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Macroscopic behaviour of a biaxial mixture of uniaxial nematics versus hydrodynamics of a biaxial nematic liquid crystal

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Abstract. — We discuss to what extent it is possible to distinguish macroscopically a truly biaxial nematic liquid crystal from a biaxial mixture of uniaxial aggregates in lyotropic systems. We propose for some cases simple experiments to resolve this question. The behaviour in an external magnetic field and the flow alignment in a shear flow experiment is discussed. We find that for small shear the flow alignment angle varies linearly with the shear rate $S$ whereas for truly biaxial nematics it is independent of $S$. In a static external magnetic field the orthorhombic symmetry can be lost above a first critical field and uniaxiality can be reached above a second critical field. The complete set of hydrodynamic equations for a biaxial mixture of uniaxial aggregates is presented in an appendix.

1. Introduction.

After the discovery of biaxial nematic liquid crystals [1] in 1980 in a lyotropic three component system, most experimental [2-4] and theoretical studies [5-8] concentrated on the macroscopic properties close to the uniaxial to biaxial phase transition. More recently, also the macroscopic and microscopic properties of the biaxial phase itself have been investigated [3, 9] and the question has been raised [9] whether the biaxial phase is actually made of biaxial aggregates or whether it is a mixture of dislike and rodlike micelles whose principal axes do not coincide so that this mixture also shows biaxial symmetry. The purpose of the present note is to consider the macroscopic and hydrodynamic properties of a biaxial mixture of uniaxial aggregates and to propose explicitly simple experiments which would allow for a distinction between the two cases.

The simplest case are of course phenomena which do not require spatial variations of the properties of the system. Those come in two groups, purely static phenomena like e.g. external magnetic or electric fields and stationary experiments characteristic of nematic liquid crystals like flow alignment, a phenomenon well established for uniaxial nematics [10].

A biaxial mixture of uniaxial aggregates can be characterized by two unit vectors $n$ and $m$, i.e. we associate with each species a different director field. In contrast to biaxial nematics, in particular orthorhombic nematics [11], the angle between the two...
directors is not fixed but can be changed by external forces like magnetic fields, shear, etc. Since both, the directions of \( \mathbf{n} \) and \( \mathbf{m} \) can be varied independently one has 4 independent macroscopic quantities \( \delta \mathbf{n} \) and \( \delta \mathbf{m} \) satisfying the constraints

\[
\delta \mathbf{n} \cdot \delta \mathbf{n} = 1, \quad \delta \mathbf{m} \cdot \delta \mathbf{m} = 1 \quad (1)
\]

or

\[
\delta \mathbf{n} \cdot \delta \mathbf{m} = 0 \quad \text{and} \quad \delta \mathbf{m} \cdot \delta \mathbf{n} = 0. \quad (2)
\]

In contrast to biaxial nematics the constraint \( \delta \mathbf{n} \cdot \delta \mathbf{m} = 0 \) [12] has been removed [13].

2. External magnetic field.

First we will discuss the behaviour of a biaxial mixture of aggregates in an external static magnetic field. It goes without saying that all the results can be transferred to the case of an external electric field, \( E \) (in this case one definitely needs a nonaqueous system, however).

For the free energy (spatially homogeneous) we have

\[
F = F_0(\rho, T, c, H^2) - \frac{1}{2} \chi_a(\mathbf{n} \cdot \mathbf{H})^2 - \frac{1}{2} \chi_m(\mathbf{m} \cdot \mathbf{H})^2
+ \frac{1}{2} M (\mathbf{n} \cdot \mathbf{m})^2 + \chi_a(\mathbf{n} \cdot \mathbf{H})(\mathbf{m} \cdot \mathbf{H})(\mathbf{n} \cdot \mathbf{m})
+ \frac{1}{2} \chi_m(\mathbf{n} \cdot \mathbf{m})^2 H^2. \quad (3)
\]

In truly biaxial nematics only the contributions proportional \( \chi_a \) and \( \chi_m \) exist [12]; the term \( \sim M \) is connected with the fact that the angle between \( \mathbf{n} \) and \( \mathbf{m} \) can be varied so as to minimize external constraints (steric, geometric in nature, boundary conditions, external fields, etc.). \( M \) is positive to guarantee thermostatic stability; the formal limit \( M \rightarrow \infty \) leads to the truly biaxial nematic case. \( \chi_a \) is essentially a field dependent correction to \( M \) which will not be considered explicitly in the following. \( \chi_a \) is a novel cross-coupling. Since it complicates considerably the subsequent formulae we will set it to zero first and discuss its influence towards the end of this section.

