. The free energy of the probability density $\rho(\mathbf{m})$ can be written as [8]

$$\begin{aligned} G[\rho] &= \int_S \rho(\mathbf{m}) \ln \rho(\mathbf{m}) d\mathbf{m} - \frac{\alpha}{2} \iint_S \mathbf{m}' \cdot \mathbf{m} \rho(\mathbf{m}') \rho(\mathbf{m}) d\mathbf{m}' d\mathbf{m} \\ &\quad - \frac{b}{2} \iint_S \mathbf{m}'\mathbf{m}' : \mathbf{mm} \rho(\mathbf{m}') \rho(\mathbf{m}) d\mathbf{m}' d\mathbf{m} - \frac{\alpha_0}{2} \iint_S (\mathbf{E} \cdot \mathbf{m})^2 \rho(\mathbf{m}) d\mathbf{m}' d\mathbf{m} \\ &= \int_S \rho(\mathbf{m}) \ln \rho(\mathbf{m}) d\mathbf{m} - \frac{\alpha}{2} \langle \mathbf{m} \rangle \cdot \langle \mathbf{m} \rangle - \frac{b}{2} \langle \mathbf{mm} \rangle : \langle \mathbf{mm} \rangle - \frac{\alpha_0}{2} \langle (\mathbf{E} \cdot \mathbf{m})^2 \rangle \\ &\equiv G_{\text{ent}}[\rho] + G_1[\rho] + G_2[\rho] + G_{\text{ext}}[\rho]. \end{aligned} \tag{4.1}$$

In the above expression $G_{\text{ent}}[\rho]$ corresponds to the entropic part of the free energy, $G_1[\rho]$ and $G_2[\rho]$ are free energy parts associated with the two mutual interactions, and $G_{\text{ext}}[\rho]$ is the free energy part associated with the external field. Here $\langle \cdot \rangle$ represents the mean taken with respect to the probability density $\rho(\mathbf{m})$. For the clarity of analysis below, we introduce two different notations for means taken with respect to two different probability densities.

(i) $\langle \cdot \rangle_{\text{eq}}$ denotes the mean taken with respect to the equilirium probability density, whereas $\langle \cdot \rangle$ represents the mean taken with respect to a general probability density (usually a perturbed probability density near the equilirium probability density).

Recall from Theorem 3.5 that for an equilibrium solution of the Smoluchowski equation (2.2), if the first moment $\langle \mathbf{m} \rangle$ is not parallel to the z-axis (which is the direction of the external elongational flow), then it must be parallel to either the x-axis or y-axis. We call these solutions with $\langle \mathbf{m} \rangle$ not parallel to the z-axis *nonparallel solutions*. Now we show that nonparallel equilibrium solutions are actually unstable. Therefore, we can exclude them from our study and focus on the parallel solutions.

THEOREM 4.1. *If an equilibrium solution satisfies $s_3 < s_1$ or $s_3 < s_2$, then it is unstable.*

Proof. We present the proof for the case of $s_3 < s_1$. The proof for the case of $s_3 < s_2$ is similar. □

In order to prove that an equilibrium solution with $s_3 < s_1$ is unstable, we show that the free energy $G[\rho]$ does not reach a local minimum at $\rho_{\text{eq}}(\mathbf{m})$. In particular, we construct a perturbed probability density $\tilde{\rho}(\mathbf{m})$ arbitrarily close to the equilibrium probability density $\rho_{\text{eq}}(\mathbf{m})$ such that

$$G[\tilde{\rho}(\mathbf{m})] < G[\rho_{\text{eq}}(\mathbf{m})]. \tag{4.2}$$

We rotate the equilibrium probability density $\rho_{\text{eq}}(\mathbf{m})$ about the y-axis by ε (a small angle) and use the result as the perturbed probability density $\tilde{\rho}(\mathbf{m})$. Mathematically this is equivalent to keeping the probability density fixed but rotating the external elongational flow field about the y-axis by $-\varepsilon$. After the rotation, the external elongational flow field is

