

## Hydrodynamics of biaxial nematics with arbitrary nonsingular textures

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The hydrodynamics of biaxial nematics has been obtained in the presence of an external field  $\vec{H}$ . By working with the triad order parameter, we obtain results valid for arbitrary nonsingular textures. We also present the bending energy for biaxial nematics. Our derivation of the hydrodynamics explicitly includes angular momentum conservation. The resulting hydrodynamic equations clearly possess a rotating-equilibrium solution. In an Appendix we discuss extending the theory to include the internal angular momentum of the molecules which constitute the biaxial nematic.

## I. INTRODUCTION

We have derived the hydrodynamics of biaxial nematic liquid crystals in the presence of a magnetic field  $\vec{H}$ .<sup>1</sup> Particular attention has been paid to two points. First, by working with the triad order parameter, we obtain results valid for arbitrary nonsingular textures. Second, by explicitly considering angular momentum conservation, we unify the procedures of phenomenological hydrodynamics. It then becomes apparent that the angular momentum flux  $\mathcal{J}_i^j$  has associated with it the thermodynamic "force"  $\partial_j \omega_i$  (where  $\omega_i$  is the local rotation rate); this result is not restricted only to biaxial nematics, but appears quite generally for systems possessing internal angular momentum.

To provide a context for our derivation, we note that one can derive hydrodynamic theories in a number of different ways. It is arbitrary but (for our purposes) convenient to place such theories into three classes: first, purely phenomenological theories (to which the present theory belongs); second, theories which are a mixture of phenomenology and microscopics; and third, theories which seriously attempt to make the connection to microscopics. For example, in the case of the superfluid  $^4\text{He}$ , Landau's work<sup>2</sup> would fall in the first category, whereas that of Hohenberg and Martin would be in the third.<sup>3</sup> For nematic liquid crystals, I would place the work of Forster *et al.*<sup>4</sup> in the first category, and that of Forster<sup>5,6</sup> in the third. For  $^3\text{He-A}$ , the works of Graham,<sup>7</sup> Ho,<sup>8</sup> and Hu and Saslow<sup>9</sup> would fall in the first category, and the works of Brand, Dörfle and Graham<sup>10,11</sup> would fall in the third. For magnetic fluids (suspensions of  $10^{-6}$  cm ferromagnetic parti-

cles in ordinary fluids), only works in the first category have appeared (see Schliomis for a review<sup>12</sup>). Most hydrodynamic theories, in fact, are of the first type, since it is much simpler to develop such a theory.

In principle, to develop a theory of the third type, one can employ the general procedure of Forster.<sup>13</sup> This has been done by Zeyher<sup>14</sup> for superionic conductors, and it is the only case known to this author where a theory of the third type preceded any theory of the first type. However, this approach<sup>13</sup> involves a cumbersome and somewhat opaque formalism; it is probably for this reason that it has not found a wider applicability.

Works in the second class, which have only begun to appear recently, obtain the reactive response in a more microscopic fashion than do works of the first class. We may cite the following. Lebedev and Khalatnikov have employed a Lagrangian and a Hamiltonian formalism.<sup>15</sup> Dzyaloshinskii and Volovik have employed a Hamiltonian formalism,<sup>16</sup> with an emphasis upon what can be earned about Poisson brackets when symmetry groups can be exploited.<sup>17</sup> Also, Saslow, Gabay, and Hurdequint<sup>18</sup> have recently extended the Larmor precession theorem of Halperin and Saslow,<sup>19</sup> in the end obtaining a Hamiltonian formalism.<sup>20</sup>

In the present paper we make no use of an Hamiltonian formalism or of the more complete formalism of Ref. 13. Our only *a priori* knowledge of the reactive response is taken to be the well-known force and torque of an external magnetic field  $\vec{H}$  on a magnetized system.

In spirit, our derivation is close to that given by Martin, Parodi, and Pershan, for ordinary nematics and other systems.<sup>21</sup> However, we include angular momentum as an integral part of the derivation,

rather than eliminate it by employing a purely symmetric stress tensor<sup>4,21</sup> or treat it somewhat as an afterthought.<sup>8,9</sup> We find that it is necessary to carefully consider the equilibrium conditions which determine the “thermodynamic driving forces.” It is convenient to rephrase the translational and rotational equilibrium conditions into the mathematical statements that both  $A_{ij} = \frac{1}{2}(\partial_i v_j + \partial_j v_i)$  and  $\partial_j \omega_i$  be zero in equilibrium. (Here  $\vec{v}$  is the fluid velocity and  $\vec{\omega} \equiv \frac{1}{2} \vec{\nabla} \times \vec{v}$ .) This is equivalent to saying that there be no shear ( $A_{ij}=0$ ) and that the rotation rate be spatially uniform ( $\partial_j \omega_i=0$ ).

On eliminating the antisymmetric part of the stress tensor  $\sigma_{ij}$  in favor of the divergence of the angular momentum flux  $\mathcal{J}_i^j$ , we find (as stated above), that the rate of entropy production includes the product of  $\mathcal{J}_i^j$  and  $\partial_j \omega_i$ , in addition to the (normally found) product of  $\frac{1}{2}(\sigma_{ij} + \sigma_{ji})$  and  $A_{ij}$ . Following the usual procedures of phenomenological hydrodynamics, we determine the unknown fluxes. The resulting hydrodynamic equations clearly possess a rotating-equilibrium solution. We note that, in the case of <sup>3</sup>He-*A*, rotating equilibrium has been the subject of recent discussion,<sup>22,23</sup> and this discussion has influenced our work.

In Sec. II we describe the thermodynamics of a biaxial nematic liquid crystal in the presence of a magnetic field  $\vec{H}$ . In Sec. III we consider the rotating-equilibrium conditions and thereby obtain the thermodynamic driving forces. Section IV considers the general form of the hydrodynamic equations, the behavior of the system under Galilean boosts and under uniform rotations, and the implications of angular momentum conservation for the antisymmetric part of the stress tensor. In Sec. V, we obtain the unknown fluxes which appear in the hydrodynamic equations. Section VI provides a short conclusion.

In an Appendix we sketch how one may include the internal angular momentum of the liquid crystal molecules, in addition to their spin angular momentum. There we also provide references to recent work on simple fluids with internal angular momenta, including numerical computations of their transport coefficients.

## II. THERMODYNAMICS OF BIAxIAL NEMATICS

The order parameter is assumed to be given by the rotation matrix

$$R_{ij}(\vec{r}) = b_i^{(n)}(\vec{r}) e_j^{(n)}, \quad (2.1)$$

where  $\{\hat{e}^{(n)}\}$  is a fixed right-handed triad of unit vectors with  $\hat{e}^{(3)} = \hat{e}^{(1)} \times \hat{e}^{(2)}$  and  $\{\hat{b}^{(n)}(\vec{r})\}$  is a spatially varying right-handed triad of unit vectors with  $\hat{b}^{(3)} = \hat{b}^{(1)} \times \hat{b}^{(2)}$ . Summation on repeated indices is implied. The system is assumed to be unchanged under a local 180° rotation about any of the  $\hat{b}^{(n)}$ , so that the system is invariant under, e.g.,  $(\hat{b}^{(1)}, \hat{b}^{(2)}, \hat{b}^{(3)}) \rightarrow (-\hat{b}^{(1)}, -\hat{b}^{(2)}, \hat{b}^{(3)})$ .