In the presence of an external field, there are five possible equilibrium states

I) \( \mathbf{H} \perp \mathbf{n} \perp \mathbf{m} \) \hspace{1cm} \text{with} \hspace{1cm} F = F_0.

II) \( \mathbf{H} \parallel \mathbf{n} \perp \mathbf{m} \) \hspace{1cm} \text{with} \hspace{1cm} F = F_0 - \frac{1}{2} \chi_m H^2.

III) \( \mathbf{H} \parallel \mathbf{m} \perp \mathbf{n} \) \hspace{1cm} \text{with} \hspace{1cm} F = F_0 - \frac{1}{2} \chi_a H^2.

IV) \( \mathbf{n}, \mathbf{H}, \mathbf{m} \) coplanar with finite angles \( \delta \) and \( \Theta \) between them (cf. Fig. 1) with

\[
F = F_0 - \frac{1}{2} H^2(\chi_a \cos^2 \delta + \chi_m \cos^2 \Theta) + \frac{1}{2} M \cos^2 (\delta + \Theta) \quad (4)
\]

where the angles are given by

\[
\cos^2 \Theta = (4 A)^{-1} ((A + 1)^2 - B^2) \quad (5)
\]

\[
\cos^2 \delta = (4 B)^{-1} ((B + 1)^2 - A^2) \quad (5)
\]

\[
\cos^2 (\delta + \Theta) = (4 A B)^{-1} (1 - (A - B)^2) \quad (5)
\]

\[
A = (\chi_a H^2)^{-1} M; \quad B = (\chi_m H^2)^{-1} M.
\]

V) \( \mathbf{n} \not\parallel \mathbf{m} \not\parallel \mathbf{H} \) with

\[
F = F_0 - \frac{1}{2} (\chi_a + \chi_m) H^2 + \frac{1}{2} M. \quad (5)
\]

In a truly biaxial nematic only the case I)-III) can occur. Their relative stability, depending on the signs of \( \chi_a, \chi_m \) and \( \chi_a - \chi_m \) is discussed in reference [12], figure 1.

The same stability diagram also applies for the biaxial mixture of uniaxial nematics if at least one of the \( \chi_a, \chi_m \) is negative. If both are positive, qualitatively new behaviour can occur. For low field strength, state II) is stable like in truly biaxial nematics (assuming for definiteness — but without loss of generality — \( \chi_a > \chi_m \)). However, above a critical field \( H_{c_1} \), state IV) occurs, where \( \mathbf{n}, \mathbf{m} \) and \( \mathbf{H} \) are coplanar but each two of those quantities enclose an angle which is different from 0° or 90° (Fig. 1). The angles \( \Theta \) and \( \delta \) (Eq. (5)) are field dependent. Rising the field strength, \( \Theta \) decreases monotonically, while \( \delta \) is first increasing and then, above a field \( H_{\text{max}} \), decreasing (cf. Fig. 2). At a finite upper critical field \( H_{c_2} \), both, \( \Theta \) and \( \delta \), are going to zero, thus reaching state V) which is stable for all \( H > H_{c_2} \) (Fig. 3). The critical field strengths are

\[
H_{c_1} = M \left( -\frac{1}{\chi_a} + \frac{1}{\chi_m} \right) \quad (6)
\]

\[
H_{c_2} = M \left( \frac{1}{\chi_a} + \frac{1}{\chi_m} \right). \quad (6)
\]

Since state V) is uniaxial, the field \( H_{c_1} \) is the threshold for a field induced biaxial to uniaxial transition. In case \( \chi_a \) and \( \chi_m \) are known from independent measurements, the determination of \( H_{c_1} \) or \( H_{c_2} \) would yield a

