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Supplementary Materials for

Controlled deformation of vesicles by flexible structured media

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A. Calculation of the best-fit surface in bead-spring model

Here we detail how we determine surface s_i. Say bead P_i has l neighbors, the position vectors of which are denoted by $\mathbf{r}_1$, $\mathbf{r}_2$, ..., $\mathbf{r}_l$. In Cartesian coordinates, $r_j = (x_j, y_j, z_j)$, j=1, 2, ..., l. We are looking for a best-fit plane s_i that is the closest to these points. The equation of s_i in Cartesian coordinates is written as

$$Ax + By + Cz = 1$$

where A, B and C are to be determined. By defining three matrices **M**, **b**, and x

$$\mathbf{M} = \begin{pmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ \vdots & \vdots & \vdots \\ x_m & y_m & z_m \end{pmatrix} \qquad \mathbf{b} = \begin{pmatrix} 1 \\ 1 \\ \vdots \\ 1 \end{pmatrix} \qquad \mathbf{x} = \begin{pmatrix} A \\ B \\ C \end{pmatrix}$$

Our goal becomes to look for a best solution x to the following linear equation

$$\mathbf{Mx} = \mathbf{b}$$

This well-known linear regression problem has the following solution

$$x = \left(M^T M\right)^{-1} M^T b = \begin{pmatrix} \sum x_j^2 & \sum x_j y_j & \sum x_j z_j \\ \sum x_j y_j & \sum x_j^2 & \sum y_j z_j \\ \sum x_j z_j & \sum y_j z_j & \sum x_j^2 \end{pmatrix}^{-1} \begin{pmatrix} \sum x_j \\ \sum y_j \\ \sum z_j \end{pmatrix}$$

The surface normal at bead P_i is then approximated by the normal to the plane s_i.

B. Analytical expressions for LC surface torque and pressure

Here we derive expressions for the surface torque, Eq. 3, and surface pressure, Eq. 4. We first demonstrate that 3 different surface torque expressions can be reduced into a single form, given that the nematic LC is uniaxial and the Euler-Lagrange equation for the surface holds. The director on the surface is denoted by **n** and the surface-preferred orientation is denoted by $\mathbf{n}^S$. The angle between **n** and $\mathbf{n}^S$ is θ. We start with Eq. 3, as given by Mackay and Denniston (*34*). Stress τ is analogous to the antisymmetric stress of the bulk LC. By introducing the surface molecular field $\mathbf{H}^W = W(\mathbf{Q} - \mathbf{Q}^S)$, one has

$$\tau = \mathbf{Q}\mathbf{H}^W - \mathbf{H}^W \mathbf{Q}$$

The above torque is not explicitly dependent on LC elastic constants. With a uniaxial assumption, the surface-preferred **Q**-field is written as $\mathbf{Q}^S = q^S (\mathbf{n}^S \mathbf{n}^S - \mathbf{I}/3)$, where q^S is the surface-preferred scalar order parameter. If we assume that the LC is uniaxial, one has $\mathbf{Q} = q(\mathbf{n}\,\mathbf{n} - \mathbf{I}/3)$, where q is the scalar order parameter. Thus

$$\tau = Wqq^S (\mathbf{n}\cdot\mathbf{n}^S)(\mathbf{n}^S\mathbf{n} - \mathbf{n}\mathbf{n}^S)$$

Substituting the above expression into Eq. 3, one has

$$\Gamma = 2Wqq^S (\mathbf{n}\cdot\mathbf{n}^S)(\mathbf{n}^S \times \mathbf{n}) = 2Wqq^S \sin\theta\cos\theta\hat{\Gamma}$$

where a unit vector $\hat{\Gamma}$ denotes the direction of Γ.

A second form of surface torque is given by de Gennes and Prost (*35*)

$$\Gamma = (\nu\cdot\Pi)\times\mathbf{n} \tag{5}$$

where ν is the surface normal and Π is related to the (bulk) distortion energy density f_d of the LC by

$$\Pi = \frac{\partial f_d}{\partial \nabla \mathbf{n}} \tag{6}$$

The above form of surface torque is not explicitly dependent on surface anchoring W. The total free energy of the LC in the director representation is (*35*)

$$F = \int_V f_d\, dV + \int_S f_S\, dS$$

where the surface free energy density f_S has an alternative form in terms of the Q-tensor

$$f_S^{(Q)} = \frac{1}{2} W (\mathbf{Q} - \mathbf{Q}^S)^2$$

We again adopt a uniaxial assumption, and the above equation reduces to

$$f_S^{(Q)} = \frac{W}{2}\left[q^2 + (q^S)^2 + \frac{5}{9}(q - q^S)^2\right] - Wqq^S(\mathbf{n}\cdot\mathbf{n}^S)^2$$