$$\widetilde{\mathbf{E}} = (\sin\varepsilon, 0, \cos\varepsilon). \tag{4.3}$$

We have

$$\begin{aligned}
G_{\text{ent}}[\tilde{\rho}] &= G_{\text{ent}}[\rho_{\text{eq}}], \\
G_1[\tilde{\rho}] &= G_1[\rho_{\text{eq}}], \\
G_2[\tilde{\rho}] &= G_2[\rho_{\text{eq}}], \\
G_{\text{ext}}[\tilde{\rho}] &= -\frac{\alpha_0}{2}\langle(\mathbf{E}\cdot\mathbf{m})^2\rangle = -\frac{\alpha_0}{2}\langle(\widetilde{\mathbf{E}}\cdot\mathbf{m})^2\rangle_{\text{eq}} \\
&= -\frac{\alpha_0}{2}\langle(m_1\sin\varepsilon + m_3\cos\varepsilon)^2\rangle_{\text{eq}} \\
&= -\frac{\alpha_0}{2}\left(\langle m_3^2\rangle_{\text{eq}} + \varepsilon\langle m_1 m_3\rangle_{\text{eq}} + \varepsilon^2[\langle m_1^2\rangle_{\text{eq}} - \langle m_3^2\rangle_{\text{eq}}] + \cdots\right) \\
&= G_{\text{ext}}[\rho_{\text{eq}}] - \varepsilon^2\frac{\alpha_0}{2}(s_1 - s_3) + \cdots.
\end{aligned} \tag{4.4}$$

If $s_3 < s_1$, then for ε sufficiently small we have $G_{\text{ext}}[\tilde{\rho}] < G_{\text{ext}}[\rho_{\text{eq}}]$. It follows that $G[\tilde{\rho}] < G[\rho_{\text{eq}}]$ and the equilibrium solution ρ_{eq} is unstable.

Theorem 4.2. *If the first moment of an equilibrium solution satisfies $q_1 > 0$, then the dipole-dipole interaction strength α is related to the second moment through the inequality $\alpha s_1 > 1$.*

Proof. Suppose, on the contrary, that $\alpha s_1 \le 1$.

The parameter q_1 has a fixed value in the equilibrium solution $\rho_{\text{eq}}(\mathbf{m})$. Here we rename it v and treat it as an independent variable. We consider the probability density

$$\begin{aligned}
\rho(\mathbf{m}, v) &= \frac{1}{Z}\exp\left[\alpha(vm_1 + q_2 m_2 + q_3 m_3) + b(s_1 m_1^2 + s_2 m_2^2 + s_3 m_3^2) + \frac{\alpha_0}{2}m_3^2\right], \\
Z &= \int_S \exp\left[\alpha(vm_1 + q_2 m_2 + q_3 m_3) + b(s_1 m_1^2 + s_2 m_2^2 + s_3 m_3^2) + \frac{\alpha_0}{2}m_3^2\right]d\mathbf{m}.
\end{aligned} \tag{4.5}$$

For clarity, let $\langle\cdot\rangle_v$ denote the mean taken with respect to the probability density $\rho(\mathbf{m}, v)$. Note that $\rho(\mathbf{m}, v)|_{v=q_1} = \rho_{\text{eq}}(\mathbf{m})$. Consider the function

$$F(v) = v - \langle m_1\rangle_v, \tag{4.6}$$

which satisfies $F(0) = 0$ and $F(q_1) = 0$. We are going to show that $\alpha s_1 \leq 1$ implies that $F'(v) > 0$ for $v \in (0, q_1)$, which contradicts $F(0) = F(q_1) = 0$. Hence $\alpha s_1 > 1$. We do it in several steps.