Fig. 1. — Geometry of state IV); \( \mathbf{n}, \mathbf{m} \) and \( \mathbf{H} \) are coplanar and the angles \( \delta, \Theta \) and \( \delta + \Theta \) are different from zero or \( \pi/2 \) (Eq. (5)).
value for the «stiffness» constant $\mathcal{M}$. If the $\chi'$ and $\chi''$ terms of equation (3) are incorporated we obtain instead of equations (5) and (6) more complicated expressions. In (5) one has to replace $\mathcal{M} \rightarrow \mathcal{M} + (\chi' + \chi'')/H^2$ and $\chi'^m \rightarrow \chi'^m - \chi$ while (6) reads now

$$
H_{c_1}^2 = \mathcal{M}(\chi' + \chi'') \left[ \chi'' + \chi'' \chi' + \chi'' (\chi' - \chi'' - 2 \chi' \chi'') \right]^{-1} \\
H_{c_2}^2 = \mathcal{M}(\chi' + \chi'') \left[ \chi'' \chi' - 2 \chi' \chi' + \chi''' \chi'' - \chi''' \chi' \chi'' + \chi''' \right]^{-1}.
$$

(7)

---

Fig. 2. — Angles $\Theta$ and $\delta$ as a function of the field strength in state IV).

Fig. 3. — Stability of states II), IV) and V) as a function of magnetic field strength (for $\chi'' > \chi' > 0$). State IV) is as plotted in figure 1; $H_{c_1}$ and $H_{c_2}$ are given in equation (6).

Therefore, if a lyotropic biaxial nematic belongs to the class $\chi' > 0, \chi'' > 0$ one can check by applying a magnetic field whether one is investigating a biaxial mixture of uniaxial aggregates or a truly biaxial nematic. In the other 3 possible cases ($\chi' < 0, \chi'' < 0$; $\chi' < 0, \chi'' > 0$; $\chi' > 0, \chi'' < 0$) only the states I-III can occur in both systems so that a different method is necessary for discrimination.

3. Flow alignment.

An obvious method which is very sensitive in probing nematic behaviour is flow alignment in a shear flow. We consider a planar geometry with the velocity field

$$
V = S\tau \hat{x}_x
$$

with the shear rate $S$ and $\hat{x}_x$ the unit vector in $x$ direction. Since the hydrodynamic equations for truly biaxial nematics [12, 15, 16] no longer apply we have to generalize them suitably by removing the constraint $\mathbf{n} \cdot \mathbf{m} = 0$. The full hydrodynamic equations for a biaxial mixture of uniaxial aggregates are presented in the appendix. Here we concentrate on the parts which are necessary to describe the experimentally easily accessible case of flow alignment. We have for the quasicurrents of $\mathbf{n}$ and $\mathbf{m}$

$$
X_i^\tau = (\alpha_i \delta_i^3 + \chi_i m_i m_i) n_i (V_i V_k + V_k V_i) + \frac{1}{2} n_i (V_i V_k - V_k V_i) + (\zeta_i \delta_i^3 + \zeta_i m_i m_k) H_k^m \\
X_i^m = (\alpha_i \delta_i^3 + \chi_i m_i m_i) m_i (V_i V_k + V_k V_i) + \frac{1}{2} m_i (V_i V_k - V_k V_i) + (\zeta_i \delta_i^3 + \zeta_i n_i n_k) H_k^n
$$

(9)

(10)

with

$$
H_k^m = 2 M n_k (\mathbf{n} \cdot \mathbf{m}) \\
H_k^n = 2 M n_k (\mathbf{n} \cdot \mathbf{m}) \delta_i^3 = \delta_i^3 - n_i n_i - m_i m_i
$$

(11)

in the spatially homogeneous limit (no distortion of the director fields $\mathbf{n}$ and $\mathbf{m}$). As can be read off immediately from equations (9), (10), there are four reversible flow parameters $\alpha_i$ and four direction friction coefficients $\zeta_i$. The terms with the «coefficients» $1/2$ describe the solid body rotation $\mathbf{n} \sim \omega \times \mathbf{n}$ and $\mathbf{m} \sim \omega \times \mathbf{m}$, respectively.