The energy density  $\epsilon$  is assumed to be a function of the following thermodynamic variables: the entropy density  $S$ , the mass density  $\rho$ , the momentum density  $\vec{g}$ , the magnetization density  $\vec{m}$ , the rotation matrix  $R_{ij}$ , and the spatial gradients  $\partial_k R_{ij}$ . We will assume a static external field  $\vec{H}$ , and we include the  $-\vec{m} \cdot \vec{H}$  interaction in  $\epsilon$  (so  $\epsilon = \epsilon_{\text{int}} - \vec{m} \cdot \vec{H}$ , where  $\epsilon_{\text{int}}$  is the internal energy density). Then, at a given position  $\vec{r}$ , we take the thermodynamics to be given by

$$d\epsilon = TdS + \mu d\rho + \vec{v} \cdot d\vec{g} + (\vec{h} - \vec{H}) \cdot d\vec{m} + \phi_{ij} dR_{ij} + \phi_{ijk} d(\partial_k R_{ij}). \quad (2.2)$$

With (2.1), we have

$$\phi_{ij} dR_{ij} = \phi_{ij} e_j^{(n)} db_i^{(n)} \equiv \phi_i^{(n)} db_i^{(n)}, \quad (2.3)$$

$$\phi_{ijk} d(\partial_k R_{ij}) \equiv \phi_{ijk} e_j^{(n)} d(\partial_k R_{ij}) \equiv \phi_{ij}^{(n)} d(\partial_j b_i^{(n)}), \quad (2.4)$$

so that (2.2) can be written as

$$d\epsilon = TdS + \mu d\rho + \vec{v} \cdot d\vec{g} + (\vec{h} - \vec{H}) \cdot d\vec{m} + \phi_i^{(n)} db_i^{(n)} + \phi_{ij}^{(n)} d(\partial_j b_i^{(n)}), \quad (2.5)$$

Here  $T \equiv \partial\epsilon/\partial S$  is the temperature,  $\mu$  is the chemical potential,  $\vec{v} \equiv \partial\epsilon/\partial\vec{g}$  is the fluid velocity,  $\vec{h} \equiv \partial\epsilon_{\text{int}}/\partial\vec{m}$  is the “internal” magnetic field, and  $\phi_i^{(n)}$  and  $\phi_{ij}^{(n)}$  are the internal fields associated with  $b_i^{(n)}$  and  $\partial_j b_i^{(n)}$ . We define the pressure  $P$  via

$$\epsilon + P = TS + \mu\rho + \vec{v} \cdot \vec{g} + (\vec{h} - \vec{H}) \cdot \vec{m} + \phi_i^{(n)} b_i^{(n)} + \phi_{ij}^{(n)} \partial_j b_i^{(n)}, \quad (2.6)$$

so that (2.5) and (2.6) yield the Gibbs-Duhem relation:

$$dP = SdT + \rho d\mu + \vec{g} \cdot d\vec{v} + \vec{m} \cdot d(\vec{h} - \vec{H}) + b_i^{(n)} d\phi_i^{(n)} + \partial_j b_i^{(n)} d\phi_{ij}^{(n)}. \quad (2.7)$$

Because, under an infinitesimal “boost” by  $d\vec{v}$ ,

$$d\epsilon = \vec{g} \cdot d\vec{v}, \quad d\vec{g} = \rho d\vec{v}, \quad (2.8)$$

use of (2.5) gives

$$\vec{g} = \rho \vec{v}, \quad (2.9)$$

thus establishing that  $\vec{v}$  is indeed the fluid velocity.

Under a rotation of the system by  $d\vec{\Theta}$ , we have  $d\epsilon = d(\epsilon_{\text{int}} - \vec{H} \cdot \vec{m}) = -\vec{H} \cdot d\vec{m} = -\vec{m} \times \vec{H} \cdot d\vec{\Theta}$ , or  $\partial\epsilon/\partial\vec{\Theta} = -\vec{m} \times \vec{H}$ . With (2.5) this yields

$$0 = \vec{v} \times \vec{g} + \vec{h} \times \vec{m} + \vec{\phi}^{(n)} \times \vec{b}^{(n)} + \vec{\phi}_{i-}^{(n)} \times \vec{\nabla} b_i^{(n)} + \vec{\phi}_{-j}^{(n)} \times \partial_j \vec{b}^{(n)}. \quad (2.10)$$

Here  $(\phi_{i-}^{(n)})_j \equiv \phi_{ij}^{(n)}$ . Use of (2.9) and an integration by parts permits us to rewrite (2.10) as

$$0 = \vec{h} \times \vec{m} + \vec{\Phi}^{(n)} \times \vec{b}^{(n)} + \partial_j (\vec{\phi}_{-j}^{(n)} \times \vec{b}^{(n)}) + \vec{\phi}_{i-}^{(n)} \times \vec{\nabla} b_i^{(n)}, \quad (2.11)$$

where

$$\vec{\Phi}^{(n)} \equiv \vec{\phi}^{(n)} - \partial_j \vec{\phi}_{-j}^{(n)}. \quad (2.12)$$

We will find (2.11) helpful later when we employ it in conjunction with angular momentum conservation.

As another preliminary, we wish to rewrite  $\partial_j b_i^{(n)}$ , which appears in (2.5)–(2.7). First we note that there are nine gradients associated with the three gradients of the three  $\vec{b}^{(n)}$ . They may be conveniently expressed in terms of the “velocity”:

$$V_i^{(n)} = \frac{1}{2} \epsilon_{npq} \vec{b}^{(p)} \cdot \partial_i \vec{b}^{(q)}, \quad (2.13)$$

which represents the “rate of rotation” (in the  $i$ th direction) of the triad about its  $n$ th axis. This quantity is precisely the same as the quantity  $K_{in}$  employed by Nye,<sup>24</sup> and called by him the curvature tensor. Bilby and Smith,<sup>25</sup> and Kléman<sup>26</sup> have shown (in Nye’s notation) the equivalent of

$$\partial_i V_j^{(n)} - \partial_j V_i^{(n)} = \vec{b}^{(n)} \cdot \partial_i \vec{b}^{(n)} \times \partial_j \vec{b}^{(n)}, \quad (2.14)$$

a relationship also found for <sup>3</sup>He-A by Mermin and Ho.<sup>27</sup>

From (2.13),

$$\epsilon_{npq} \epsilon_{np'q'} = \delta_{pp'} \delta_{qq'} - \delta_{pq'} \delta_{qp'},$$

and  $\partial_i (\vec{b}^{(p)} \cdot \vec{b}^{(q)}) = 0$ , we have that

$$\epsilon_{npq} V_i^{(n)} = \vec{b}^{(p)} \cdot \partial_i \vec{b}^{(q)}. \quad (2.15)$$

Then

$$\begin{aligned} \partial_i \vec{b}^{(n)} &= \vec{b}^{(p)} (\vec{b}^{(p)} \cdot \partial_i \vec{b}^{(n)}) \\ &= \frac{1}{2} \epsilon_{pq'r} \vec{b}^{(q')} \times \vec{b}^{(r')} \epsilon_{pnq} V_i^{(q)} \\ &= \vec{b}^{(n)} \times (\vec{b}^{(q)} V_i^{(q)}). \end{aligned} \quad (2.16)$$

This explicitly shows that gradients of the unit vector  $\vec{b}^{(n)}$  are perpendicular to  $\vec{b}^{(n)}$ . We will employ (2.16) later, when we consider the dynamics of  $\vec{b}^{(n)}$ .

