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# A MODEL FOR TOTAL ENERGY OF NEMATIC ELASTOMERS WITH NON-UNIFORM PROLATE SPHEROIDS

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Abstract. We provide a model for the total energy of nematic liquid crystal elastomers whose elastic solid backbones have an average shape of non-uniform prolate spheroids along the director of crosslinked liquid crystalline molecules. This energy is a generalization of the Oseen-Frank free energy for nematic liquid crystals, but incorporates coherently the elastic deformation with changes in the shape and the director of the network. The total free energy we obtain for the elastomer is suitable for the existence of energy minimizers and other mathematical studies of the elastomers and also provides a concrete example that is consistent with both the continuum mechanics theory and the molecular-statistical theory.

1. **Introduction.** Nematic elastomers are unusual materials that simultaneously combine the elastic properties of rubbers with the anisotropy of liquid crystals. They consist of polymer networks of elastic solid chains (the backbones) formed by the cross-linking of nematic molecules (the mesogens) as the elements of their main-chains and/or pendant side-groups. Because of this structure, any stress on the polymer network influences the nematic order of the elastomer, and, conversely, any change in the nematic order will affect the mechanical shape of the elastomer. The interplay between elastic and orientational changes is responsible for many fascinating properties of such materials that are different from the classical elastic solids and liquid crystals. These materials have been actively studied for both fundamental research and industrial applications; see, e.g., References [2, 4, 7, 8, 9, 11, 12, 14, 16].

The nematic elastomers we shall model in this paper have a common feature that the average shape of backbone nematic polymer is a prolate spheroid with the nematic director as its long axis. Such an elastomer can be formed in two steps: First, in the melt the material is an isotropic elastic rubber with no defined orientation. Under some preferred procedures (via applied fields or temperature change) the nematic chooses an initial shape of a spheroid with a fixed direction $\mathbf{n}_0$ and *asphericity* $q_0 \geq 1$ (i.e., the aspect ratio of lengths parallel and perpendicular to the spheroid axis $\mathbf{n}_0$). Then, the network undergoes a deformation $\mathbf{y}$ from the reference domain to a current domain where the nematic takes a shape of a new shape of prolate spheroid at a deformed point $z$ with axis $\tilde{\mathbf{n}}(z)$ and asphericity $\tilde{q}(z)$. We assume the elastic deformation is *incompressible* and the shape spheroids are

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all prolate in the direction $\tilde{\mathbf{n}}(z)$; this is to assume the asphericity $\tilde{q}(z) \geq 1$ at all points $z$ in the current configuration.

In this paper we provide a model for the total free energy of the elastomer that consists of three contributions: (1) the nematic elastic energy due to the anisotropy of the elastic solid backbone, (2) the asphericity energy that, to some extent, memorizes the initial spheroid shape of the network, and, (3) the Oseen-Frank penalty energy due to the coupling of non-uniform nematic orders and elastic deformations. This last part of the energy has been usually ignored in the study of nematic elastomers ([4, 7, 9, 11]). We stress that our main contribution here is to provide a mathematically workable and physically sound theory for the total energy of the elastomer that includes the reasonable Oseen-Frank penalty energy for the cross-linking and that, like Ericksen's theory for nematic liquid crystals (see [10, 13, 15]), our model permits the defects in the elastomer network.

We now outline the plan of this paper. In section 2, we introduce the physical quantities that describe the nematic elastomers we want to model. In particular, we introduce the *shape parameters* that combines both the director and the asphericity of the prolate spheroid. In section 3, we build up our mathematical model for the total free energy of the nematic elastomer network that includes three contributions: the nematic elastic energy, the asphericity energy, and the modified Oseen-Frank penalty energy. We remark that our theory is consistent with both the continuum mechanics theory and the molecular-statistical theory. In final two sections of the paper, sections 4 and 5, we discuss the variational properties of our total free energy functional and the existence of energy-minimizers. We omit most of the details here and refer to [5] for proofs and more discussions.

## 2. Physical State Variables and Shape Parameters.

We first introduce the physical variables that describe the nematic elastomer and then define some new variables that will be used in our theory.

### 2.1. The state variables.

The nematic elastomer modeled here can be described by the following state variables.

*(a) Elastic deformation* $z = \mathbf{y}(x)$, where $x \in \Omega$, the reference (Lagrangian) domain, $z \in D$, the current (Eulerian) domain. The incompressibility of the elastomer network requires that the deformation gradient $F = \nabla \mathbf{y}$ satisfy

$$\det \nabla \mathbf{y}(x) = 1, \quad \forall x \in \Omega. \tag{2.1}$$

*(b) Nematic director* $\tilde{\mathbf{n}} = \tilde{\mathbf{n}}(z)$ of the average axis of the crosslinked nematic mesogenic groups (rod-like segments) after deformation; this is a unit vector in $\mathbf{R}^3$, that is,

$$|\tilde{\mathbf{n}}(z)| = 1. \tag{2.2}$$

For the nematic elastomers we study here, $\tilde{\mathbf{n}}$ coincides with the principal axis of the shape spheroid of the elastic solid backbone.

*(c) Current shape* of the spheroidal elastic solid backbone of the nematic elastomer. The *asphericity* $\tilde{q} = \tilde{q}(z)$ of the spheroidal backbone is the ratio of lengths parallel and perpendicular to the principal axis (which is $\tilde{\mathbf{n}}(z)$) of the current spheroid shape. For prolate spheroids, $\tilde{q}(z) \geq 1$. The shape of the spheroid can be described by the so-called *step-length tensor* $\tilde{L}$, which is given by matrix

$$\tilde{L} = l_\perp I + (l_\parallel - l_\perp)\tilde{\mathbf{n}} \otimes \tilde{\mathbf{n}} = l_\perp(\tilde{q})[I + (\tilde{q} - 1)\tilde{\mathbf{n}} \otimes \tilde{\mathbf{n}}], \tag{2.3}$$

where $\tilde{q} = l_\parallel / l_\perp$ is the asphericity, and we have assumed the perpendicular length $l_\perp = l_\perp(\tilde{q})$ is a function of asphericity $\tilde{q}$ only. In this case the current shape is completely described by the step-length tensor; that is, $\tilde{L} = \tilde{L}(\tilde{q}, \tilde{\mathbf{n}})$.