The equations

$$
\dot{n}_i + X_i^\tau = 0; \quad \dot{m}_i + X_i^m = 0
$$

(12)

are then investigated for planar stationary solutions. We assume $\mathbf{n}$ and $\mathbf{m}$ to lie in the shear plane (see Fig. 4)

$$
\dot{n} = l_x \cos \phi + l_z \sin \phi \\
\dot{m} = l_x \cos \psi - l_z \sin \psi
$$

(13)

which can probably be achieved with the help of external fields. The requirement of vanishing torques (or stationarity) leads for small shear rates to the result

$$
\phi - \psi = \frac{1}{4 \mathcal{M} \zeta_4 \alpha_3 - \zeta_4 \alpha_7} S
$$

(14)

where we have linearized in $\phi - \psi$, an approximation.
which is justified when the flow alignment angle is small, a condition which is usually satisfied. A non-linear treatment would change the basic equations because the shear changes the structure for the biaxial mixture of uniaxial aggregates.

This result (Eq. (14)) is in striking contrast to the case of truly biaxial nematics where the flow alignment angles $\phi$ and $\psi$ are constant, i.e. they do not depend on the shear rate at all [15]. Here we find that for a biaxial mixture of uniaxial nematics the difference in the alignment angles is proportional to the shear rate for small shear. This is a result which allows for a qualitative distinction between the two possibilities.

The basic reason for the dependence on $S$ might be traced back to the fact that even in the spatially homogeneous limit the viscous torque enters via the term $\sim M$ which is geometric and/or steric in origin and allows for a non-zero difference $\phi - \psi$, which means that $\hat{n}$ and $\hat{m}$ are no longer orthogonal. If the hydrodynamic parameters $\alpha_2$, $\alpha_7$, $\zeta_2$ and $\zeta_4$ are known, the shear flow experiment allows to measure $M$, the specific $\langle$ stiffness $\rangle$ constant of the biaxial mixture. In order to get a rough estimate of the values of the hydrodynamic parameters one can assume that the two constituents of the mixture still have their uniaxial character, i.e. the hydrodynamic parameters are those, each constituent would have in its (unmixed) uniaxial nematic phase. In this approximation

$$\begin{align*}
\alpha_2 &= \alpha_1 = -\frac{1}{2} \lambda^{(u)} \\
\alpha_5 &= \alpha_7 = -\frac{1}{2} \lambda^{(m)} \\
\zeta_1 &= \zeta_2 = \zeta^{(u)} \\
\zeta_3 &= \zeta_4 = \zeta^{(m)}
\end{align*}$$

(15)

where $\lambda^{(u,m)}$ are the usual reversible flow parameters $\dot{\lambda}$ ($\equiv -\gamma_2/\gamma_1$ in de Gennes's notation) and $\zeta^{(u,m)}$ the director friction coefficients $\zeta$ ($\equiv 1/\gamma_1$) of the two different uniaxial nematics, which form the mixture, with director $\hat{n}$ and $\hat{m}$, respectively.

Of course there is the possibility that in the mixture the two uniaxial constituents influence each other inducing a biaxiality resulting in $\alpha_2 \neq \alpha_1$ etc. This is quite different from the case of a truly biaxial nematic, where the biaxial objects induce the macroscopic biaxial ordering while in the biaxial mixture the macroscopic ordering probably induces a biaxiality in the originally uniaxial constituents. If this induced biaxiality is small, equation (15) is a good approximation.


In closing we want to mention another possibility to distinguish mixtures from a truly biaxial nematic. Mixtures of fluids and, of course, also of liquid crystals have static and viscous cross-couplings between temperature variations and changes in the concentration. If one applies e.g. a temperature gradient [17] to a binary mixture of miscible, isotropic fluids in the heat conduction state one obtains always simultaneously a concentration gradient which can be detected easily by a beam deflection technique via measurement of the change in index of refraction. A similar type of experiment sensitive to density changes could be feasible for the distinction of mixtures and truly biaxial nematics for which the cross-coupling between temperature and concentration becomes anisotropic.