Note that the biaxial nematic is expected to have an anisotropic magnetic susceptibility  $\vec{\chi}$ . Assuming that  $\vec{\chi}$  is diagonalized in the local reference frame  $\{\vec{b}^{(n)}\}$ , we have

$$\vec{m} = \vec{\chi} \cdot \vec{h}, \quad (2.17)$$

$$\vec{\chi} = \sum_{n=1}^3 \chi_n \vec{b}^{(n)} \vec{b}^{(n)}. \quad (2.18)$$

Thus, in (2.11),  $\vec{h} \times \vec{m} \neq \vec{0}$  in general.

For completeness, we give the bending energy density  $\epsilon_B$ , which yields the terms  $\phi_{ij}^{(n)}$  in (2.5). It is convenient to write it in terms of the  $V_i^{(n)}$ ’s

$$\begin{aligned} \epsilon_B &= \frac{1}{2} \sum_{m,n} A_{m,n} (\vec{b}^{(m)} \cdot \vec{\nabla}^{(n)})^2 \\ &+ \frac{1}{2} \sum_{m \neq n} B_{m,n} (\vec{b}^{(m)} \cdot \vec{\nabla}^{(m)}) (\vec{b}^{(n)} \cdot \vec{\nabla}^{(n)}) \\ &+ \frac{1}{2} \sum_{m \neq n} C_{m,n} (\vec{b}^{(m)} \cdot \vec{\nabla}^{(n)}) (\vec{b}^{(n)} \cdot \vec{\nabla}^{(m)}). \end{aligned} \quad (2.19)$$

The stability of the uniform state requires that  $\epsilon_B > 0$  for all nonuniform textures. This yields the stability conditions:

$$\begin{aligned} A_{m,n} &> 0, \quad \text{all } m, n \\ A_{m,m} A_{n,n} &> B_{m,n}^2, \quad m \neq n \\ A_{m,n} A_{n,m} &> C_{m,n}^2, \quad m \neq n. \end{aligned} \quad (2.20)$$

One may reduce (2.19) from 15 terms to 12, by introducing three surface terms (M. Kléman, private communication). The identities which make this possible depend upon (2.14). [Note that Trebin has given a form equivalent to (2.19), employing a set of variables which make evident the biaxial nematic  $\rightarrow$  uniaxial nematic transition.<sup>28</sup> However, (2.19) is more appropriate to fully developed biaxial symmetry.] Explicitly, the identities have the form

$$\begin{aligned} \vec{v}^{(3)} \cdot \vec{\nabla} \times \vec{b}^{(3)} &= \vec{\nabla} \cdot [\vec{b}^{(3)} \times \vec{v}^{(3)} + \frac{1}{2} \vec{b}^{(3)} \cdot \vec{\nabla} \cdot \vec{b}^{(3)} \\ &+ \frac{1}{2} \vec{b}^{(3)} \times \vec{\nabla} \times \vec{b}^{(3)}]. \end{aligned} \quad (2.21)$$

To be applied to (2.19), we rewrite  $\vec{v}^{(3)} \cdot \vec{\nabla} \times \vec{b}^{(3)}$  as

$$\begin{aligned} \vec{v}^{(3)} \cdot \vec{\nabla} \times \vec{b}^{(3)} &= \vec{b}^{(1)} \cdot \vec{\nabla}^{(1)} \vec{b}^{(3)} \cdot \vec{v}^{(3)} - \vec{b}^{(1)} \cdot \vec{\nabla}^{(3)} \vec{b}^{(3)} \cdot \vec{v}^{(1)} \\ &+ \vec{b}^{(2)} \cdot \vec{\nabla}^{(2)} \vec{b}^{(3)} \cdot \vec{v}^{(3)} - \vec{b}^{(2)} \cdot \vec{\nabla}^{(3)} \vec{b}^{(3)} \cdot \vec{v}^{(2)}. \end{aligned} \quad (2.22)$$

Thus the six  $B_{m,n}$  and  $C_{m,n}$  terms can be reduced to three surface terms and three bulk terms. One can eliminate the three  $B_{m,n}$  terms or the three  $C_{m,n}$  terms, for example.

### III. THERMODYNAMIC EQUILIBRIUM AND THERMODYNAMIC DRIVING FORCES

To obtain the conditions for thermodynamic equilibrium, we permit the system to interchange entropy, mass, momentum, angular momentum, and magnetization with a reservoir. In addition, we must vary the order parameter. When the energy of the system plus reservoir has been minimized, equilibrium has been reached. Alternately, we may minimize the “free energy”

$$F = \int d\vec{r} [\epsilon - T_0 S - \mu_0 \rho - \vec{v}_0 \cdot \vec{g} - \vec{\omega}_0 \cdot (\vec{r} \times \vec{g} + \gamma^{-1} \vec{m})]. \quad (3.1)$$

Thus, with (2.5) and (3.1), we obtain the following equilibrium conditions:

$$T = T_0, \text{ or } \partial_i T = 0, \quad (3.2)$$

$$\mu = \mu_0, \text{ or } \partial_i \mu = 0, \quad (3.3)$$

$$\vec{v} = \vec{v}_0 + \omega_0 \times \vec{r}, \text{ or } A_{ij} = 0, \quad \partial_j \omega_i = 0,$$

$$A_{ij} \equiv \frac{1}{2}(\partial_i v_j + \partial_j v_i), \quad \omega_i \equiv \frac{1}{2}(\vec{\nabla} \times \vec{v})_i, \quad (3.4)$$

$$\vec{h}^{(e)} = 0, \quad \vec{h}^{(e)} \equiv \vec{h} - \vec{H} - \gamma^{-1} \vec{\omega}. \quad (3.5)$$

[In (3.5), we have used  $\vec{\omega} = \vec{\omega}_0$  in equilibrium.]

Note that, in (3.1), we include both the orbital ( $\vec{r} \times \vec{g}$ ) and spin ( $\gamma^{-1} \vec{m}$ ) angular momentum densities. In practice, however, when a macroscopic object rotates, its total orbital angular momentum vastly exceeds its total spin angular momentum. Also, note that (usually)  $\gamma^{-1} \omega \ll H$ , so that  $\vec{h} \approx \vec{H}$  in rotating equilibrium. In fact, neglect of the spin angular momentum in (3.1) yields  $\vec{h} = \vec{H}$  in rotating equilibrium.