*(d) Initial shape* of the nematic elastomer formed at the crosslinking. We assume this shape is constant and given by matrix

$$L_0 = l_0[I + (q_0 - 1)\mathbf{n}_0 \otimes \mathbf{n}_0], \tag{2.4}$$

with *constant* numbers $l_0 > 0, q_0 > 1$ and unit vector $\mathbf{n}_0$.

2.2. **The current shape parameter and material defects.** For unit director $\tilde{\mathbf{n}}$ and prolate asphericity $\tilde{q} \geq 1$ we introduce the *shape parameter* $\tilde{\mathbf{d}} = \tilde{\mathbf{d}}(z)$ by

$$\tilde{\mathbf{d}} = \sqrt{\tilde{q} - 1}\tilde{\mathbf{n}}. \tag{2.5}$$

Using this new shape parameter $\tilde{\mathbf{d}}$ we can write the step-length tensor $\tilde{L}$ above as

$$\tilde{L} = \mathbf{L}(\tilde{\mathbf{d}}) = a(|\tilde{\mathbf{d}}|)(I + \tilde{\mathbf{d}} \otimes \tilde{\mathbf{d}}), \tag{2.6}$$

where $a(s) > 0$ is a given function satisfying

$$|\ln a(s)| \leq a_0 s^r + a_1, \quad \forall\, s \geq 0 \tag{2.7}$$

for some constants $a_0, a_1$ and $1 \leq r < 6$. In [5], we have used $a(s) = (1 + s^2)^{-1/3}$ to assure the condition $\det \mathbf{L}(\tilde{\mathbf{d}}) = 1$; but we don't need this condition here.

Notice that one can always recover $\tilde{q}$ from $\tilde{\mathbf{d}}$ through $\tilde{q} = 1 + |\tilde{\mathbf{d}}|^2$. However, one can recover $\tilde{\mathbf{n}}$ from $\tilde{\mathbf{d}}$ only when $\tilde{\mathbf{d}} \neq 0$, in which case $\tilde{\mathbf{n}} = \frac{\tilde{\mathbf{d}}}{|\tilde{\mathbf{d}}|}$. Note that when $\tilde{\mathbf{d}} = 0$ and hence $\tilde{q} = 1$, the backbone shape is a *sphere* and the elastomer is in the isotropic solid phase and has no orientation preference; in this case, we consider the elastomer has a *material defect.* In this aspect, our theory is similar to that of Ericksen's on the nematic liquid crystals with variable degree of orientation (see [10, 15]). Under our theory, the *defect set* of the nematic elastomer is determined by the points of $z$ where $\tilde{\mathbf{d}}(z) = 0$.

2.3. **The reference shape parameter.** Given elastic deformation $z = \mathbf{y}(x)$ and shape parameter function $\tilde{\mathbf{d}}(z)$, define the *reference shape parameter* $\mathbf{d} = \mathbf{d}(x)$ by

$$\mathbf{d}(x) = \tilde{\mathbf{d}}(\mathbf{y}(x)). \tag{2.8}$$

If $\mathbf{y}$ is one-to-one, then this equation also uniquely defines $\tilde{\mathbf{d}}(z)$ from $\mathbf{d}(x)$. However, if $\mathbf{y}$ is not injective, one cannot always recover $\tilde{\mathbf{d}}$ from $\mathbf{d}$; but this problem can be overcome both mathematically and physically. We intend to address this issue elsewhere. (See also Remarks 3.2 and 3.3 below.)

In the following, we assume the deformation $\mathbf{y}$ is one-to-one and use $\mathbf{y}$ and the (independent) reference shape parameter $\mathbf{d}$ as the state variables for modeling the total energy of the elastomer network. One advantega of using the non-physical $\mathbf{d}(x)$ instead of the true shape parameter $\tilde{\mathbf{d}}(z)$ is that $\mathbf{d}(x)$ and $\mathbf{y}(x)$ are now both functions on the reference domain $\Omega$.

3. **The Mathematical Model for Total Free Energy.** The total free energy of the nematic elastomer consists of three contributions: the nematic elastic energy, the asphericity energy, and finally the Oseen-Frank penalty energy due to the non-uniform distribution of the nematic order. Before we build up these energies, we first introduce some notation, some of which has been already used before.

3.1. **Notation.** Let $\mathbf{M}^{3\times 3}$ denote the space of $3\times 3$ matrices with standard inner product and Euclidean norm. The components of matrix $P\in\mathbf{M}^{3\times 3}$ are written as $P_{ij}$ with $1\le i,j\le 3$. Given vectors $\mathbf{a}$, $\mathbf{b}\in\mathbf{R}^3$, $\mathbf{a}\otimes\mathbf{b}$ will denote the (rank-one) matrix in $\mathbf{M}^{3\times 3}$ with components $(\mathbf{a}\otimes\mathbf{b})_{ij}=\mathbf{a}_i\mathbf{b}_j$. For $P\in\mathbf{M}^{3\times 3}$, denote by $\det P$ the determinant of $P$ and $\mathrm{adj}P$ the cofactor matrix of $P$ that satisfies

$$P(\mathrm{adj}P)=(\mathrm{adj}P)P=(\det P)I\quad\forall\ P\in M^{3\times 3}.$$

In particular, if $P$ is invertible (i.e., $\det P\neq 0$), we have

$$(\det P)P^{-1}=\mathrm{adj}P. \tag{3.1}$$

Let $\Omega$ be a bounded domain in $\mathbf{R}^3$. For a Sobolev function $u\colon\Omega\to\mathbf{R}^3$ we denote by $\nabla u(x)$ the Jacobi matrix of $u$ as a $3\times 3$ matrix whose elements are defined almost everywhere in $x\in\Omega$ by the weak partial derivatives:

$$(\nabla u)_{ij}=\frac{\partial u^i}{\partial x_j},\quad i,j=1,2,3.$$

3.2. **The nematic elastic energy.** Following [4, 16], the ideal nematic elastic free energy is obtained from a *molecular-statistical theory* and is given by an energy density function that is a simple extension of the classical isotropic Gaussian elasticity. From this, the nematic elastic energy density is given by the so-called *trace formula*:

$$E_{el}=E_{el}(\mathbf{d},F)=\frac{\mu}{2}\left[\mathrm{tr}(L_0F^TL^{-1}F)-\ln\frac{\det L_0}{\det L}-3\right], \tag{3.2}$$

where $\mu>0$ is an elasticity constant of the network, $L_0$ and $L=\mathbf{L}(\mathbf{d})$ are the step-length tensors defined above by (2.4) and (2.6). Note that $L_0=\mathbf{L}(\mathbf{d}_0)$, where $\mathbf{d}_0=(q_0-1)^{1/2}\mathbf{n}_0\neq 0$ is a constant vector. The additive constant 3 in the formula is to assure the lowest energy is zero (see below).

