Supplementary information for "Calculation Of Free Energies In Fluid Membranes Subject To Heterogeneous Curvature Fields" by Neeraj J. Agrawal and Ravi Radhakrishnan.

S1. FOURIER TRANSFORM OF A ONE DIMENSIONAL ANALOG OF THE HELFRICH HAMILTONIAN

For the case when $H_0 = 0$, Lin et. al. [1] have shown that the Helfrich Hamiltonian in the Monge gauge (see Eq. 2) can be written as a sum of harmonic terms involving the coefficients of a Fourier series:

$$E = \sum_{\mathbf{q}} \left[\frac{\kappa L q^4}{4} + \frac{\sigma L q^2}{4} \right] \left(a_q^2 + b_q^2 \right). \tag{S1.1}$$

Here, $q = 2\pi m/L$, m is an integer such that $0 < m \leq L/h$, and the coefficients a_q and b_q are defined by the transform (assuming a spatial domain periodic in L) [2],

$$z(x) = \frac{a_0}{2} + \sum_{q=1} a_q \cos(qx) + \sum_{q=1} b_q \sin(qx). \tag{S1.2}$$

For $H_0 = 0$, based on Eq. S1.1, the Fourier modes are indeed the independent (eigen) modes, each contributing to a Harmonic term in the total energy. We seek to find a similar Harmonic representation when $H_0 \neq 0$. To analytically represent the stiffness along undulating modes of the of Helfrich Hamiltonian (Eq. 2) within the Monge gauge when $H_0 \neq 0$, we consider a one-dimensional analogue, i.e. $z(x, y) = z(x)$. Since we employ periodic boundary conditions, $z(x)$ is periodic over length L, and hence we expand $z(x)$ in Fourier series [2] as:

$$z(x, y) = z(x) = \frac{a_0}{2} + \sum_{q=1} a_q \cos(qx) + \sum_{q=1} b_q \sin(qx) \tag{S1.3}$$

where $q = 2\pi m/L$ and m is an integer greater than zero. The upper limit on the wave-number m is dictated by the number of grid-points, L/h. We also use the orthogonality conditions that for $m \neq 0$:

$$\int_0^L \sin(2\pi mx/L)\sin(2\pi nx/L)dx = (L/2)\delta_{m,n}, \tag{S1.4}$$

$$\int_0^L \cos(2\pi mx/L)\cos(2\pi nx/L)dx = (L/2)\delta_{m,n}. \tag{S1.5}$$

$$\int_0^L \sin(2\pi mx/L)\cos(2\pi nx/L)dx = 0. \tag{S1.6}$$

The terms $\nabla^2 z$ and ∇z are given by:

$$\nabla^2 z = -\sum a_q q^2 \cos(qx) - \sum b_q q^2 \sin(qx), \tag{S1.7}$$

$$\nabla z = -\sum a_q q \sin(qx) + \sum b_q q \cos(qx). \tag{S1.8}$$

We substitute the Fourier expansion of $z(x)$ in the following Helfrich Hamiltonian:

$$E = \int_0^L \left[\frac{\kappa}{2}\left(\left(\nabla^2 z\right)^2 - 2\nabla^2 z C_0 \Gamma(x_0/2) + C_0^2 \Gamma(x_0/2)\right) + \left(\frac{\kappa C_0^2 \Gamma(x_0/2)}{4} + \frac{\sigma}{2}\right)\left(\nabla z\right)^2\right] dx. \tag{S1.9}$$

Considering term by term, we get:

$$\begin{aligned}
\int_0^L \frac{\kappa}{2}\left(\nabla^2 z\right)^2 dx &= \frac{\kappa}{2}\int_0^L \left(\sum a_q q^2 \cos(qx) + \sum b_q q^2 \sin(qx)\right)^2 dx \\
&= \frac{\kappa}{2}\int_0^L \left(\sum_q \sum_r a_q a_r q^2 r^2 \cos(qx)\cos(rx)\right) + \left(\sum_q \sum_r a_q b_r q^2 r^2 \cos(qx)\sin(rx)\right) \\
&\quad + \left(\sum_q \sum b_q b_r q^2 r^2 \sin(qx)\sin(rx)\right) dx,
\end{aligned} \tag{S1.10}$$

which using the orthogonlity conditions reduces to:

$$\int_0^L \frac{\kappa}{2}\left(\nabla^2 z\right)^2 dx = \sum_q \frac{\kappa L q^4}{4}\left(a_q^2 + b_q^2\right). \tag{S1.11}$$