To vary the order parameter, we subject *it alone* to a rotation  $\delta \vec{\Theta}_0$ , finding that:

$$\vec{0} = \frac{\delta F}{\delta \vec{\Theta}_0}, \text{ or } \vec{\Psi} = 0,$$

$$\vec{\Psi} \equiv \vec{b}^{(n)} \times \vec{\Phi}^{(n)}. \quad (3.6)$$

From (3.1)–(3.6) we have the thermodynamic driving forces for biaxial nematic liquid crystals. In particular, note that  $\partial_j v_i$  is not a thermodynamic driving force, but that  $A_{ij}$  and  $\partial_j \omega_i$  are. Other “forces” are  $\partial_i T$ ,  $\partial_i \mu$ , and  $h_i^{(e)}$ . We will also find it necessary to consider  $\partial_j h_i^{(e)}$ , which has the same behavior under time reversal and space inversion as does  $\partial_j \omega_i$ . Finally,  $\vec{\Psi}$  is the force which drives the “texture” of the system, and is specifically associated with the biaxial nematic degrees of freedom.

Note that, for fluids possessing internal angular

momentum, one must also consider a new quantity, similar to  $h_i^{(e)}$ , as a thermodynamic driving force. This and related points are discussed in the Appendix.

It should be observed that rotating equilibrium is not simply a rotated version of static equilibrium. First, for all systems the chemical potential  $\mu = \mu_{\text{rest}} - \frac{1}{2} \omega^2 r^2$ , where  $r$  is the distance to the axis of rotation, a point discussed microscopically by Ref. 22. It is  $\mu$  that remains constant; the extra term  $-\frac{1}{2} \omega^2 r^2$  (the “centrifugal potential”) tends to drive matter away from the axis. Second,  $\vec{h}$  now develops a new  $\gamma^{-1} \vec{\omega}$  term, causing  $\vec{m}$  to develop a new  $\vec{\chi} \cdot \gamma^{-1} \vec{\omega}$  term. Furthermore, because  $\vec{\chi}$  depends on the orientation of the triad axes, the triad axes rotate in order to minimize the interaction  $-\vec{m} \cdot \vec{H}$  with the field  $\vec{H}$ .

### IV. EQUATIONS OF MOTION

We now must present equations of motion or conservation laws for  $\epsilon$ ,  $S$ ,  $\rho$ ,  $\vec{g}$ ,  $\vec{m}$ , and  $\vec{b}^{(n)}$ . In addition, we must ensure that the angular momentum density is conserved. We have

$$\frac{\partial \epsilon}{\partial t} + \partial_i j_i^\epsilon = 0, \quad (4.1)$$

where  $j_i^\epsilon$  is the (unknown) energy flux,

$$\frac{\partial S}{\partial t} + \partial_i j_i^s = R \geq 0, \quad (4.2)$$

where  $j_i^s$  is the (unknown) entropy flux and  $R$  is the (unknown) rate of entropy production per unit volume,

$$\frac{\partial \rho}{\partial t} + \partial_i j_i^\rho = 0, \quad (4.3)$$

where the mass flux  $j_i^\rho$  is known (from microscopic physics) to be equal to  $g_i$ ,

$$\frac{\partial g_i}{\partial t} + \partial_j \sigma_{ij} = m_j \partial_i H_j, \quad (4.4)$$

where  $\sigma_{ij}$  is the unknown momentum flux density and  $m_j \partial_i H_j$  is the magnetic force acting on the system. Because of

$$\vec{B} \equiv \vec{H} + 4\pi \vec{m}, \quad \vec{\nabla} \cdot \vec{B} = 0, \quad \vec{\nabla} \times \vec{H} = 0, \quad (4.5)$$

the last of these holding only for no current-flow or time-varying electric field  $\vec{E}$ , we may rewrite (4.4) as

$$\frac{\partial g_i}{\partial t} + \partial_j \sigma'_{ij} = 0, \quad (4.6)$$

$$\sigma'_{ij} \equiv \sigma_{ij} - (4\pi)^{-1} (B_i H_j - \frac{1}{2} \delta_{ij} H^2) . \quad (4.7)$$

We also have

$$\frac{\partial m_i}{\partial t} + \partial_i J_{ij} = J_i + \gamma [\vec{m} \times (\vec{H} - \vec{h})]_i , \quad (4.8)$$

where  $J_{ij}$  is the (unknown) magnetization flux density,  $\gamma \vec{m} \times (\vec{H} - \vec{h})$  is the known torque,  $\gamma$  is the gyromagnetic ratio, and  $j_i$  represents any unknown torque.<sup>29</sup> For the order parameter, we write

$$\frac{\partial \vec{b}^{(n)}}{\partial t} = \vec{\Omega} \times \vec{b}^{(n)} , \quad (4.9)$$

where  $\vec{\Omega}$  is an (unknown) rotation rate.

Note that, by assuming  $\vec{H}$  fixed, we are able to permit the electromagnetic field to exchange momentum and angular momentum, but not energy. We may think of the source of  $\vec{H}$  as being rigidly attached to an infinitely massive rigid body. Thus energy is not exchanged when it absorbs momentum or angular momentum.

We now consider angular momentum conserva-

tion in detail. With the total angular momentum density given by

$$\vec{\mathcal{J}} = \vec{r} \times \vec{g} + \gamma^{-1} \vec{m} , \quad (4.10)$$

we have

$$\frac{\partial \mathcal{J}_i^j}{\partial t} + \partial_j \mathcal{J}_i^j = (\vec{m} \times \vec{H})_i , \quad (4.11)$$

where  $\mathcal{J}_i^j$  is the (unknown) angular momentum flux and  $(\vec{m} \times \vec{H})_i$  is the external torque. For a fixed position  $\vec{r}$ , use of (4.6), (4.8), (4.10), and (4.11) yield that the quantity

$$\sigma'_i \equiv \epsilon_{ijk} \sigma'_{jk} \quad (4.12)$$

is given by

$$\sigma'_i = \gamma^{-1} J_i - (\vec{m} \times \vec{h})_i + \partial_l [\mathcal{J}_i^l - \gamma^{-1} J_{il} - \epsilon_{ijk} r_j \sigma'_{kl}] . \quad (4.13)$$

This relation enables us to eliminate the antisymmetric part of the momentum flux density in terms of other quantities, most notably the angular momentum flux density.

## V. HYDRODYNAMIC EQUATIONS

The standard approach of phenomenological hydrodynamics is to obtain the rate of heat production, per unit volume, in terms of a product of thermodynamic forces and fluxes. We thus start with (4.2) multiplied by  $T$ :

$$0 \leq TR \leq T \frac{\partial S}{\partial t} + T \partial_i j_i^s . \quad (5.1)$$

We then utilize (2.5) to obtain  $\partial S / \partial t$  in terms of the other time derivatives given in Sec. IV. We then obtain (with  $j_i^p = g_i$ )

$$0 \leq TR = -\partial_i j_i^e + \mu \partial_i g_i + T \partial_i j_i^s + v_i \partial_j \sigma'_{ij} - \vec{\Omega} \cdot \vec{b}^{(n)} \times \vec{\phi}^{(n)} - \phi_{ik}^{(n)} \partial_k (\vec{\Omega} \times \vec{b}^{(n)})_i - J_i (h_i - H_i) + (h_j - H_j) \partial_i J_{ji} . \quad (5.2)$$