Step 1. Differentiating with respect to v yields

$$\begin{aligned}\frac{\partial}{\partial v}\rho(\mathbf{m}, v) &= \alpha\left(m_1 - \langle m_1\rangle_v\right)\rho(\mathbf{m}, v),\\ \frac{d}{dv}\langle m_1\rangle_v &= \alpha\langle m_1\left(m_1 - \langle m_1\rangle_v\right)\rangle_v = \alpha\left(\langle m_1^2\rangle_v - \langle m_1\rangle_v^2\right),\\ F'(v) &= 1 - \alpha\langle m_1^2\rangle_v + \alpha\langle m_1\rangle_v^2.\end{aligned} \tag{4.7}$$

To show $F'(v) > 0$ for $v \in (0, q_1)$, we only need to show $\alpha\langle m_1^2\rangle_v < 1$ for $v \in (0, q_1)$. Consider function $g(v) = \langle m_1^2\rangle_v$. Because of the supposition $\alpha s_1 \leq 1$, we have $\alpha g(q_1) \leq 1$. Thus, to show $\alpha g(v) < 1$ for $v \in (0, q_1)$, we only need to show $g'(v) > 0$ for $v \in (0, q_1)$.

Step 2. $g(v)$ satisfies the property that $g'(v_0) = 0$ implies $g''(v_0) > 0$. To see this, differentiating $g(v)$ with respect to v yields

$$g'(v) = \langle m_1^2\left(m_1 - \langle m_1\rangle_v\right)\rangle_v. \tag{4.8}$$

Differentiating a second time respect to v, we get

$$\begin{aligned}g''(v) &= \langle m_1^2\left(m_1 - \langle m_1\rangle_v\right)^2\rangle_v - \langle m_1^2\rangle_v\langle m_1\left(m_1 - \langle m_1\rangle_v\right)\rangle_v\\ &= \langle m_1^4\rangle_v - 2\langle m_1^3\rangle_v\langle m_1\rangle_v + \langle m_1^2\rangle_v\langle m_1\rangle_v^2 - \langle m_1^2\rangle_v^2 + \langle m_1^2\rangle_v\langle m_1\rangle_v^2\\ &= \langle m_1^4\rangle_v - \langle m_1^2\rangle_v^2 - 2\langle m_1^2\left(m_1 - \langle m_1\rangle_v\right)\rangle_v\langle m_1\rangle_v\\ &= \operatorname{var}(m_1^2) - 2g'(v)\langle m_1\rangle_v.\end{aligned} \tag{4.9}$$

Therefore, if $g'(v_0) = 0$, then we have $g''(v_0) = \operatorname{var}(m_1^2) > 0$.

Step 3. When $v = 0$, the probability density $\rho(\mathbf{m}, 0)$ is symmetric with respect to m_1. So we have

$$g'(0) = \langle m_1^3\rangle\big|_{v=0} - \langle m_1^2\rangle\big|_{v=0}\langle m_1\rangle\big|_{v=0} = 0. \tag{4.10}$$

Using the result of Step 2, we conclude that $g'(v) > 0$ for $v \in (0, q_1)$. This leads immediately to $F'(v) > 0$ for $v \in (0, q_1)$, which contradicts $F(0) = F(q_1) = 0$. □

Theorem 4.3. *If an equilibrium solution satisfies $q_3 = 0$ and $\alpha s_3 > 1$, then it is unstable.*

Proof. This theorem does not specify any condition on q_1. Recall the method we used in selecting the axes: if the equilibrium solution is not parallel to $\mathbf{E}$, then we select the axes to make $q_1 > 0$ and $q_2 \geq 0$. Theorem 3.5 tells us that if $q_1 > 0$, then $q_2 = 0$ and $q_3 = 0$. So we always have $q_2 = 0$. □

We only need to show that the free energy $G[\rho]$ does not attain a local minimum at $\rho_{\text{eq}}(\mathbf{m})$. More precisely, we show that there exists a perturbed probability density $\tilde{\rho}(\mathbf{m})$

arbitrarily close to the equilibrium probability density $\rho_{eq}(\mathbf{m})$ such that

$$G[\tilde{\rho}(\mathbf{m})] < G[\rho_{eq}(\mathbf{m})]. \tag{4.11}$$

We consider

$$\tilde{\rho}(\mathbf{m}) = (1 + \varepsilon m_3)\rho_{eq}(\mathbf{m}). \tag{4.12}$$

Since $\langle m_3 \rangle_{eq} = q_3 = 0$, we have $\int_S \tilde{\rho}(\mathbf{m}) d\mathbf{m} = 1$, which means $\tilde{\rho}(\mathbf{m})$ is a probability density. We calculate the four parts of the free energy of the perturbed probability density $\tilde{\rho}(\mathbf{m})$.