Let $A_0=L_0^{1/2}$ and $B(\mathbf{d})=L^{-1/2}=\mathbf{L}(\mathbf{d})^{-1/2}$. Then we can write

$$E_{el}(\mathbf{d},F)=\frac{\mu}{2}\left[|B(\mathbf{d})FA_0|^2-2\ln\det(B(\mathbf{d})A_0)-3\right]. \tag{3.3}$$

Therefore, for any given $\mathbf{d}$, $E_{el}(\mathbf{d},F)$ is *convex* in $F$; moreover, for all $F$ with $\det F=1$, one can show that $E_{el}(\mathbf{d},F)\ge 0$, and that $E_{el}(\mathbf{d},F)=0$ if and only if $B(\mathbf{d})FA_0\in SO(3)$; that is,

$$F\in\mathcal{U}(\mathbf{d})\equiv B(\mathbf{d})^{-1}SO(3)A_0^{-1}, \tag{3.4}$$

where $SO(3)$ is the set of all rotations in $\mathbf{R}^3$ with determinant 1. $\mathcal{U}(\mathbf{d})$ is the so-called *energy-well* (see, e.g., [5, 9]).

Suppose now $\mathbf{y}(x)$ is an elastic deformation which deforms the reference domain $\Omega$ to the current domain $D$ and at each point $z\in D$ the shape parameter $\tilde{d}(z)$ of the nematic elastomer is given. Throughout the paper we assume the reference domain $\Omega$ is a bounded open domain with Lipschitz boundary $\Omega$ in $\mathbf{R}^3$. Let $\mathbf{d}(x)=\tilde{\mathbf{d}}(\mathbf{y}(x))$ be the reference parameter. Then the *nematic elastic energy* is defined by

$$\mathcal{E}_{el}=\mathcal{E}_{el}(\mathbf{d},\mathbf{y})=\int_\Omega E_{el}(\mathbf{d}(x),\nabla\mathbf{y}(x))\,dx. \tag{3.5}$$

3.3. **The asphericity energy.** We postulate that after deformation the nematic elastomer penalizes the change of asphericity except for the isotropic sphere case and remembers only the initial asphericity constant. The total *asphericity energy* is given by

$$\tilde{\mathcal{E}}_{asph} = \tilde{\mathcal{E}}_{asph}(\tilde{\mathbf{d}}) = \int_D \Phi(|\tilde{\mathbf{d}}(z)|)\, dz, \tag{3.6}$$

where $\Phi(s)$ is a continuous function satisfying, for some constants $c_0 > 0$, $c_1 > 0$ and $1 \le p < 6$,

$$\max\{0, c_0|s|^2 - 1\} \le \Phi(s) \le c_1(|s|^p + 1), \tag{3.7}$$

$$\Phi(s) = 0 \iff s = |\mathbf{d}_0| \text{ or } s = 0. \tag{3.8}$$

The growth condition (3.7) and the similar one (2.7) on function $a(s)$ above will be used in the proof of existence of energy-minimizers later. The usual Ginzburg-Landau type of *double-well* functions can be chosen as such a $\Phi$. For instance,

$$\Phi(s) = s^2(s - |\mathbf{d}_0|)^2.$$

3.4. **The modified Oseen-Frank curvature energy.** For a nematic elastomer in the current configuration $D$, we use a *modified* Oseen-Frank curvature energy to penalize the spatial change of the shape parameter $\tilde{\mathbf{d}}$ wherever $\tilde{\mathbf{d}}(z) \neq 0$.

3.4.1. *The Oseen-Frank curvature energy.* In general, in the absence of applied fields, the total *Oseen-Frank curvature energy* for unit vector field $\tilde{\mathbf{n}}(z)$ is given by the integral

$$\int_D \Psi_{\tilde{\mathbf{n}}}(z)\, dz,$$

where the energy density $\Psi_{\tilde{\mathbf{n}}}(z)$ is defined by

$$\Psi_{\tilde{\mathbf{n}}} = k_1(\mathrm{div}\tilde{\mathbf{n}})^2 + k_2(\tilde{\mathbf{n}} \cdot \mathrm{curl}\tilde{\mathbf{n}})^2 + k_3|\tilde{\mathbf{n}} \times \mathrm{curl}\tilde{\mathbf{n}}|^2 + k_4[\mathrm{tr}(\nabla\tilde{\mathbf{n}})^2 - (\mathrm{div}\tilde{\mathbf{n}})^2] \tag{3.9}$$

with the Frank constants $k_1, \cdots, k_4$. The terms with $k_1, k_2$ and $k_3$ represent the corresponding *splay, twist* and *bend* curvature energies, respectively; the $k_4$-term is a *null-Lagrangian* representing the *surface energy* contribution. (See, e.g., [10, 13, 15].)

There is a way to write $\Psi_{\tilde{\mathbf{n}}}$ as a function of $\tilde{\mathbf{n}}$ and $\nabla\tilde{\mathbf{n}}$. To do so, we define the *Oseen-Frank energy density function* $\mathcal{W}(\mathbf{n}, P)$ as follows.

$$\mathcal{W}(\mathbf{n}, P) = W_0(\mathbf{n}, P) + k_4 N(P), \tag{3.10}$$

$$W_0(\mathbf{n}, P) = k_1(\mathrm{tr}P)^2 + k_2(\mathbf{n} \cdot \mathrm{ax}(P))^2 + k_3|\mathbf{n} \times \mathrm{ax}(P)|^2, \tag{3.11}$$

$$N(P) = \mathrm{tr}(P^2) - (\mathrm{tr}P)^2, \tag{3.12}$$

where, for each matrix $P \in \mathbf{M}^{3\times 3}$, the vector $\mathrm{ax}(P) \in \mathbf{R}^3$ denotes the *axial vector* of the matrix $P$, which is uniquely defined through the identity

$$(P - P^T)\mathbf{v} = \mathrm{ax}(P) \times \mathbf{v}, \quad \forall\ \mathbf{v} \in \mathbf{R}^3. \tag{3.13}$$