In conclusion, we have proposed simple experiments to decide whether the biaxial systems in lyotropics are truly biaxial nematics or biaxial mixtures of uniaxial nematics via the application of an external magnetic field (for the anisotropies $\chi^{(u,m)}$ both positive) or the measurement of the flow alignment angle in shear flow where we predict that the angle $(\phi - \psi)$ is proportional to the shear rate for a mixture.

Very recent synchrotron experiments [18] in the system K-laurate, 1-decanol and D$_2$O done at LURE indicate that at least in this system one has a truly biaxial nematic. We infer this from the extension ($\sim 400$ Å) of the positional ordering in all nematic phases along the axis of the capillary, a range which is difficult to reconcile with the picture of a mixture of discs and rods in this system.

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Appendix.

HYDRODYNAMIC EQUATIONS FOR THE BIAXIAL MIXTURE OF UNIAXIAL AGGREGATES. — The purpose of this appendix is to summarize the basic equations for biaxial mixtures of uniaxial nematics and to contrast them with those for biaxial nematics as derived in references [12, 16] or, in the Leslie-Eriksen terminology, in reference [14]. As hydrodynamic variables we have the conserved quantities density $\rho$, entropy density $s$,
density of linear momentum $g$, and the concentration of one of the components $c$. As discussed in detail in the text, the variables associated with the broken symmetries about the two preferred directions $\hat{n}$ and $\hat{m}$ are $\hat{n}$ and $\hat{m}$, respectively with $\hat{n} \cdot \hat{m} = 0$ and $\hat{m} \cdot \hat{m} = 0$. Thus the dynamic equations are

$$
\begin{align*}
\dot{\rho} + V_i \dot{g}_i &= 0 \\
\dot{g}_i + V_j \sigma_{ij} &= 0 \\
\dot{\sigma} + V_i \sigma_i &= 0 \\
\dot{c} + V_i c_i &= 0 \\
\dot{\hat{n}}_i + X_i^\rho &= 0 \\
\dot{\hat{m}}_i + X_i^\rho &= 0.
\end{align*}
$$

(A.1)

In local thermodynamic equilibrium the hydrodynamic variables satisfy the Gibbs relation

$$
d\varepsilon = T \, d\sigma + \mu \, d\rho + Z \, dc + V \cdot dg + \phi_{ij} d(V m_i) + \phi_{ij} d(V m_j) + h_i^\rho d\hat{n}_i + h_i^\rho d\hat{m}_i. \tag{A.2}
$$

For the thermodynamic conjugate quantities defined via equation (A.2) we have

$$
\begin{align*}
\delta \mu &= \lambda \delta \rho + \gamma \delta \sigma + \chi \delta c \\
\delta T &= T \varepsilon T^{-1} \delta \sigma + \gamma \delta \rho + \chi \delta c \\
\delta Z &= \chi \delta \sigma + \gamma \delta \rho + \xi \delta c \\
\delta V &= \frac{\partial F^g}{\partial \rho}, \quad \delta \sigma_i = \frac{\partial F^g}{\partial m_i}, \\
\delta h_i^\rho &= \frac{\partial F^g}{\partial \hat{n}_i}, \quad \delta h_i^\rho = \frac{\partial F^g}{\partial \hat{m}_i}
\end{align*}
$$

(A.3)

where $F^g$ is the gradient free energy containing as a special case both uniaxial nematics and orthorhombic biaxial nematics (we assume invariance under $\hat{n} \rightarrow -\hat{n}$ and $\hat{m} \rightarrow -\hat{m}$ separately)

$$
F^g = K_{ijkl}(V m_k) (V m_l) + K_{ijkl}(V m_k) (V m_l) + K_{ijkl}(V m_k) (V m_l)
$$

(A.4)

with

$$
K_{ijkl} = \delta_{ij}^3 \{ K_1 \delta_{kl}^3 + K_2 m_k m_l + K_3 n_k n_l \} + m_k m_l \{ K_4 \delta_{kl}^3 + K_5 m_k m_l + K_6 n_k n_l \} + \frac{1}{4} K_7 \{ m_k m_l \delta_{kl}^3 + m_k m_l \delta_{kl}^3 + m_k m_l \delta_{kl}^3 + m_k m_l \delta_{kl}^3 \}
$$