Because $q_2 = 0$ and $q_3 = 0$, the equilibrium probability density $\rho_{eq}(\mathbf{m})$ is symmetric with respect to m_2 and m_3. Using the Taylor expansion

$$(a + \Delta x)\ln(a + \Delta x) = a\ln a + (\ln a + 1)\Delta x + \frac{1}{2a}(\Delta x)^2 + \cdots \tag{4.13}$$

we have

$$\begin{aligned} G_{\mathrm{ent}}[\tilde{\rho}] &= G_{\mathrm{ent}}[\rho_{eq}] + \varepsilon^2 \frac{1}{2} \langle m_3^2 \rangle_{eq} + \cdots, \\ G_1[\tilde{\rho}] &= G_1[\rho_{eq}] - \varepsilon^2 \frac{\alpha}{2} \langle m_3^2 \rangle_{eq}^2. \end{aligned} \tag{4.14}$$

The symmetry of $\rho_{eq}(\mathbf{m})$ gives us

$$\langle \mathbf{mm} m_3 \rangle_{eq} = \begin{pmatrix} 0 & 0 & \langle m_1 m_3^2 \rangle_{eq} \\ 0 & 0 & 0 \\ \langle m_1 m_3^2 \rangle_{eq} & 0 & 0 \end{pmatrix}. \tag{4.15}$$

Substituting into $G_2[\tilde{\rho}]$ yields

$$\begin{aligned} G_2[\tilde{\rho}] &= G_2[\rho_{eq}] - \varepsilon^2 b \langle m_1 m_3^2 \rangle_{eq}^2, \\ G_{\mathrm{ext}}[\tilde{\rho}] &= G_{\mathrm{ext}}[\rho_{eq}]. \end{aligned} \tag{4.16}$$

Combining (4.14) and (4.16), we obtain

$$\begin{aligned} G[\tilde{\rho}] - G[\rho_{eq}] &= \varepsilon^2 \frac{1}{2} \langle m_3^2 \rangle_{eq} - \varepsilon^2 \frac{\alpha}{2} \langle m_3^2 \rangle_{eq}^2 - \varepsilon^2 b \langle m_1 m_3^2 \rangle_{eq}^2 \\ &\leq -\varepsilon^2 \frac{1}{2} s_3 (\alpha s_3 - 1). \end{aligned} \tag{4.17}$$

If $\alpha s_3 > 1$, then for ε sufficiently small we have $G[\tilde{\rho}] < G[\rho_{eq}]$, which means the equilibrium solution ρ_{eq} is unstable.

Theorem 4.4. *If an equilibrium solution satisfies $q_1 > 0$, then it is unstable.*

Proof. Theorem 3.5 tells us that $q_1 > 0$ implies $q_2 = 0$ and $q_3 = 0$. We discuss two cases.

(i) Case 1: $s_3 < s_1$. Theorem 4.1 tells us that the equilibrium solution is unstable.

(ii) Case 2: $s_3 \geq s_1$. Using Theorem 4.2, we have $\alpha s_1 > 1$. It follows that $\alpha s_3 > 1$. Using Theorem 4.3, we conclude that the equilibrium solution is unstable.

Therefore, all nonparallel solutions are unstable. In this way, we have excluded all nonparallel solutions from our study. □

5. All stable equilibria are axisymmetric

In the previous section we have concluded that all nonparallel solutions are unstable. So from now on we only consider parallel solutions. It is well known that for pure (nondipolar) nematic rod ensembles where $\langle \mathbf{m} \rangle = 0$ [8, 11], the stable equilibria satisfy either $s_1 = s_2 = s_3$ (isotropic phase) or $s_3 > s_1 = s_2$ (prolate uniaxial phase) in the selected coordinate system. In [15] we showed that this is also the case for extended (dipolar) nematics. In this section we are going to show that in the presence of imposed uniaxial elongational flow the system still retains this axisymmetry.