Integrating by parts on the second, third, fourth, sixth, and eighth terms, we obtain

$$0 \leq TR = -\partial_i [j_i^e - \mu g_i - T j_i^s - v_j \sigma'_{ji} + \vec{\Omega} \cdot \vec{b}^{(n)} \times \vec{\phi}_{-i}^{(n)} - J_{ji} (h_j - H_j)] - g_i \partial_i \mu - j_i^s \partial_i T - \sigma'_{ij} \partial_j v_i - \vec{\Omega} \cdot \vec{\Psi} - J_i (h_i - H_i) - J_{ji} \partial_i (h_j - H_j) . \quad (5.3)$$

Now we take the step which brings in the Galilean transformation properties of the system. We rewrite the second term of (5.3), using (2.7)

$$\begin{aligned} -g_i \partial_i \mu &= -v_i (\rho \partial_i \mu) \\ &= -v_i [\partial_i P - S \partial_i T - g_j \partial_i v_j - m_j \partial_i (h_j - H_j) - b_j^{(n)} \partial_i \phi_j^{(n)} - \partial_k b_j^{(n)} \partial_i \phi_{jk}^{(n)}] \\ &= -\partial_i [v_i (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_k b_j^{(n)} \phi_{jk}^{(n)}) + \phi_{ji}^{(n)} (\vec{v} \cdot \vec{\nabla}) b_j^{(n)}] \\ &\quad + [g_i v_j + \delta_{ij} (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_k b_j^{(n)} \phi_{jk}^{(n)}) + \partial_i b_n^{(n)} \phi_{kj}] \partial_j v_i \\ &\quad + S v_i \partial_i T + m_j v_i \partial_i (h_j - H_j) - \Phi_j^{(n)} v_i \partial_i b_j^{(n)} . \end{aligned} \quad (5.4)$$

Putting (5.4) into (5.3), we obtain

$$\begin{aligned}
0 \leq TR = & -\partial_i [j_i^\epsilon - \mu g_i - T j_i^s - v_j \sigma'_{ji} + \vec{\Omega} \cdot \vec{b}^{(n)} \times \vec{\phi}_{-i}^{(n)} - J_{ji} (h_j - H_j) \\
& + v_i (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_k b_j^{(n)} \phi_{jk}^{(n)}) + \phi_{ji}^{(n)} (\vec{\nabla} \cdot \vec{\nabla}) b_j^{(n)}] \\
& - (j_i^s - S v_i) \partial_i T - (J_{ji} - m_j v_i) \partial_i (h_j - H_j) - J_i (h_i - H_i) \\
& - [\sigma'_{ij} - g_i v_j - \delta_{ij} (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_n b_j^{(n)} \phi_{jk}^{(n)}) - \partial_i b_k^{(n)} \phi_{kj}] \partial_j v_i - (\vec{\Omega} - \vec{b}^{(q)} \vec{\nabla} \cdot \vec{\nabla}^{(q)}) \cdot \vec{\Psi} . \quad (5.5)
\end{aligned}$$

Note that (2.16) was employed to rewrite the last term of (5.4):

$$-\Phi_j^{(n)} v_i \partial_i b_j^{(n)} = -\vec{\Phi}^{(n)} \cdot v_i (\vec{b}^{(n)} \times \vec{b}^{(q)}) \mathcal{V}_i^{(q)} = (\vec{b}^{(n)} \times \vec{\Phi}^{(n)}) \cdot \vec{b}^{(q)} (\vec{\nabla} \cdot \vec{\nabla}^{(q)}) = (\vec{\Psi} \cdot \vec{b}^{(q)}) (\vec{\nabla} \cdot \vec{\nabla}^{(q)}) . \quad (5.6)$$

Equation (5.5) is quite close to the desired form, but we must still replace  $\vec{h} - \vec{H}$  by  $\vec{h}^{(e)}$  [cf. (3.5)], and  $\partial_j v_i$  by  $A_{ij}$  and  $\omega_i$  [cf. (3.4)]. Let us define

$$\Sigma'_{ij} \equiv \sigma'_{ij} - g_i v_j - \delta_{ij} (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_k b_j^{(n)} \phi_{jk}^{(n)}) - \partial_i b_k^{(n)} \phi_{kj} \quad (5.7)$$

and decompose it as follows:

$$\Sigma'_{ij} = \Sigma'_{ij}{}^{(s)} + \frac{1}{2} \epsilon_{ijk} \Sigma'_k , \quad (5.8)$$

$$\Sigma'_{ij}{}^{(s)} \equiv \frac{1}{2} (\Sigma'_{ij} + \Sigma'_{ji}) , \quad (5.9)$$

$$\Sigma'_k \equiv \epsilon_{ijk} \Sigma'_{jk} . \quad (5.10)$$

With

$$\partial_j v_i = A_{ij} + \epsilon_{ijk} \omega_k , \quad (5.11)$$

which follows from (3.4), we have

$$\Sigma'_{ij} \partial_j v_i = \Sigma'_{ij}{}^{(s)} A_{ij} - \Sigma'_k \omega_k . \quad (5.12)$$

Use of (5.12) and (3.5) in (5.5) then yields [with the notation  $\dots$  to indicate that the corresponding terms from (5.5) are to be carried over]

$$\begin{aligned}
0 \leq TR = & -\partial_i [j_i^\epsilon + \dots + \gamma^{-1} (J_{ji} - m_j v_i) \omega_j] - (j_i^s - S v_i) \partial_i T - (J_{ji} - m_j v_i) h_j^{(e)} - J_i h_i^{(e)} \\
& - \Sigma'_{ij}{}^{(s)} A_{ij} - (\vec{\Omega} - \vec{b}^{(q)} \vec{\nabla} \cdot \vec{\nabla}^{(q)}) \cdot \vec{\Psi} + \{ \Sigma'_i - \gamma^{-1} [J_i - \partial_j (J_{ij} - m_i v_j)] \} \omega_i . \quad (5.13)
\end{aligned}$$

Equation (5.13) is almost in the desired form. Except for the first and last terms, it involves products of (unknown) fluxes times (presumably known) forces. We now rewrite the last term, employing (4.12), (4.13), (5.7), and (2.11). This step incorporates angular momentum conservation and the invariance of the internal energy under rotations. It yields

$$\begin{aligned}
\Sigma'_i - \gamma^{-1} [J_i - \partial_j (J_{ij} - m_i v_j)] &= \sigma'_i - \epsilon_{ijk} \partial_j b_l^{(n)} \phi_{lk}^{(n)} - \gamma^{-1} [J_i - \partial_j (J_{ij} - m_i v_j)] \\
&= -(\vec{m} \times \vec{h})_i - \epsilon_{ijk} \partial_j b_l^{(n)} \phi_{lk}^{(n)} + \partial_l (\mathcal{J}_i^l - \epsilon_{ijk} r_j \sigma'_{kl} - \gamma^{-1} m_i v_l) \\
&= \Psi_i + \partial_l (\mathcal{J}_i^l - \epsilon_{ijk} r_j \sigma'_{kl} - \gamma^{-1} m_i v_l - \epsilon_{ijk} \phi_{jl}^{(n)} b_k^{(n)}) . \quad (5.14)
\end{aligned}$$