For a smooth vector field $\tilde{\mathbf{m}} \colon D \to \mathbf{R}^3$, one can easily check that

$$\mathrm{tr}(\nabla\tilde{\mathbf{m}}) = \mathrm{div}\tilde{\mathbf{m}}, \quad \mathrm{ax}(\nabla\tilde{\mathbf{m}}) = \mathrm{curl}\tilde{\mathbf{m}}.$$

Hence one has

$$\Psi_{\tilde{\mathbf{n}}}(z) = \mathcal{W}(\tilde{\mathbf{n}}(z), \nabla\tilde{\mathbf{n}}(z)).$$

For unit vectors $\mathbf{n}$, $|\mathbf{n}| = 1$ we have the identity

$$|P|^2 = (\mathrm{tr}P)^2 + (\mathbf{n} \cdot \mathrm{ax}(P))^2 + |\mathbf{n} \times \mathrm{ax}(P)|^2 + N(P). \tag{3.14}$$

Therefore if $k_2 = k_3$ then $W_0(\mathbf{n}, P)$ depends only on $P$.

Throughout this paper, we assume

$$\kappa = \min\{k_1, k_2, k_3\} > 0. \tag{3.15}$$

It is easily seen that, for $|\mathbf{n}| = 1$,

$$\kappa|P|^2 \le \mathcal{W}(\mathbf{n}, P) + (\kappa - k_4)N(P). \tag{3.16}$$

If $k_1 = k_2 = k_3 = k_4 = \kappa > 0$, one obtains the so-called *one-constant* Oseen-Frank energy formula:

$$\mathcal{W}(\mathbf{n}, P) = \kappa|P|^2. \tag{3.17}$$

3.4.2. *Extending the Oseen-Frank energy function.* We extend the function $\mathcal{W}(\mathbf{n}, P)$ to arbitrary vectors $\mathbf{d}$. We use a simple extension to penalize the directional changes of $\mathbf{d}$ only; the penalty for $|\mathbf{d}|$ is already included in the asphericity energy defined above. Define

$$\mathcal{K}(\mathbf{d}, P) = \mathcal{W}(\omega(\mathbf{d}), P); \quad \omega(\mathbf{d}) = \begin{cases} \mathbf{d}/|\mathbf{d}| & \text{if } \mathbf{d} \neq 0, \\ \mathbf{n}_0 & \text{if } \mathbf{d} = 0, \end{cases} \tag{3.18}$$

where $\mathbf{n}_0 = \mathbf{d}_0/|\mathbf{d}_0|$ is the initial constant director as above. Note that the assumption $\mathcal{K}(0, P) = \mathcal{W}(\mathbf{n}_0, P)$ reflects a postulation that the elastomer may have some memory of its initial director when the current shape is isotropic.

3.4.3. *The modified Oseen-Frank energy.* We define the modified Oseen-Frank energy for shape parameter $\tilde{\mathbf{d}}(z)$ by

$$\tilde{\mathcal{E}}_{OF} = \tilde{\mathcal{E}}_{OF}(\tilde{\mathbf{d}}) = \int_D \mathcal{K}(\tilde{\mathbf{d}}(z), \nabla\tilde{\mathbf{d}}(z))\, dz \tag{3.19}$$

with the density function $\mathcal{K}(\mathbf{d}, P)$ defined above.

3.5. **The total free energy and the reference energy density function.** For the total free energy of the nematic elastomer network we simply add the three energies defined above. So the total free energy is

$$\mathcal{E}_{total} = \mathcal{E}_{el} + \tilde{\mathcal{E}}_{asph} + \tilde{\mathcal{E}}_{OF}. \tag{3.20}$$

To derive the total free energy density function in the reference configuration, we need to write

$$\mathcal{E}_{total} = \mathcal{E}_{total}(\mathbf{d}, \mathbf{y}) = \int_\Omega f_{\mathbf{d},\mathbf{y}}(x)\, dx \tag{3.21}$$

and determine the density $f_{\mathbf{d},\mathbf{y}}$ as a function of $\mathbf{d}, \mathbf{y}, \nabla\mathbf{d}, \nabla\mathbf{y}$.

As mentioned above, we assume $\mathbf{y}\colon \Omega \to D$ is a *bi-Lipschitz* map. We associate a function $\tilde{\mathbf{d}}\colon D \to \mathbf{R}^3$ to a function $\mathbf{d}\colon \Omega \to \mathbf{R}^3$ by $\mathbf{d}(x) = \tilde{\mathbf{d}}(\mathbf{y}(x))$ and, *vice versa*, a function $\mathbf{d}\colon \Omega \to \mathbf{R}^3$ to a function $\tilde{\mathbf{d}}\colon D \to \mathbf{R}^3$ through $\tilde{\mathbf{d}}(z) = \mathbf{d}(\mathbf{y}^{-1}(z))$. Note that

$$\nabla\mathbf{d}(x) = \nabla\tilde{\mathbf{d}}(\mathbf{y}(x))\nabla\mathbf{y}(x)$$

and hence

$$\nabla\tilde{\mathbf{d}}(z) = \nabla\mathbf{d}(x)(\nabla\mathbf{y}(x))^{-1}.$$

We can change all the energy integrals on current domain $D$ to the integrals on the reference configuration $\Omega$ by the (bi-Lipschitz) coordinate change $z = \mathbf{y}(x)$. Since $\det\nabla\mathbf{y}(x) = 1$, we have

$$\tilde{\mathcal{E}}_{OF} = \mathcal{E}_{OF}(\mathbf{d}, \mathbf{y}) = \int_\Omega \mathcal{K}(\mathbf{d}(x), \nabla\mathbf{d}(x)(\nabla\mathbf{y}(x))^{-1})\, dx$$

and

$$\tilde{\mathcal{E}}_{asph} = \mathcal{E}_{asph}(\mathbf{d}, \mathbf{y}) = \int_\Omega \Phi(|\mathbf{d}(x)|)\, dx.$$