Below for the case of extended nematic equilibria in which $\langle \mathbf{m} \rangle$ may be nonzero, we will show that if $\langle \mathbf{m} \rangle \neq 0$ (i.e., $q_1 = q_2 = 0$ and $q_3 > 0$ by the selection of the coordinate system and by the result of Theorem 3.1), then a stable equilibrium solution must be uniaxial. Furthermore, the axis of symmetry must be the major director (i.e., the eigenvector of the second moment corresponding to the largest eigenvalue). That is, $\langle m_1^2 \rangle = \langle m_2^2 \rangle < \langle m_3^2 \rangle$.

It should be point out that in [15] we proved five lemmas that paved the roads to reach axisymmetry. In order to prove axisymmetry for the extended nematics in the presence of an external elongational flow, we will take full advantage of the well-established results in [15].

First, we recall that in [15] there is no external flow field and the probability density is given by

$$\begin{aligned} \rho_1(\mathbf{m}) &= \frac{1}{Z} \exp\left[\alpha q_3 m_3 + b\left(s_1 m_1^2 + s_2 m_2^2 + s_3 m_3^2\right)\right], \\ Z &= \int_S \exp\left[\alpha q_3 m_3 + b\left(s_1 m_1^2 + s_2 m_2^2 + s_3 m_3^2\right)\right] d\mathbf{m}, \end{aligned} \tag{5.1}$$

where the components of the second moment and the first moment are

$$\langle m_i^2 \rangle = s_i, \tag{5.2}$$

$$\langle m_3 \rangle = q_3. \tag{5.3}$$

Under the constraints $s_1 < s_3$ and $s_2 < s_3$ we proved in [15] that there is no equilibrium solution such that $s_1 < s_2$. This holds for all $\alpha \geq 0$ and $b \geq 0$. Since the nonexistence of $s_1 < s_2$ (where $s_1 < s_3$ and $s_2 < s_3$) is true for all $\alpha \geq 0$ and $b \geq 0$, we introduce $\lambda = \alpha q_3$ as a parameter and treat α as unknown. Equation (5.3) becomes

$$\langle m_3 \rangle = \frac{\lambda}{\alpha}. \tag{5.4}$$

So (5.3) can be satisfied by selecting a suitable value of α. Therefore, (5.2) cannot be satisfied with $s_1 < s_2$ (where $s_1 < s_3$ and $s_2 < s_3$). Again, this is true for all λ and $b \geq 0$.

Let us introduce $r_j = bs_j$ as unknowns. Notice that the pdf in (5.1) does not depend on b any more once we know (r_1, r_2, r_3). Equation (5.2) yields

$$\langle m_1^2 \rangle = \frac{r_1}{b}, \qquad \langle m_2^2 \rangle = \frac{r_2}{b}, \qquad \langle m_3^2 \rangle = \frac{r_3}{b}. \tag{5.5}$$

It follows that (5.5) cannot be satisfied with $r_1 < r_2$ (where $r_1 < r_3$ and $r_2 < r_3$). This is true for all λ and $b \geq 0$. Further, we introduce

$$\eta_1 = r_1 - r_3 < 0, \qquad \eta_2 = r_2 - r_3 < 0. \tag{5.6}$$

Using the fact that $m_1^2 + m_2^2 + m_3^2 = 1$, the pdf in (5.1) can be rewritten as

$$\begin{aligned} \rho_1(\mathbf{m}) &= \frac{1}{Z} \exp\left(\lambda m_3 + \eta_1 m_1^2 + \eta_2 m_2^2\right), \\ Z &= \int_S \exp\left[\lambda m_3 + \eta_1 m_1^2 + \eta_2 m_2^2\right] d\mathbf{m}. \end{aligned} \tag{5.7}$$

Note that the parameter b does not appear in (5.7). Similarly, (5.5) turns into

$$\langle m_1^2 - m_3^2 \rangle = \frac{\eta_1}{b}, \qquad \langle m_2^2 - m_3^2 \rangle = \frac{\eta_2}{b}. \tag{5.8}$$

One concludes that (5.8) cannot be satisfied with $\eta_1 < \eta_2 < 0$. This conclusion holds for all λ and $b \geq 0$. Next, we prove two theorems for the extended nematics without external fields.