The next term can be written as:

$$\begin{aligned}
-\kappa C_0 \int_0^L \Gamma(x_0/2)\nabla^2 z dx &= -\kappa C_0 \int_{L/2-x_0/2}^{L/2+x_0/2} \nabla^2 z dx \\
&= \kappa C_0 \int_{L/2-x_0/2}^{L/2+x_0/2} \left(\sum a_q q^2 \cos(qx) + \sum b_q q^2 \sin(qx)\right) dx \\
&= \kappa C_0 \left[\int_{L/2-x_0/2}^{L/2+x_0/2} \sum a_q q^2 \cos(qx)\, dx + \int_{L/2-x_0/2}^{L/2+x_0/2} \sum b_q q^2 \sin(qx)\, dx\right] \\
&= \kappa C_0 \left[\sum a_q q \sin(qx) - \sum b_q q \cos(qx)\right]_{L/2-x_0/2}^{L/2+x_0/2} \\
&= 2\kappa C_0 \left[\sum a_q q \cos(qL/2)\sin(qx_0/2) + b_q q \sin(qL/2)\cos(qx_0/2)\right] \\
&= 2\kappa C_0 \sum a_q q \cos(qL/2)\sin(qx_0/2).
\end{aligned} \tag{S1.12}$$

The next term can be written as:

$$\frac{\kappa C_0^2}{4}\int_0^L \Gamma(x_0/2)\,(\nabla z)^2\,dx = \frac{\kappa C_0^2}{4}\int_{L/2-x_0/2}^{L/2+x_0/2} (\nabla z)^2\,dx$$
$$= \frac{\kappa C_0^2}{4}\int_{L/2-x_0/2}^{L/2+x_0/2} \Big(-\sum a_q q\sin(qx) + \sum b_q q\cos(qx)\Big)^2\,dx. \tag{S1.13}$$

We denote above integral by I. To evaluate I, one needs to consider two cases: $q = r$ and $q \neq r$ separately. For $\mathbf{q} = \mathbf{r}$,

$$I = \frac{\kappa C_0^2}{8}\sum_q \big[\left(a_q^2 + b_q^2\right) q^2 x_0 + \left(-a_q^2 + b_q^2\right) q\sin(qx_0)\big]. \tag{S1.14}$$

When $\mathbf{q} \neq \mathbf{r}$, we get,

$$I = \frac{\kappa C_0^2}{2}\sum_q\sum_r \frac{qr}{q^2 - r^2}\cos(qL/2)\cos(rL/2)$$
$$\times\left\{\sin(qx_0/2)\cos(rx_0/2)\,[ra_q a_r + qb_q b_r] - \cos(qx_0/2)\sin(rx_0/2)\,[qa_q a_r + rb_q b_r]\right\}. \tag{S1.15}$$

Collectively, we can express I as:

$$I = \frac{\kappa C_0^2}{2}\sum_q\sum_r \frac{\delta_{q,r}}{4}\big[\left(a_q^2 + b_q^2\right) q^2 x_0 + \left(-a_q^2 + b_q^2\right) q\sin(x_0)\big] + (1-\delta_{q,r})\frac{qr}{q^2 - r^2}\cos(qL/2)\cos(rL/2)$$
$$\times\left\{\sin(qx_0/2)\cos(rx_0/2)\,[ra_q a_r + qb_q b_r] - \cos(qx_0/2)\sin(rx_0/2)\,[qa_q a_r + rb_q b_r]\right\}. \tag{S1.16}$$