(A.5)

and

$$
K_{ijkl} = \delta_{ij}^3 \{ K_8 \delta_{kl}^3 + K_9 m_k m_l + K_{10} n_k n_l \} + n_k n_l \{ K_{11} \delta_{kl}^3 + K_{12} m_k m_l + K_{13} n_k n_l \} + \frac{1}{4} K_{14} \{ n_k n_l \delta_{kl}^3 + n_k n_l \delta_{kl}^3 + n_k n_l \delta_{kl}^3 + n_k n_l \delta_{kl}^3 \}
$$

(A.6)

There are 20 bulk coefficients in the gradient free energy. The case of a truly biaxial nematic is enclosed in equations (A.5)-(A.7) and obtained if the additional constraint $\hat{n} \cdot \hat{m} = -\hat{m} \cdot \hat{n}$ is implemented, i.e. $F^g$ must not depend on $\hat{n} \cdot \hat{m} + \hat{m} \cdot \hat{n}$. In that case $K_4 = K_{11}, K_5 = K_{12}, K_6 = K_{13}, K_{18} = -K_{14}, K_7 = K_{15}, K_{16} = K_{17}, K_{20} = 0$. There are 12 independent coefficients.

For the flow alignment tensor we have — as already discussed in the text — 4 parameters (index $R$ reversible)

$$
X_{ij}^{\text{rk}} = x_1 (n_j \delta_{ik}^3 + n_k \delta_{ij}^3) + x_2 (m_j m_k n_i + m_j m_i n_k)
$$

(A.8)

and

$$
X_{ij}^{\text{rd}} = x_3 (n_j \delta_{ik}^3 + n_k \delta_{ij}^3) + x_2 (m_j m_k n_i + m_j m_i n_k)
$$

(A.9)

In truly biaxial nematics there are only three parameters since $x_2 = -x_3$; this leads immediately to $\varphi = \psi$ in equation (14). For the dissipative currents, we find no change with respect to $V_{ijkl}$ when compared with biaxial nematics. For the other currents we have (index $D$ dissipative)

$$
X_{ij}^{\text{rd}} = x_3 (n_j \delta_{ik}^3 + n_k \delta_{ij}^3) + x_2 (m_j m_k n_i + m_j m_i n_k)
$$

(A.9)
with $H^m_n \equiv h^m_n - \nabla \phi_{j}^{m}$ (where again the case of truly biaxial nematics is enclosed via $\zeta_2 = - \zeta_2$), and

\begin{align}
j_i^{\alpha \beta} &= (\kappa_1 \delta_i^{\alpha j} + \kappa_2 n_i n_j + \kappa_3 m_i m_j) \nabla_j T + \\
&+ (D_1 \delta_i^{\alpha j} + D_2 n_i n_j + D_3 m_i m_j) \nabla_j Z \quad (A.10)
\end{align}

\begin{align}
j_i^{\alpha \beta} &= (D_1 \delta_i^{\alpha j} + D_2 n_i n_j + D_3 m_i m_j) \nabla_j Z + \\
&+ (D_1 \delta_i^{\alpha j} + D_2 n_i n_j + D_3 m_i m_j) \nabla_j T \quad (A.11)
\end{align}

where the $D$'s and $D''$'s are absent in truly biaxial nematics. Equations (A.10) and (A.11) show clearly the anisotropic dissipative (viscous) cross-coupling between variations of the temperature and the chemical potential for species $c$. The static cross-coupling between entropy and concentration variations is still isotropic, however (cf. Eq. (A.3)).

Finally, we would like to stress that there are no anholonomity relations [12] for the variables $\delta n$ and $\delta m$ of a biaxial mixture, due to the fact that $\tilde{\mathbf{n}}$ and $\tilde{\mathbf{m}}$ can rotate independently to each other while in truly biaxial nematics rotations are not independent because $\tilde{\mathbf{n}}$, $\tilde{\mathbf{m}}$ and $\tilde{\mathbf{n}} \times \tilde{\mathbf{m}}$ form a rigid triad.

References

[13] Throughout the paper we will use the notation introduced in Ref. [12]. A description using the Leslie-Ericksen type approach for truly biaxial nematics has been given in Ref. [14].