Therefore, we have the total energy is given by

$$\mathcal{E}_{total} = \mathcal{E}_{total}(\mathbf{d}, \mathbf{y}) = \int_\Omega f_{\mathbf{d},\mathbf{y}}(x)\, dx = \int_\Omega \psi(\mathbf{d}, \nabla\mathbf{y}, \nabla\mathbf{d})\, dx, \tag{3.22}$$

where the density function

$$f_{\mathbf{d},\mathbf{y}}(x) = \psi(\mathbf{d}(x), \nabla\mathbf{y}(x), \nabla\mathbf{d}(x))$$

is given by *total free energy density function* $\psi = \psi(\mathbf{d}, F, G)$ defined by

$$\psi(\mathbf{d}, F, G) = E_{el}(\mathbf{d}, F) + \Phi(|\mathbf{d}|) + \mathcal{K}(\mathbf{d}, GF^{-1}), \tag{3.23}$$

with $E_{el}$, $\Phi$ and $\mathcal{K}$ defined as above. The state variables $(\mathbf{d}, F, G)$ for this free energy density function $\psi$ are

$$\mathbf{d} \in \mathbf{R}^3, \quad F,\, G \in M^{3\times 3} \quad \text{with } \det F = 1. \tag{3.24}$$

### 3.6. **A few remarks about our total energy formula.**

*Remark* 3.1. We derived the total energy formula (3.22) under the assumption that $\mathbf{y}\colon \Omega \to D$ is a *bi-Lipschitz* map. However, our final formula (3.22) or (3.23) makes no requirement that the deformation $\mathbf{y}$ be one-to-one from $\Omega$ onto $D = \mathbf{y}(\Omega)$. So we can use this total energy formula for *all* $\mathbf{d}$ and $\mathbf{y}$.

*Remark* 3.2. Mathematically, the injectivity of the deformations in nonlinear elasticity has been addressed in Ball [3] and Ciarlet & Nečas [6] and can be guaranteed either by a pure displacement (Dirichlet) boundary condition with one-to-one boundary data [3] or by imposing an inequality condition [6], which in our case $(\det \nabla\mathbf{y}(x) = 1)$ reduces simply to the condition: $|\Omega| \le |\mathbf{y}(\Omega)|$.

*Remark* 3.3. There may be still a way to recover the current shape parameter $\tilde{\mathbf{d}}(z)$ by restricting the admissible class to a subset of the *joint class* $\mathcal{C}$ of $(\mathbf{d}, \mathbf{y})$ defined by

$$\mathcal{C} = \{(\mathbf{d}, \mathbf{y}) \,|\, \mathbf{y} \in W^{1,\infty}(\Omega; \mathbf{R}^3),\ \mathbf{d}(x) = \tilde{\mathbf{d}}(\mathbf{y}(x))\ \exists \tilde{\mathbf{d}} \in W^{1,2}(\mathbf{y}(\Omega); \mathbf{R}^3)\};$$

this is equivalent to requiring $\mathbf{d}$ be constant on each level set of $\mathbf{y}$.

*Remark* 3.4. Our density function $\psi(\mathbf{d}, F, G)$ defined by (3.23), when restricted to the unit vectors $\mathbf{d} \in S^2$, satisfies all the frame-indifference and material symmetry properties of the continuum mechanics theory as required in Andersen et al [2] for their density functions (*cf.* (4.38), (4.49) and (4.52) in [2]):

$$\psi(R\mathbf{d}, RF, RG) = \psi(\mathbf{d}, F, G)\ \forall\, R \in SO(3), \tag{3.25}$$

$$\psi(\mathbf{d}, FQ, GQ) = \psi(\mathbf{d}, F, G),\ \forall\, Q \in SO(3) \text{ with } Q\mathbf{d}_0 = \mathbf{d}_0, \tag{3.26}$$

$$\psi(-\mathbf{d}, F, -G) = \psi(\mathbf{d}, F, G). \tag{3.27}$$

Therefore, our energy density function is also consistent with the continuum mechanics theory.

*Remark* 3.5. For the one-constant Oseen-Frank energy model, our modified Oseen-Frank energy density reduces to $\kappa|\nabla\mathbf{d}(\nabla\mathbf{y})^{-1}|^2$ and the model used in [2] reduces to the density $\kappa|\nabla\mathbf{y}^T\nabla\mathbf{d}|^2$. In this case, the advantage of our model is that this part of energy density is *lower semicontinuous* under a weak convergence of the

state variables $(\mathbf{d}, \mathbf{y})$ and so is compatible with the existence of energy-minimizers, while the one used in [2] is not.

*Remark* 3.6. One can assume the initial shape is an isotropic sphere; that is $A_0 = I$. To see this, let $\det A_0 = \alpha^3$, $\alpha > 0$. By a linear change of coordinate $x = A_0\hat{x}$ with $x \in \Omega$ and $\hat{x} \in \hat{\Omega}$ and the change of any function $f(x)$ to $\hat{f}(\hat{x})$ by $\alpha\hat{f}(\hat{x}) = f(x)$, we deduce

$$\mathcal{E}_{total}(\mathbf{d}, \mathbf{y}) = \hat{\mathcal{E}}_{total}(\hat{\mathbf{d}}, \hat{\mathbf{y}}) = \alpha^3 \int_{\hat{\Omega}} \hat{\psi}(\hat{\mathbf{d}}, \nabla\hat{\mathbf{y}}, \nabla\hat{\mathbf{d}})\, d\hat{x}, \tag{3.28}$$

where the new energy density function $\hat{\psi}(\hat{\mathbf{d}}, \hat{F}, \hat{G})$ is given by

$$\hat{\psi}(\hat{\mathbf{d}}, \hat{F}, \hat{G}) = \frac{\mu}{2}\left[|\hat{B}(\hat{\mathbf{d}})\hat{F}|^2 - 2\ln\det\hat{B}(\hat{\mathbf{d}}) - 3\right] + \hat{\Phi}(|\hat{\mathbf{d}}|) + \mathcal{K}(\hat{\mathbf{d}}, \hat{G}\hat{F}^{-1}) \tag{3.29}$$

with the new functions $\hat{B}, \hat{\Phi}$, and $\hat{\mathcal{K}}$ given by

$$\hat{B}(\hat{\mathbf{d}}) = \alpha B(\alpha\hat{\mathbf{d}}), \quad \hat{\Phi}(s) = \Phi(\alpha s), \quad \hat{\mathcal{K}}(\hat{\mathbf{d}}, \hat{P}) = \mathcal{K}(\hat{\mathbf{d}}, \hat{P}),$$

where $B(\mathbf{d}), \Phi(s)$ and $\mathcal{K}$ as the same functions as defined above. An interesting fact is that $\hat{\mathcal{K}}$ remains the same as $\mathcal{K}$. Note that the information on $\mathbf{d}_0$ is still encoded in the asphericity energy function $\hat{\Phi}(|\mathbf{d}|)$ and, of course, also in $\hat{\mathcal{K}}(\hat{\mathbf{d}}, \hat{P})$.