In the director model, $q = q^S = 1$. Thus the first term of f_S is a constant and can be dropped. The equivalent surface energy in director representation is

$$f_S = -Wqq^S(\mathbf{n}\cdot\mathbf{n}^S)^2 \tag{7}$$

In order to compare to other forms for the surface torque, we keep q and q^S in the above expressions. So the total free energy becomes

$$F = \int_V f_d \, dV - \int_S Wqq^S \left(\mathbf{n} \cdot \mathbf{n}^S\right)^2 dS$$

By free energy minimization, one has the Euler-Lagrange equation

$$\nu \cdot \frac{\partial f_d}{\partial \nabla \mathbf{n}} = 2Wqq^S \left(\mathbf{n} \cdot \mathbf{n}^S\right) \mathbf{n}^S$$

Plugging the above equation and Eq. 6 into Eq. 5, one has

$$\Gamma = \left(\nu \cdot \frac{\partial f_d}{\partial \nabla \mathbf{n}}\right) \times \mathbf{n} = 2Wqq^S \left(\mathbf{n} \cdot \mathbf{n}^S\right)\left(\mathbf{n} \times \mathbf{n}^S\right) = 2Wqq^S \sin\theta \cos\theta \hat{\Gamma}$$

A third way of calculating the magnitude of the surface torque Γ is via the derivative of f_S with respect to the angle θ. Eq. 7 is written in θ as

$$f_S = -Wqq^S \cos^2 \theta$$

Therefore

$$\Gamma = \frac{\delta f_S}{\delta \theta} = 2Wqq^S \sin\theta \cos\theta$$

In summary, the two forms of surface torque given by Ref. (*34*) and Ref. (*35*) are distinct. The former is not explicitly dependent on elasticity, and the latter is not explicitly dependent on surface anchoring. However, we have shown that if the LC is uniaxial, and if it obeys the Euler-Lagrange equation, the two forms are equivalent. The advantage of using Eq. 3 by Mackay and Denniston is two-fold: first, its tensorial form takes into account order parameter variations. Second, it avoids tedious calculations of elastic forces.

We next consider the contribution from symmetric stress. For neutral (zero) anchoring, the vesicle can still feel stress by the surrounding LC. The local stress is (*44*)

$$\sigma = f_B \mathbf{I} - \left(\nabla \mathbf{Q}\right) : \left(\frac{\partial f_d}{\partial \nabla \mathbf{Q}}\right)^T$$

where f_B is the bulk free energy density with $f_B = f_{Ld} + f_E$. For a third order tensor a_{ijk}, we define $a_{ijk}^T = a_{kji}$. Normal pressure on the surface is the only relevant component of the stress responsible for surface deformation. Thus we are interested in calculating p_S

$$p_S \equiv \nu \cdot \sigma \cdot \nu = f_B - \left(\nu \cdot \nabla \mathbf{Q}\right) : \left(\nu \cdot \frac{\partial f_d}{\partial \nabla \mathbf{Q}}\right)^T \tag{8}$$

Recall that the boundary condition on the surface for finite anchoring in Q-tensor form is

$$\nu \cdot \frac{\partial f_d}{\partial \nabla \mathbf{Q}} = W\left(\mathbf{Q} - \mathbf{Q}^S\right) \tag{9}$$

one has

$$p_S = f_B - \left(\nu \cdot \nabla \mathbf{Q}\right) : W\left(\mathbf{Q} - \mathbf{Q}^S\right)$$

By generalizing surface energy density f_S into a function of Q

$$f_S\left(\mathbf{Q}\right) = \frac{W}{2}\left(\mathbf{Q} - \mathbf{Q}^S\right)^2$$

one obtains

$$p_S = f_B - \nu \cdot \nabla f_S\left(\mathbf{Q}\right) \tag{10}$$

If a single-elastic-constant assumption is adopted, Eq. 9 reduces to

$$\boldsymbol{\nu} \cdot \boldsymbol{L_1} \boldsymbol{\nabla} \mathbf{Q} = \boldsymbol{W}(\mathbf{Q} - \mathbf{Q}^{\boldsymbol{S}})$$

and Eq. 8 becomes

$$\boldsymbol{p_s} = \boldsymbol{f_B} - \frac{\mathbf{2}\boldsymbol{W}}{\boldsymbol{L_1}} \boldsymbol{f_S} = \boldsymbol{f_B} - \frac{\mathbf{4}\boldsymbol{q_0}^{\mathbf{2}}}{\boldsymbol{\xi_{Kd}}} \boldsymbol{f_S} \tag{11}$$