Theorem 5.1. *In the region $\eta_1 < \eta_2 < 0$, it is true for all λ that*

$$\eta_2 \langle m_1^2 - m_3^2 \rangle - \eta_1 \langle m_2^2 - m_3^2 \rangle \neq 0. \tag{5.9}$$

Proof. Suppose, on the contrary, that $\eta_2 \langle m_1^2 - m_3^2 \rangle - \eta_1 \langle m_2^2 - m_3^2 \rangle = 0$. Then we get

$$\frac{\langle m_1^2 - m_3^2 \rangle}{\eta_1} = \frac{\langle m_2^2 - m_3^2 \rangle}{\eta_2}. \tag{5.10}$$

Select the value in (5.10) as $1/b$ and we obtain (5.8), which violates the fact that (5.8) cannot be satisfied with $\eta_1 < \eta_2 < 0$.

Note that b does not appear in Theorem 5.1. □

Theorem 5.2. *In the region $\eta_1 < \eta_2 < 0$, it is true for all λ that*

$$\eta_2 \langle m_1^2 - m_3^2 \rangle - \eta_1 \langle m_2^2 - m_3^2 \rangle < 0. \tag{5.11}$$

Proof. Let us denote $\eta_2 \langle m_1^2 - m_3^2 \rangle - \eta_1 \langle m_2^2 - m_3^2 \rangle$ by $H(\eta_1, \eta_2)$. It is easy to verify that $H(\eta_1, \eta_2)$ is a continuous function of (η_1, η_2). From Theorem 5.1, $H(\eta_1, \eta_2)$ is nonzero in the region $\eta_1 < \eta_2 < 0$ and thus it does not change its sign in this region.

Consider the case where $\eta_1 < \eta_2$ and both η_1 and η_2 approach $-\infty$. Then $\langle m_3^2\rangle \to 1$, $\langle m_1^2\rangle \to 0$, and $\langle m_2^2\rangle \to 0$. Therefore,

$$H(\eta_1,\eta_2) = (\eta_1-\eta_2)\langle m_3^2\rangle + \eta_2\langle m_1^2\rangle - \eta_1\langle m_2^2\rangle < 0 \tag{5.12}$$

as $\eta_1 \to -\infty$ and $\eta_2 \to -\infty$ $(\eta_1 < \eta_2)$. This completes the proof of Theorem 5.2. □

Now we use the above results to consider the case of $\alpha_0 > 0$, which corresponds to the coupling of an imposed uniaxial elongational flow. In this case, the pdf is given by

$$\begin{aligned} \rho_2(\mathbf{m}) &= \frac{1}{Z}\exp\left[\alpha q_3 m_3 + b(s_1m_1^2+s_2m_2^2+s_3m_3^2) + \frac{\alpha_0}{2}m_3^2\right], \\ Z &= \int_S \exp\left[\alpha q_3 m_3 + b(s_1m_1^2+s_2m_2^2+s_3m_3^2) + \frac{\alpha_0}{2}m_3^2\right]d\mathbf{m}. \end{aligned} \tag{5.13}$$

As before, we introduce

$$\lambda = \alpha q_3, \qquad r_j = bs_j, \qquad \eta_1 = r_1 - r_3 - \frac{\alpha_0}{2}, \qquad \eta_2 = r_2 - r_3 - \frac{\alpha_0}{2}. \tag{5.14}$$

Then the pdf (5.13) becomes

$$\begin{aligned} \rho_2(\mathbf{m}) &= \frac{1}{Z}\exp\left[\lambda m_3 + \eta_1 m_1^2 + \eta_2 m_2^2\right], \\ Z &= \int_S \exp\left[\lambda m_3 + \eta_1 m_1^2 + \eta_2 m_2^2\right]d\mathbf{m}. \end{aligned} \tag{5.15}$$

Note that this pdf is exactly the same as the previous case (5.7) with η_1, η_2 defined slightly differently. Consequently, (5.8) is also modified slightly,

$$\langle m_1^2 - m_3^2\rangle = \frac{\eta_1 + \alpha_0/2}{b}, \qquad \langle m_2^2 - m_3^2\rangle = \frac{\eta_2 + \alpha_0/2}{b}. \tag{5.16}$$

Our next theorem generalizes earlier conclusions to include the coupling of external elongational flow.