In the rest of the paper, we shall assume $A_0 = I$, so the density function $\psi(\mathbf{d}, F, G)$ is similar to $\hat{\psi}$ above.

4. **Variational Properties of the Total Energy.** By (3.1) above, the free energy density $\psi$ above can be write

$$\psi(\mathbf{d}, F, G) = E_{el}(\mathbf{d}, F) + \Phi(|\mathbf{d}|) + \mathcal{K}(\mathbf{d}, G\mathrm{adj}F). \tag{4.1}$$

For all $P \in M^{3\times3}$, one also has

$$N(P) = -2\mathrm{tr}(\mathrm{adj}P). \tag{4.2}$$

Since the map $\mathrm{ax}\colon \mathbf{M}^{3\times3} \to \mathbf{R}^3$ defined by (3.13) above is linear, and constants $k_1, k_2, k_3$ are all positive, the function $W_0(\mathbf{n}, P)$ (in the definition of $\mathcal{W}(\mathbf{n}, P)$) is convex in $P$ for any given $\mathbf{n} \in \mathbf{R}^3$. Hence, for any $P, Q \in \mathbf{M}^{3\times3}$,

$$W_0(\mathbf{n}, P + Q) \geq W_0(\mathbf{n}, P) + \frac{\partial W_0}{\partial P}(\mathbf{n}, P)\colon Q. \tag{4.3}$$

4.1. **Compensated compactness and lower semicontinuities.** The total energy $\mathcal{E}_{total}(\mathbf{d}, \mathbf{y})$ contains terms like $\nabla\mathbf{d}(x)\mathrm{adj}\nabla\mathbf{y}(x)$. Such terms have some compensated compactness property and we list the following two theorems and refer to [5] for the proof and more discussions.

**Theorem 4.1.** *Let $f_\nu \in W^{1,\infty}(\Omega; \mathbf{R}^3)$ and $g_\nu \in W^{1,2}(\Omega; \mathbf{R}^3)$ satisfy*

$$f_\nu \rightharpoonup \bar{f} \quad \textit{weakly * in } W^{1,\infty}, \quad g_\nu \rightharpoonup \bar{g} \quad \textit{weakly in } W^{1,2}.$$

*Then it follows that*

$$adj\nabla f_\nu \rightharpoonup adj\nabla\bar{f} \quad \textit{weakly * in } L^\infty(\Omega; \mathbf{M}^{3\times3}), \tag{4.4}$$

$$\nabla g_\nu\, adj\nabla f_\nu \rightharpoonup \nabla\bar{g}\, adj\nabla\bar{f} \quad \textit{weakly in } L^2(\Omega; \mathbf{M}^{3\times3}), \tag{4.5}$$

*and therefore*

$$\int_\Omega |\nabla\bar{g}\, adj\nabla\bar{f}|^2\, dx \leq \liminf_{\nu\to\infty} \int_\Omega |\nabla g_\nu\, adj\nabla f_\nu|^2\, dx.$$

**Theorem 4.2.** *Let* $\mathbf{y}_\nu \in W^{1,\infty}(\Omega;\mathbf{R}^3)$ *and* $\mathbf{d}_\nu \in W^{1,2}(\Omega;\mathbf{R}^3)$ *satisfy*

$$\mathbf{y}_\nu \rightharpoonup \bar{\mathbf{y}} \quad \textit{weakly * in } W^{1,\infty}, \quad \mathbf{d}_\nu \rightharpoonup \bar{\mathbf{d}} \quad \textit{weakly in } W^{1,2}.$$

*Let* $\Phi(|\mathbf{d}|)$, $W_0(\mathbf{n},P)$ *and* $\omega(\mathbf{d})$ *be the functions defined above. Then one has*

$$\int_\Omega \Phi(|\bar{\mathbf{d}}|)\,dx \le \liminf_{\nu\to\infty} \int_\Omega \Phi(|\mathbf{d}_\nu|)\,dx, \tag{4.6}$$

$$\int_\Omega W_0(\omega(\bar{\mathbf{d}}), \nabla\bar{\mathbf{d}}\, adj\nabla\bar{\mathbf{y}}) \le \liminf_{\nu\to\infty} \int_\Omega W_0(\omega(\mathbf{d}_\nu), \nabla\mathbf{d}_\nu\, adj\nabla\mathbf{y}_\nu). \tag{4.7}$$

4.2. **The null-Lagrangian term and coercivity of the energy.** In view of (4.2), the null-Lagragian term $N(P)$ in $\mathcal{W}(\mathbf{n},P)$ has the following property.

**Theorem 4.3.** *Let* $f_1,\ f_2 \in W^{1,\infty}(\Omega;\mathbf{R}^3)$ *and* $g_1,\ g_2 \in W^{1,2}(\Omega;\mathbf{R}^3)$ *with*

$$\det\nabla f_i(x) = 1 \quad a.e.\ x\in\Omega, \quad (i=1,2)$$

*satisfy* $f_1 = f_2$ *and* $g_1 = g_2$ *on the boundary* $\partial\Omega$ *in the sense of trace. Then*

$$\int_\Omega N(\nabla g_1\, adj\nabla f_1)\,dx = \int_\Omega N(\nabla g_2\, adj\nabla f_2)\,dx. \tag{4.8}$$

From this theorem and (3.16), we easily have the following *coercivity* result.