Placing (5.14) into (5.13), rearranging the  $\Psi_i \omega_i$  term, and integrating by parts on the remaining  $\omega_i$  terms, we finally obtain

$$\begin{aligned}
0 \leq TR = & -\partial_i [j_i^\epsilon - \tilde{j}_i^\epsilon - \omega_j (\mathcal{J}_i^j - \tilde{\mathcal{J}}_i^j)] - (j_i^s - S v_i) \partial_i T - (J_{ji} - m_j v_i) \partial_i h_j^{(e)} - J_i h_i^{(e)} - \Sigma'_{ij}{}^{(s)} A_{ij} \\
& - (\Omega_i - b_i^{(q)} \vec{\nabla} \cdot \vec{\nabla}^{(q)} - \omega_i) \Psi_i - (\mathcal{J}_i^j - \tilde{\mathcal{J}}_i^j) \partial_j \omega_i , \quad (5.15)
\end{aligned}$$

$$\tilde{j}_i^\epsilon \equiv \mu g_i + T j_i^s + v_j [\sigma'_{ji} - \delta_{ij} (P - \vec{b}^{(n)} \cdot \vec{\phi}^{(n)} - \partial_i b_k^{(n)} \phi_{kl}^{(n)}) - \partial_j b_k^{(n)} \phi_{ki}] + J_{ji} h_j^{(e)} - \vec{\Omega} \cdot \vec{b}^{(n)} \times \vec{\phi}_{-i}^{(n)} + v_i \gamma^{-1} \vec{\omega} \cdot \vec{m} , \quad (5.16)$$

$$\tilde{\mathcal{J}}_i^j \equiv \epsilon_{ijk} (r_j \sigma'_{kl} + \phi_{jl}^{(n)} b_k^{(n)}) = \gamma^{-1} m_i v_l . \quad (5.17)$$

Since the divergence term in (5.15) could be of either sign, to avoid violating  $0 \leq TR$ , we set

$$J_i^\epsilon = \tilde{J}_i^\epsilon + \omega_j (\mathcal{J}_i^j - \tilde{\mathcal{J}}_i^j). \quad (5.18)$$

Of course,  $\tilde{J}_i^\epsilon$ ,  $\mathcal{J}_i^j$ , and  $\tilde{\mathcal{J}}_i^j$  contain other fluxes whose forms have not been determined. Nevertheless, the other fluxes can be found independently of  $\tilde{J}_i^\epsilon$ ,  $\mathcal{J}_i^j$ , and  $\tilde{\mathcal{J}}_i^j$ , so the logic is not impaired by employing (5.10) first. We are thus left with

$$0 \leq TR = -\tilde{J}_i^s \partial_i T - \tilde{J}_{ji} \partial_i h_j^{(e)} - J_i h_i^{(e)} - \Sigma_{ij}^{(s)} A_{ij} - (\mathcal{J}_i^j - \tilde{\mathcal{J}}_i^j) \partial_j \omega_i - \tilde{\Omega}_i \Psi_i, \quad (5.19)$$

$$\tilde{J}_i^s \equiv J_i^s - S v_i, \quad (5.20)$$

$$\tilde{J}_{ji} \equiv J_{ji} - m_i v_i, \quad (5.21)$$

$$\tilde{\Omega}_i \equiv \Omega_i - b_i^{(q)} \vec{\nabla} \cdot \vec{V}^{(q)} - \omega_i. \quad (5.22)$$

The object, then, is to find the unknown set of quantities ( $\tilde{J}_i^s$ ,  $\tilde{J}_{ji}$ ,  $J_i$ ,  $\Sigma_{ij}^{(s)}$ ,  $\mathcal{J}_i^j$ ,  $\tilde{\Omega}_i$ ) in terms of the set of thermodynamic forces ( $\partial_i T$ ,  $\partial_i h_j^{(e)}$ ,  $h_i^{(e)}$ ,  $A_{ij}$ ,  $\partial_j \omega_i$ ,  $\Psi_i$ ).

We now construct the reactive terms, employing only  $\epsilon_{ijk}$  and terms of the form

$$B_{ij} = \sum_{n=1}^3 B^{(n)} b_i^{(n)} b_j^{(n)}. \quad (5.23)$$

This omits terms proportional to  $\vec{m}$  and  $\vec{\omega}$ , which we expect to be small. Consistent with no contribution to  $TR$  in (5.19), we find (using the superscript R to denote the reactive part)

$$\tilde{J}_i^{(R)} = B_{ijk}^{(1)} \partial_k h_j^{(e)} + B_{ijk}^{(2)} \partial_k \omega_j, \quad (5.24)$$

$$B_{ijk}^{(1)} = \epsilon_{ijl} B_{lk}^{(1)} + \epsilon_{ilk} B_{lj}^{(2)} + \epsilon_{ijk} B_{li}^{(3)}, \quad (5.25)$$

where the  $B_{ij}^{(n)}$ 's have the same form as  $B_{ij}$  in (5.23), and  $B_{ijk}^{(2)}$  has the same form as  $B_{ijk}^{(1)}$ :

$$\tilde{J}_{ji}^{(R)} = -B_{kji}^{(1)} \partial_k T, \quad (5.26)$$

$$J_i^{(R)} = B_{ij}^{(4)} \Psi_j, \quad (5.27)$$

$$\Sigma_{ij}^{(s)(R)} = B_{ijk}^{(3)} \Psi_k, \quad (5.28)$$

$$\mathcal{J}_i^{j(R)} = \tilde{\mathcal{J}}_i^{j(R)} - B_{kij}^{(2)} \partial_k T, \quad (5.29)$$

$$\tilde{\Omega}_i^{(R)} = -B_{ji}^{(4)} h_j^{(e)} - B_{kji}^{(3)} A_{kj}. \quad (5.30)$$

Since  $\Sigma_{ij}^{(s)} = \Sigma_{ji}^{(s)}$ , we have  $B_{ijk}^{(3)} = B_{jik}^{(3)}$ , so

$$B_{ijk}^{(3)} = \epsilon_{ikl} B_{jl}^{(5)} + \epsilon_{jkl} B_{il}^{(5)}, \quad (5.31)$$

where  $B_{jl}^{(5)}$  has the same form as  $B_{ij}$  in (5.23).

The dissipative terms have the form (using the superscript D to denote the dissipative part)

$$\tilde{J}_i^{(D)} = -\frac{1}{T} \kappa_{ij} \partial_j T, \quad (5.32)$$

where  $\kappa_{ij}$  has the same form as  $B_{ij}$ :

$$\tilde{J}_{ji}^{(D)} = -D_{jikl}^{(1)} \partial_l h_k^{(e)} - D_{jikl}^{(2)} \partial_l \omega_k, \quad (5.33)$$

where  $D_{jikl}^{(1)}$  is a complicated magnetization diffusion tensor, possessing 15 independent components, and  $D_{jikl}^{(2)}$  has 21 independent components (the Onsager principle gives the symmetry  $D_{jikl}^{(1)} = D_{klji}^{(1)}$ , thus reducing the number of components in  $D_{jikl}^{(1)}$ ). Also,

$$J_i^{(D)} = -D_{ij}^{(1)} h_j^{(e)} - D_{kji}^{(1)} A_{kj}, \quad (5.34)$$

where  $D_{ij}^{(1)}$  is a relaxation tensor (giving  $T_1$  and  $T_2$  processes) having the same form as  $B_{ij}$ , and  $D_{kji}^{(1)}$  has the same form as  $B_{ijk}^{(3)}$ :

$$\Sigma_{ij}^{(s)(D)} = -D_{ijk}^{(1)} h_k^{(e)} - \eta_{ijkl} A_{kl}, \quad (5.35)$$

where  $\eta_{ijkl}$  is a viscosity tensor possessing 9 independent components ( $\eta_{ijkl} = \eta_{klji}$ ,  $\eta_{ijkl} = \eta_{jikl}$ ):

$$\mathcal{J}_i^{j(D)} = \tilde{\mathcal{J}}_i^{j(D)} - D_{ijk}^{(3)} \partial_l \omega_k - D_{klji}^{(2)} \partial_l h_k^{(e)}, \quad (5.36)$$

where  $D_{jikl}^{(3)}$  has the same form as  $D_{jikl}^{(1)}$ :

$$\tilde{\Omega}_i^{(D)} = -D_{ij}^{(2)} \Psi_j, \quad (5.37)$$

where  $D_{ij}^{(2)}$  has the same form as  $B_{ij}$ .