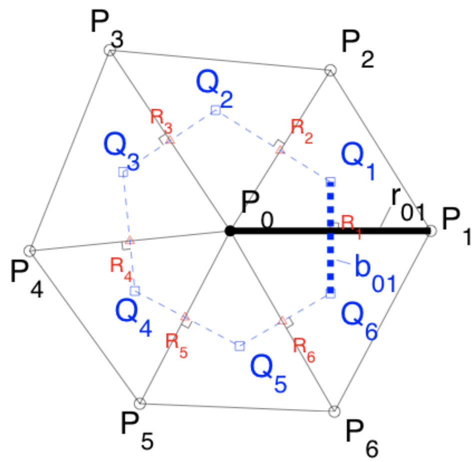

fig. S1. Illustration of the stress calculation due to surface torque. Say node P_0 has $m = 6$ neighbors, P_i (open circles), with distance $|P_0P_i| = r_{0i}$, $i = 1, 2..., 6$. Torque Γ_{0i} is defined on R_{0i} (open triangles, R_i for short), the middle point of P_0P_i. $Q_{0i,i+1}$ (open squares, Q_i for short) is the center of the circumscribed circle of triangle $P_0P_iP_{i+1}$ with $P_7 \equiv P_1$. F^{torq} is calculated on polygon $R_1Q_1R_2Q_2...R_6Q_6$ which constructs node P_0's area S_0. The bead-spring surface can be covered by these polygons. The reciprocal length of P_0P_1 is defined as $b_{01} = |Q_6R_1| + |R_1Q_1|$; the reciprocal length of P_0P_2 is $b_{02} = |Q_1R_2| + |R_2Q_3|$; ..., etc.

C. Surface torque to force calculation

Here we show how to convert the surface torque into a stress distribution. Given a surface torque Γ (units of N/m), the equivalent force distribution is

$$\boldsymbol{F^{torq}} = \boldsymbol{\nu} \cdot \boldsymbol{\nabla} \times \boldsymbol{\Gamma}$$

where v is the surface normal. $\boldsymbol{F^{torq}}$ has units of force per area. In a spherical coordinate system, the surface of interest is a sphere of radius R at the origin, and the torques are along the ϕ axis and are only dependent on θ. The resultant stress is

$$\boldsymbol{F^{torq}} = \frac{\boldsymbol{1}}{\boldsymbol{R\sin\theta}} \frac{\boldsymbol{\partial}}{\boldsymbol{\partial\theta}}(\boldsymbol{\Gamma_\phi \sin\theta})\boldsymbol{\hat{r}}.$$

In order to numerically calculate the force, we employ Stokes' theorem. Say we are calculating $\boldsymbol{F^{torq}}$ for node P_0 and it has m edges, the length of which is denoted by r_{0j}, $j = 1, 2, ..., m$. The reciprocal length for r_{0j} is b_{0j}, as illustrated in fig. S1. If the torque is defined on the middle point of edge j as Γ_{0j}, the local stress $\boldsymbol{F_0^{torq}}$ on node P_i can be estimated by

$$\boldsymbol{F_0^{torq}} \approx \frac{\boldsymbol{1}}{\boldsymbol{S_0}} \iint_{\boldsymbol{S_0}} (\boldsymbol{\nabla} \times \boldsymbol{\Gamma}) \cdot \boldsymbol{dS_0} = \frac{\boldsymbol{1}}{\boldsymbol{S_0}} \oint_{\boldsymbol{\partial S_0}} \boldsymbol{\Gamma} \cdot \boldsymbol{db} \approx \frac{\boldsymbol{1}}{\boldsymbol{S_0}} \sum_{\boldsymbol{j=1}}^{\boldsymbol{m}} \boldsymbol{\Gamma_{0j} b_{0j}}$$

where the area of S_0 of polygon $R_1Q_1R_2Q_2...R_mQ_m$ is the summation of the areas of triangle $S_{P_0R_jQ_j}$ and $S_{P_0Q_jR_{j+1}}$, $j = 1, 2 ..., m$ with $R_{m+1} \equiv R_1$. Thus

$$S_0 = \sum_{j=1}^{m} S_{P_0R_jQ_j} + S_{P_0Q_jR_{j+1}} = \frac{1}{4}\sum_{j=1}^{m} r_{0j}b_{0j}$$

Thus the surface area of the network is

$$S = \sum_i S_i = \sum_i \frac{1}{4}\sum_{(i,j)} r_{ij}b_{ij} = \frac{1}{2}\sum_{(i,j)} r_{ij}b_{ij}$$

where (i, j) sweeps all neighboring pairs.