The last term can be expressed as:

$$\frac{\sigma}{2}\int_0^L (\nabla z)^2\,dx = \frac{\sigma}{2}\int_0^L \Big(-\sum a_q q\sin(qx) + \sum b_q q\cos(qx)\Big)^2\,dx$$
$$= \frac{\sigma}{2}\int_0^L \left(\sum_q\sum_r [a_q a_r qr\sin(qx)\sin(rx) - a_q b_r qr\sin(qx)\cos(rx) + b_q b_r qr\cos(qx)\cos(rx)]\right) dx, \tag{S1.17}$$

which using orthogonality, reduces to:

$$\frac{\sigma}{2}\int_0^L (\nabla z)^2\,dx = \sum_q \frac{\sigma L q^2}{4}\left(a_q^2 + b_q^2\right). \tag{S1.18}$$

Hence, we obtain,

$$E = \frac{\kappa}{2} C_0^2 x_0 + \sum_q \sum_r \delta_{q,r} \left\{ \frac{\kappa L q^4}{4} \left(a_q^2 + b_q^2\right) + 2\kappa C_0 a_q q \cos(qL/2) \sin(qx_0/2) + \frac{\sigma L q^2}{4} \left(a_q^2 + b_q^2\right) \right.$$
$$\left. + \frac{\kappa C_0^2}{8} \left[\left(a_q^2 + b_q^2\right) q^2 x_0 + \left(-a_q^2 + b_q^2\right) q \sin(qx_0)\right] \right\} + \frac{\kappa C_0^2}{2} (1 - \delta_{q,r}) \frac{qr}{q^2 - r^2} \cos(qL/2) \cos(rL/2)$$
$$\times \left\{ \sin(qx_0/2) \cos(rx_0/2) \left[r a_q a_r + q b_q b_r\right] - \cos(qx_0/2) \sin(rx_0/2) \left[q a_q a_r + r b_q b_r\right] \right\}, \tag{S1.19}$$

where, $\delta_{q,r}$ is the Kronecker delta function. From above expression, it becomes clear that the energy contribution from a given Fourier mode is not decoupled from another, since the off-diagonal elements (obtained by setting $q \neq r$) are non-zero. This mode-mixing in Fourier coefficients implies that the Fourier modes are not exactly the eigenmodes of the system, when $H_0 \neq 0$. Moreover, when $H_0 \neq 0$, for a given mode, the energy of the sine (asymmetric about $L/2$) and the cosine (symmetric about $L/2$) modes are not equal to each other, since Eq. S1.19 is not invariant when a_q and b_q are swapped. The Helfrich Hamiltonian is nevertheless harmonic with respect to the Fourier coefficients as shown below. Differentiating E twice with respect to a_q, we obtain the stiffness (rigidity) associated with the q^{th} sine mode:

$$\frac{\partial^2 E}{\partial a_q^2} = \left[\frac{\kappa L q^4}{2} + \frac{\sigma L q^2}{2}\right] + \frac{\kappa C_0^2 q^2 x_0}{4} - \frac{\kappa C_0^2 q}{4} \sin(qx_0). \tag{S1.20}$$

In order to determine whether the effective stiffness of mode q increases or decreases with x_0, we differentiate above expression to obtain,

$$\frac{\partial}{\partial x_0} \left(\frac{\partial^2 E}{\partial a_q^2}\right) = \frac{\kappa C_0^2 q^2}{4} \left(1 - \cos(qx_0)\right). \tag{S1.21}$$

The right-hand side is always positive, which indicates that the effective stiffness of the sine mode q increases (or remains constant) with x_0. Differentiating Eq. S1.20 with respect to C_0, we obtain,

$$\frac{\partial}{\partial C_0} \left(\frac{\partial^2 E}{\partial a_q^2}\right) = \frac{\kappa C_0 q}{2} \left(q x_0 - \sin(qx_0)\right). \tag{S1.22}$$

The right-hand side is always positive which indicates that the effective stiffness of the sine mode q increases with increasing C_0. Differentiating E twice with respect to b_q, we obtain the stiffness of the q^{th} cosine mode:

$$\frac{\partial^2 E}{\partial b_q^2} = \left[\frac{\kappa L q^4}{2} + \frac{\sigma L q^2}{2}\right] + \frac{\kappa C_0^2 q^2 x_0}{4} + \frac{\kappa C_0^2 q}{4} \sin(qx_0). \tag{S1.23}$$

Indeed, the cosine (symmetric) and sine (asymmetric) modes have different effective stiffness values when $C_0 \neq 0$, however, the qualitative dependence of the stiffness with changing x_0 and C_0 remain the same.