From (5.32) to (5.37), (5.19) becomes

$$0 \leq TR = T^{-1} \kappa_{ij} \partial_i T \partial_j T + D_{ij}^{(1)} h_i^{(e)} h_j^{(e)} + 2D_{ijk}^{(1)} h_k^{(e)} A_{ij} + \eta_{ijkl} A_{ij} A_{kl} + D_{ijk}^{(2)} \Psi_i \Psi_j + D_{ijk}^{(1)} \partial_j h_i^{(e)} \partial_l h_k^{(e)} + 2D_{ijk}^{(2)} \partial_j h_i^{(e)} \partial_l \omega_k + D_{ijk}^{(3)} \partial_j \omega_i \partial_l \omega_k. \quad (5.38)$$

This enforces a number of restrictions on the dissipative coefficients, which we will not attempt to enumerate.

We close this section by noting that, in rotating equilibrium, where the driving forces go to zero, it is straightforward to show that the entropy and mass densities only convect at the local velocity  $\vec{v}$ . Further, the momentum density, magnetization, and the rotation rate  $\vec{\Omega}$  all change both with the local velocity  $\vec{v}$  and the local angular velocity  $\vec{\omega}$ . In particular, one may show, in rotating equilibrium, that the triad vectors satisfy

$$\frac{\partial \vec{b}^{(n)}}{\partial t} = \vec{\omega} \times \vec{b}^{(n)} - (\vec{v} \cdot \vec{\nabla}) \vec{b}^{(n)}, \quad (5.39)$$

a result which reduces to that for nematics if we restrict  $\vec{b}^{(n)}$  to the nematic axis  $\hat{n}$ .

## VI. CONCLUSION

We have derived the hydrodynamics of biaxial nematics with arbitrary nonsingular textures. The choice of independent thermodynamic variables ( $S, \rho, \vec{g}, \vec{m}, \{\vec{b}^{(n)}\}$ ) led to the corresponding set of thermodynamic "forces" ( $\partial_i T, \partial_i \mu, A_{ij}, \partial_j \omega_i, h_i^{(e)}, \Psi_i$ ). Had we neglected  $\vec{m}$  and solved for the normal modes of the uniform system, we would have found a pair of propagating modes (ordinary sound, associated with  $\rho$  and  $\vec{k} \cdot \vec{g}$ , where  $\vec{k}$  is the wave vector) a thermal diffusion mode (associated with  $S$ ), and five other diffusion modes (associated with  $\vec{k} \times \vec{g}$  and  $\{\vec{b}^{(n)}\}$ ). Of course, it is possible that some of the modes associated with  $\{\vec{b}^{(n)}\}$  are propagating, as is also possible in uniaxial nematics<sup>4</sup>; but, since such modes are diffusive in uniaxial nematics, one anticipates that they will be diffusive in biaxials. The presence of an additional mode distinguishes biaxials from uniaxial nematics and may serve as a definitive signature of this phase. Note that, by increasing the symmetry of various tensors, one can make the hydrodynamics of biaxial nematics go into that for uniaxial nematics. In that way, for example, one can extend the reversible nonlinear uniaxial nematic hydrodynamics,<sup>30</sup> to include the dissipative terms.

In closing, we repeat that explicit treatment of angular momentum conservation, as well as detailed consideration of the equilibrium conditions, leads to hydrodynamic equations which clearly possess a rotating-equilibrium solution. We should mention, however, that there exist other treatments in the literature where angular momentum is explicitly conserved. (Certain of these are mentioned in Ref. 12.) Probably, the work of Snider and Lewchuk,<sup>31</sup> on fluids whose molecules possess internal angular momentum, is most like ours in spirit. In the Appendix we indicate how to incorporate internal angular momentum into the present problem.

*Note added in proof.* Since submitting this paper, I have become aware of three other related works. They are by H. Brand and H. Pleiner, by E. A. Jacobsen and J. Swift, and by M. Liu.

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## APPENDIX

In the bulk of the present paper, we have completely neglected the effects of the internal angular

momentum of the molecules constituting the biaxial nematic. In this Appendix, we will indicate how the theory can be so extended, and we will present some references to work on ordinary fluids with internal angular momentum.

Let us assume, therefore, that the system also possesses a thermodynamic variable  $\vec{l}$ , the internal angular momentum density. This requires that we add  $\vec{\Gamma} \cdot d\vec{l}$  to  $d\epsilon$  [the right-hand sides of Eqs. (2.2) and (2.5)], that we add  $\vec{\Gamma} \cdot \vec{l}$  to  $\epsilon + P$  [the right-hand side of Eq. (2.6)], and that we add  $\vec{l} \cdot d\vec{\Gamma}$  to  $dP$  [Eq. (2.7)]. Here  $\vec{\Gamma} \equiv \partial\epsilon/\partial\vec{l}$ , with the appropriate variables held constant. As a consequence we must add  $\vec{\Gamma} \times \vec{l}$  to the right-hand side of (2.10) and (2.11), to express  $d\epsilon_{\text{int}}=0$  under uniform rotations.

We must also add a term

$$- \int \vec{\omega}_0 \cdot \vec{l} d\vec{r}$$

to  $F$  in (3.1), and we then find an additional condition, on varying  $\vec{l}$ :  $\vec{\Gamma} - \vec{\omega} = 0$  in equilibrium.

The equation of motion for  $\vec{l}$  is taken to be

$$\frac{\partial l_i}{\partial t} + \partial_j \mathcal{L}_{ij} = \mathcal{L}_i + (\vec{l} \times \vec{\Gamma})_i,$$

and in (4.10) for  $\vec{\mathcal{F}}$  we must add  $\vec{l}$  to the right-hand side. This redefinition of  $\vec{\mathcal{F}}$  requires a redefinition of  $\vec{\sigma}'$ : to the right-hand side of (4.13) we must add  $-\vec{l} \times \vec{\Gamma}$  (which will be zero if  $\vec{l}$  is along  $\vec{\Gamma}$ ).