**Theorem 4.4.** *Assume* $\kappa = \min\{k_1,k_2,k_3\} > 0$. *Then*

$$\kappa\int_\Omega |\nabla\mathbf{d}(\nabla\mathbf{y})^{-1}|^2\,dx \le \int_\Omega \mathcal{K}(\mathbf{d},\nabla\mathbf{d}(\nabla\mathbf{y})^{-1})\,dx + C(\mathbf{d}_1,\mathbf{y}_1) \tag{4.9}$$

*for all* $\mathbf{d}\in W^{1,2}(\Omega;\mathbf{R}^3)$, $\mathbf{y}\in W^{1,\infty}(\Omega;\mathbf{R}^3)$ *with* $\det\nabla\mathbf{y}(x) = 1$ *a.e. and* $\mathbf{d}|_{\partial\Omega} = \mathbf{d}_1$, $\mathbf{y}|_{\partial\Omega} = \mathbf{y}_1$, *where* $\mathbf{d}_1 \in W^{1,2}(\Omega;\mathbf{R}^3)$ *and* $\mathbf{y}_1 \in W^{1,\infty}(\Omega;\mathbf{R}^3)$ *with* $\det\nabla\mathbf{y}_1(x) = 1$ *a.e. are given functions, and* $C(\mathbf{d}_1,\mathbf{y}_1)$ *is a constant only depending on* $\mathbf{d}_1$, $\mathbf{y}_1$.

5. **Existence of Energy Minimizers.** We study the minimization problem for the total nematic elastomer energy given above by

$$\mathcal{E}_{total}(\mathbf{d},\mathbf{y}) = \int_\Omega \psi(\mathbf{d},\nabla\mathbf{y},\nabla\mathbf{d})\,dx$$

with $\psi$ defined by (3.23) above or equivalently by (4.1).

5.1. **Admissible classes.** The natural admissible class for the reference shape parameter $\mathbf{d}$ is the Sobolev space

$$\mathcal{D} = W^{1,2}(\Omega;\mathbf{R}^3).$$

The natural class for deformation $\mathbf{y}$ is all the *volume-preserving* Lipschitz maps. However since we can not have *a priori* bounds on the Lipschitz constant or the $W^{1,\infty}$-norm of $\mathbf{y}$, we have to assume such a bound in advance. So we define the admissible class of deformations to be the volume-preserving Lipschitz maps with a given uniform Lipschitz constant $\Lambda > 0$. Let

$$\mathcal{Y} = \{\mathbf{y}\in W^{1,\infty}(\Omega;\mathbf{R}^3) \mid \det\nabla\mathbf{y}(x) = 1 \ a.e.\}, \tag{5.1}$$

$$\mathcal{Y}_\Lambda = \{\mathbf{y}\in\mathcal{Y} \mid \|\nabla\mathbf{y}\|_{L^\infty(\Omega)} \le \Lambda\}. \tag{5.2}$$

5.2. **Minimization with given Dirichlet boundary conditions.** Let $\bar{\mathbf{d}} \in \mathcal{D}$, $\bar{\mathbf{y}} \in \mathcal{Y}_\Lambda$ be given. We define the following admissible *Dirichlet classes* for shape and deformation with given boundary anchoring:

$$\mathcal{D}_{\bar{\mathbf{d}}} = \{\mathbf{d} \in \mathcal{D} \,|\, \mathbf{d}|_{\partial\Omega} = \bar{\mathbf{d}}\}, \tag{5.3}$$

$$\mathcal{Y}_{\Lambda,\bar{\mathbf{y}}} = \{\mathbf{y} \in \mathcal{Y}_\Lambda \,|\, \mathbf{y}|_\Omega = \bar{\mathbf{y}}\}. \tag{5.4}$$

For simplicity, we denote $\mathcal{A}_1 = \mathcal{D}_{\bar{\mathbf{d}}} \times \mathcal{Y}_{\Lambda,\bar{\mathbf{y}}}$. The strong and weak convergences in these Dirichlet classes will be those induced by the same convergence in the Banach space $W^{1,2} \times W^{1,\infty}$. We easily see that they are sequentially compact in the weak topology.

We prove the following existence result.

**Theorem 5.1.** *Assume $\mathcal{E}_{total}$ is defined by* (3.22) *and* (3.23). *Let $\Lambda < \infty$. Then there exists a minimizer $(\mathbf{d}^*, \mathbf{y}^*) \in \mathcal{A}_1$ such that*

$$\mathcal{E}_{total}(\mathbf{d}^*, \mathbf{y}^*) = \min_{(\mathbf{d},\mathbf{y})\in\mathcal{A}_1} \mathcal{E}_{total}(\mathbf{d}, \mathbf{y}).$$

*Proof.* The proof follows a standard direct method of the calculus of variations. Let $(\mathbf{d}_\nu, \mathbf{y}_\nu) \in \mathcal{A}_1$ be a minimizing sequence; that is,

$$\mathcal{E}_\nu = \mathcal{E}_{total}(\mathbf{d}_\nu, \mathbf{y}_\nu) \to \mathcal{E}_0 = \inf_{(\mathbf{d},\mathbf{y})\in\mathcal{A}_1} \mathcal{E}_{total}(\mathbf{d}, \mathbf{y}).$$

1. Without loss of generality, we assume $\mathbf{y}_\nu \rightharpoonup \mathbf{y}^*$ weakly * in $W^{1,\infty}(\Omega; \mathbf{R}^3)$. Then $\mathbf{y}^* \in \mathcal{Y}_{\Lambda,\bar{\mathbf{y}}}$.

2. We derive a bound on $\{\mathbf{d}_\nu\}$. By (4.9), we have $\{\nabla\mathbf{d}_\nu \mathrm{adj}\nabla\mathbf{y}_\nu\}$ is bounded in $L^2(\Omega; \mathbf{M}^{3\times3})$. Note that, for $E$, $F \in \mathbf{M}^{3\times3}$ with $\det F = 1$ and $|F| \le \Lambda$, it follows that

$$|E| \le |F||EF^{-1}| \le \Lambda|E\mathrm{adj}F|. \tag{5.5}$$

This gives a uniform $L^2$-bound on $\{\nabla\mathbf{d}_\nu\}$. Since $\mathbf{d}_\nu \in \mathcal{D}_{\bar{\mathbf{d}}}$, this implies $\{\mathbf{d}_\nu\}$ is bounded in $W^{1,2}(\Omega; \mathbf{R}^3)$. Without loss of generality we assume that $\mathbf{d}_\nu \rightharpoonup \mathbf{d}^*$ weakly in $W^{1,2}(\Omega; \mathbf{R}^3)$ and $\mathbf{d}^* \in \mathcal{D}_{\bar{\mathbf{d}}}$.