Theorem 5.3. *In the region $\eta_1 < \eta_2 < 0$, it is true for all λ and $b \geq 0$ that (5.16) cannot be satisfied.*

Proof. We use proof by contradiction. Suppose (5.16) can be satisfied with $\eta_1 < \eta_2 < 0$ for some value of b. Then

$$\left(\eta_2 + \frac{\alpha_0}{2}\right)\langle m_1^2 - m_3^2\rangle - \left(\eta_1 + \frac{\alpha_0}{2}\right)\langle m_2^2 - m_3^2\rangle = 0, \tag{5.17}$$

or

$$\eta_2\langle m_1^2 - m_3^2\rangle - \eta_1\langle m_2^2 - m_3^2\rangle + \frac{\alpha_0}{2}\langle m_1^2 - m_2^2\rangle = 0. \tag{5.18}$$

From Theorem 5.2, $\eta_2\langle m_1^2 - m_3^2\rangle - \eta_1\langle m_2^2 - m_3^2\rangle < 0$. Therefore, $\langle m_1^2 - m_2^2\rangle > 0$.

Next, we show $\langle m_1^2 - m_2^2 \rangle < 0$ which contradicts the above result. To do so, we rewrite the pdf (5.15) as

$$\rho_2(\mathbf{m}) = \frac{1}{Z} \exp\left[\lambda m_3 + \frac{\eta_1 + \eta_2}{2}(m_1^2 + m_2^2) + \frac{\eta_1 - \eta_2}{2}(m_1^2 - m_2^2)\right], \tag{5.19}$$

and then

$$\langle m_1^2 - m_2^2 \rangle = \frac{1}{Z} \int_S \exp\left[\lambda m_3 + \frac{\eta_1 + \eta_2}{2}(m_1^2 + m_2^2)\right](m_1^2 - m_2^2) \exp\left[\frac{\eta_1 - \eta_2}{2}(m_1^2 - m_2^2)\right] d\mathbf{m}. \tag{5.20}$$

Switching the role of integration variables m_1 and m_2 in the integral on the right-hand side only (note that m_1, m_2 on the left-hand side are random variables and have different meanings) yields

$$\langle m_1^2 - m_2^2 \rangle = \frac{1}{Z} \int_S \exp\left[\lambda m_3 + \frac{\eta_1 + \eta_2}{2}(m_2^2 + m_1^2)\right](m_2^2 - m_1^2) \exp\left[\frac{\eta_1 - \eta_2}{2}(m_2^2 - m_1^2)\right] d\mathbf{m}. \tag{5.21}$$

Adding (5.20) and (5.21) and averaging, we obtain

$$\begin{aligned} &\langle m_1^2 - m_2^2 \rangle \\ &\quad = \frac{1}{Z} \int_S \exp\left[\lambda m_3 + \frac{\eta_1 + \eta_2}{2}(m_2^2 + m_1^2)\right](m_1^2 - m_2^2) \sinh\left[\frac{\eta_1 - \eta_2}{2}(m_1^2 - m_2^2)\right] d\mathbf{m} < 0, \end{aligned} \tag{5.22}$$

which contradicts earlier result. □

6. Conclusions

The stable equilibrium solutions of rigid, dipolar rod ensembles (extended nematics) under imposed elongational field are shown to be axisymmetric. Moreover, the distinguished axis of symmetry of stable anisotropic equilibria coincides with the first moment of the probability density function (pdf), the major director of the second moment of the pdf (eigenvector associated with the largest eigenvalue), and the imposed elongational flow field. This finding of axisymmetry provides reduction in the degree of freedom in the representation of the pdf solution and thereby significantly simplifies any process of obtaining physically observable equilibria.

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