A bit of reflection yields that  $\vec{l}$  and  $\vec{\Gamma}$  are analogous to  $\gamma^{-1}\vec{m}$  and  $\gamma(\vec{h} - \vec{H})$ , with  $\mathcal{L}_i$  and  $\mathcal{L}_{ij}$  analogous to  $\gamma^{-1}J_i$  and  $\gamma^{-1}J_{ij}$ . Thus, we may obtain the final results by making appropriate changes. We must redefine  $\tilde{j}_i^\epsilon$  by adding

$$\mathcal{L}_{ij}(\Gamma_j - \omega_j) + v_i \vec{\omega} \cdot \vec{l}$$

to the right-hand side of (5.16); and redefine  $\tilde{\mathcal{F}}_i^l$  by adding  $l_i v_i$  to the right-hand side of (5.17). Then (5.18) is unchanged. With the definition

$$\tilde{\mathcal{L}}_{ij} \equiv \mathcal{L}_{ij} - l_i v_j,$$

we then find that the entropy production equation, (5.19) develops the terms

$$- \tilde{\mathcal{L}}_{ji} \partial_i (\Gamma_j - \omega_j) - \mathcal{L}_i (\Gamma_i - \omega_i)$$

added to its right-hand side. With these changes, one may proceed to obtain the new terms  $\mathcal{L}_{ji}$  and  $\mathcal{L}_i$ , and the additional pieces that must be added to the old terms.



We only sketch the resulting analysis. *For the reactive parts:*

(1)  $\tilde{J}_i^{(R)}$  gets a  $\partial_k(\Gamma_j - \omega_j)$  term and  $\tilde{\mathcal{L}}_{ji}^{(R)}$  gets a corresponding  $\partial_k T$  term.

(2)  $\tilde{\Omega}_i^{(R)}$  gets a  $(\Gamma_j - \omega_j)$  term and  $\mathcal{L}_i^{(R)}$  gets a corresponding  $\Psi_k$  term. *For the dissipative parts:*

(1)  $\mathcal{L}_{ji}^{(D)}$  gets  $\partial_l h_k^{(e)}$ ,  $\partial_l \omega_k$ , and  $\partial_l(\Gamma_k - \omega_k)$  terms; and  $\tilde{J}_{ji}^{(D)}$  and  $\mathcal{F}_i^{(D)}$  get corresponding  $\partial_l(\Gamma_k - \omega_k)$  terms.

(2)  $\mathcal{L}_i^{(D)}$  gets  $h_k^{(e)}$ ,  $A_{kj}$ , and  $(\Gamma_k - \omega_k)$  terms; and  $J_i^{(D)}$  and  $\Sigma_{ij}^{(s)(D)}$  get corresponding  $(\Gamma_k - \omega_k)$  terms.

To remain true to the spirit of hydrodynamics, which only can properly include motions with no restoring forces (reactive or dissipative) in the long-wavelength limit, we must always keep  $\vec{h}^{(e)} \equiv \vec{h} - \vec{H} - \gamma^{-1} \vec{\omega}$  and  $\vec{\Gamma} - \vec{\omega}$  set to zero. The former means that  $T_1$  and  $T_2$  relaxation processes have brought the magnetization into equilibrium, and the latter means that analogous processes ("vortex viscosity"<sup>32</sup>) have brought the molecular rotation rate into equilibrium. However, because  $\vec{H}$  is at our experimental disposal, and it can be made to oscillate very quickly, in practice situations can readily be produced where  $\vec{h}^{(e)} \neq \vec{0}$ . In that case, the  $\vec{h}^{(e)} \neq 0$  equations, although not literally correct because the relaxation rate may in practice not be frequency independent, will be needed. On the other hand for most situations of hydrodynamic interest,  $\vec{\Gamma} - \vec{\omega} \approx \vec{0}$ . [Of course, one can consider probes in the optical frequency regime ( $\sim 10^{15}$  Hz), faster than the inverse of the relaxation time  $\tau$  for  $\vec{\Gamma} - \vec{\omega}$ . This is near  $10^{-11}$  sec, from computations for nitrogen near its boiling point.<sup>32</sup> However, such high-frequency probes are not hydrodynamic.] Thus, in  $d\epsilon$  the terms  $\vec{v} \cdot d\vec{g} + \vec{\Gamma} \cdot d\vec{\Gamma}$  become

$$\vec{v} \cdot d\vec{g} + \frac{1}{2}(\vec{\nabla} \times \vec{v}) \cdot d\vec{\Gamma} = \frac{1}{2} \vec{\nabla} \cdot (\vec{v} \times d\vec{\Gamma}) + \vec{v} \cdot d(\vec{g} + \frac{1}{2} \vec{\nabla} \times \vec{\Gamma}).$$

If the surface term can be dropped (e.g., if  $\vec{v} = \vec{0}$

on the surface; or if  $\vec{\Gamma}$  is normal to the surface), then by letting  $\vec{g} \rightarrow \vec{g}' = \vec{g} + \frac{1}{2} \vec{\nabla} \times \vec{\Gamma}$ , we can attempt to do the hydrodynamics with a redefined momentum density. However, the form  $\vec{v} \times \vec{g} + \vec{\Gamma} \times \vec{\Gamma}$  needed for angular momentum conservation [see (2.10)], which reduces to  $\vec{\Gamma} \times \vec{\Gamma}$ , and is  $\frac{1}{2}(\vec{\nabla} \times \vec{v}) \times \vec{\Gamma}$  for  $\vec{\Gamma} = \vec{\omega}$ , takes the form  $\vec{v} \times \vec{g}' = \vec{v} \times (\frac{1}{2} \vec{\nabla} \times \vec{\Gamma})$ . In general, these two expressions will not be equal, so a straightforward elimination of  $\vec{\Gamma}$  through the use of  $\vec{g}'$  will not work. However, if we set  $\vec{\Gamma} = \vec{\omega}$ , all of the new contributions to the "old" fluxes (which neglect  $\vec{\Gamma}$  entirely) disappear. Thus, although  $\vec{\Gamma}$  cannot be neglected in the thermodynamics, its associated variable  $\vec{\Gamma}$  does not explicitly enter into the fluxes which drive the equations of motion (except for the total angular momentum density  $\vec{J}$ , which certainly does sense the presence of  $\vec{\Gamma}$ ). For most purposes, it means that  $\vec{\Gamma}$  can be neglected. However, it should be noted that for very small samples this may not be a good approximation; then the center-of-mass angular momentum may not completely overwhelm the internal angular momentum. See, for example, the work of Kagan and Maksimov.<sup>33</sup> These authors consider, among other things, the equilibration of a cylinder containing diatomic molecules, with  $\vec{v} = \vec{0}$  and  $\vec{\Gamma} \neq \vec{0}$  initially: they find that the equilibrium value of  $\Gamma (= \omega)$  is down by a factor of  $a^2/L^2$  from its initial value, where  $a$  is the molecular radius and  $L$  is the cylinder radius. (To eliminate wall friction, one may assume all wall collisions to be specular.) This is simply the ratio of the individual molecular internal moment of inertia to the typical molecular center-of-mass moment of inertia.

Further references on the hydrodynamics of molecules with internal angular momenta may be found in Refs. 31 and 33. Note that Ref. 33 indicates that the  $D_{ijkl}^{(3)}$  of (5.36) are zero for point particles. By the positivity of (5.38), this also means the  $D_{ijkl}^{(2)}$  are zero in that case.

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