3. From (3.3) and the growth conditions (2.7) and (3.7), we see that the term $\psi_1(\mathbf{d}, F) = E_{el}(\mathbf{d}, F) + \Phi(|\mathbf{d}|)$ in the total free energy density function $\psi$ is a *non-negative convex* function in $F$ and hence, by a theorem in [1], the corresponding energy is weakly lower semicontinuous in the class $\mathcal{A}_1$. By Theorems 4.2 and 4.3, the whole energy $\mathcal{E}_{total}$ is also weakly lower semicontinuous in this class $\mathcal{A}_1$.

4. Finally, by the lower semicontinuity established above, we have

$$\mathcal{E}_{total}(\mathbf{d}^*, \mathbf{y}^*) \le \liminf_{\nu\to\infty} \mathcal{E}_{total}(\mathbf{d}_\nu, \mathbf{y}_\nu) = \mathcal{E}_0.$$

This shows that $(\mathbf{d}^*, \mathbf{y}^*) \in \mathcal{A}_1$ is a minimizer. □

5.3. **The one-constant formula of total energy.** In many situations, we need to minimize the total energy with the shape parameter fluctuating freely without anchoring to the boundary. In such cases, we do not have control on the null-Lagrangian term $N(P)$ in $\mathcal{K}(\mathbf{d}, P)$ for general choices of the Frank elasticity constants $k_i$. In what follows, we will use the one-constant formula for $\mathcal{K}(\mathbf{d}, P)$, and therefore, only consider the simplified energy functional

$$\mathcal{I}(\mathbf{d}, \mathbf{y}) = \int_\Omega \left[E_{el}(\mathbf{d}, \nabla\mathbf{y}) + \Phi(|\mathbf{d}|) + \kappa|\nabla\mathbf{d}(\nabla\mathbf{y})^{-1}|^2\right] dx \tag{5.6}$$

with $\kappa > 0$.

5.3.1. *Asphericity control.* We define, for given constants $0 \le \theta_1 \le \theta_1 \le \infty$, the class of asphericity-controlled shape parameters

$$\mathcal{D}_1 = \mathcal{D}_{\theta_1,\theta_2} = \{\mathbf{d} \in W^{1,2}(\Omega;\mathbf{R}^3) \,|\, \theta_1 \le |\mathbf{d}(x)| \le \theta_2 \ a.e.\}.$$

Note that, in this notation, $\mathcal{D} = \mathcal{D}_{0,\infty}$ and $\mathcal{D}_{1,1} = W^{1,2}(\Omega; S^2)$.

5.3.2. *Effective elastic response energy.* For given $\mathbf{y}$, we introduce an *effective elastic response energy* due to the shape fluctuation in $\mathcal{D}_1$ by

$$\mathcal{J}(\mathbf{y}) = \mathcal{J}_{\theta_1,\theta_2}(\mathbf{y}) = \inf_{\mathbf{d}\in\mathcal{D}_1} \mathcal{I}(\mathbf{d},\mathbf{y}) = \min_{\mathbf{d}\in\mathcal{D}_1} \mathcal{I}(\mathbf{d},\mathbf{y}). \tag{5.7}$$

The fact that the minimum in (5.7) is attained can be proved easily by the direct method in a similar way as in Theorem 5.1 above. Due to the property of asphericity energy density function $\Phi(|\mathbf{d}|)$ assumed before, the *energy-well* of response energy $\mathcal{J}(\mathbf{y}) = \mathcal{J}_{\theta_1,\theta_2}(\mathbf{y})$ (i.e., the set of $\mathbf{y}$ with $\mathcal{J}(\mathbf{y}) = 0$) will depend heavily on the values of $\theta_1$ and $\theta_2$ (see [5] for more).

5.3.3. *Continuities of the response energy.* We list the following properties of the effective elastic response energy defined above and refer to [5] for details and more discussions. Note that the results listed below hold for all $0 \le \theta_1 \le \theta_2 \le \infty$.

**Theorem 5.2.** *The energy $\mathcal{J}\colon \mathcal{Y} \to \mathbf{R}^+$ is continuous in the strong topology and lower semicontinuous in the weak * topology of $\mathcal{Y}$; namely,*

$$\mathcal{J}(\bar{\mathbf{y}}) \le \liminf_{\nu\to\infty} \mathcal{J}(\mathbf{y}_\nu) \quad \forall\ \mathbf{y}_\nu \rightharpoonup \bar{\mathbf{y}} \quad \textit{weakly * in } \mathcal{Y}, \tag{5.8}$$

$$\mathcal{J}(\bar{\mathbf{y}}) = \lim_{\nu\to\infty} \mathcal{J}(\mathbf{y}_\nu) \quad \forall\ \mathbf{y}_\nu \to \bar{\mathbf{y}} \quad \textit{strongly in } \mathcal{Y}. \tag{5.9}$$

*Moreover, if $\mathbf{y}_\nu \to \bar{\mathbf{y}}$ strongly in $\mathcal{Y}$ and let $\mathbf{d}_\nu \in \mathcal{D}_1$ be any minimizer of $\mathcal{J}(\mathbf{y}_\nu)$; that is, $\mathcal{J}(\mathbf{y}_\nu) = \mathcal{I}(\mathbf{d}_\nu, \mathbf{y}_\nu)$. Then $\{\mathbf{d}_\nu\}$ has a subsequence $\{\mathbf{d}_{\nu_j}\}$ strongly converging in $\mathcal{D}$ to a minimizer $\bar{\mathbf{d}}$ of $\mathcal{J}(\bar{\mathbf{y}})$.*

From this result, one easily obtains the following existence theorem.

**Corollary 5.3.** *Let $\mathcal{Y}_1$ be either the class $\mathcal{Y}_\Lambda$ or a Dirichlet class $\mathcal{Y}_{\Lambda,\bar{\mathbf{y}}}$ with $\Lambda < \infty$. Then there exists a $\mathbf{y}* \in \mathcal{Y}_1$ such that*

$$\mathcal{J}(\mathbf{y}*) = \min_{\mathbf{y}\in\mathcal{Y}_1} \mathcal{J}(\mathbf{y}) = \min_{(\mathbf{d},\mathbf{y})\in\mathcal{D}_1\times\mathcal{Y}_1} \mathcal{I}(\mathbf{d},\mathbf{y}).$$

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