We also derive the stiffness associated with mixed modes as:

$$\frac{\partial^2 E}{\partial a_q \partial a_r} = \frac{\kappa C_0^2}{2} \frac{qr}{q^2 - r^2} \cos(qL/2)\cos(rL/2) \Bigg\{ r \sin(qx_0/2)\cos(rx_0/2) - q\cos(qx_0/2)\sin(rx_0/2) \Bigg\}. \tag{S1.24}$$

While Eq. S1.20 defines the stiffness of harmonic potential given by Eq. S1.19 to a given cosine mode with wave-number q, Eq. S1.24 defines stiffness to a mixed modes of two cosines with wave-numbers r and q. We note that the stiffness associated with two mixed sine modes of different wave numbers can be derived using a similar procedure, however the stiffness associated with mixed sine and cosine modes (either $a_r b_q$ or $a_q b_r$) is zero.

Using Eq. S1.20, we construct a stiffness matrix, K for the cosine modes with wave-number, q ranging from $2\pi/L$ to $10\pi/L$. Let K_0 be the stiffness matrix when $C_0 = 0$ and K be the stiffness matrix when $C_0 \neq 0$. We numerically compute the eigenvalues Λ_0, Λ and the eigenmodes v_0, v of the matrices K_0 and K, respectively. The renormalized stiffness, κ_{renorm} is then defined as $\Lambda_i/\Lambda_{0,i}$ where i represent the mode-number and is plotted in Fig. 7. The eigenmode matrix for $C_0 = 0$ in Fourier space is then simply

$$v_0 = \mathbf{I}. \tag{S1.25}$$

For $C_0 \neq 0$, in Fourier space we get (where the parameters used are listed in Fig. 7):

$$v = \begin{bmatrix} 0.99994 & -0.0104 & 0.0033824 & -0.0011722 & 0.00031768 \\ 0.010452 & 0.99983 & -0.014253 & 0.0044099 & -0.0012582 \\ -0.0032468 & 0.014339 & 0.99983 & -0.010997 & 0.0029615 \\ 0.0010924 & -0.0042723 & 0.011083 & 0.99991 & -0.0068943 \\ -0.00028737 & 0.0011894 & -0.0029036 & 0.0069323 & 0.99997 \end{bmatrix}. \tag{S1.26}$$

Here, the columns of above matrices represent the eigenmodes of the system. The angle θ_{proj} is then defined as the angle between the column vector of v_0 with the corresponding column vector of v. We also obtain the eigenmodes in Cartesian space by multplying the above matrices by following transformation matrix, T.

$$T = \begin{bmatrix} cos(2\pi x/L) \\ cos(4\pi x/L) \\ cos(6\pi x/L) \\ cos(8\pi x/L) \\ cos(10\pi x/L) \end{bmatrix} \tag{S1.27}$$

We denote the eigenmode matrices in Cartesian space as v_0^c and v^c for $C_0 = 0$ and $C_0 \neq 0$, respectively. In order to quantify the degree of mode-mixing when C_0 is non-zero, we calculate the angle θ_{proj} between each eigenmode of the system and the eigenmode of a related system with $C_0 = 0$. In Fig. S1.1 we plot these angles when $C_0(x_0/2) = 0.04$ 1/nm with $x_0 = 60$ nm, which signify the degree of mixing among different Fourier modes. We note that an angle of zero represents a pure mode, i.e. Fourier modes of the system being the same as its eigenmodes. In Fig. S1.2, we plot the difference in the membrane deformation (in Cartesian representation) when the membrane fluctuates by a unit amount along the corresponding eigenmodes for the $C_0 \neq 0$ case and the $C_0 = 0$ case; these plots quantify the degree of mode mixing due to curvature field.

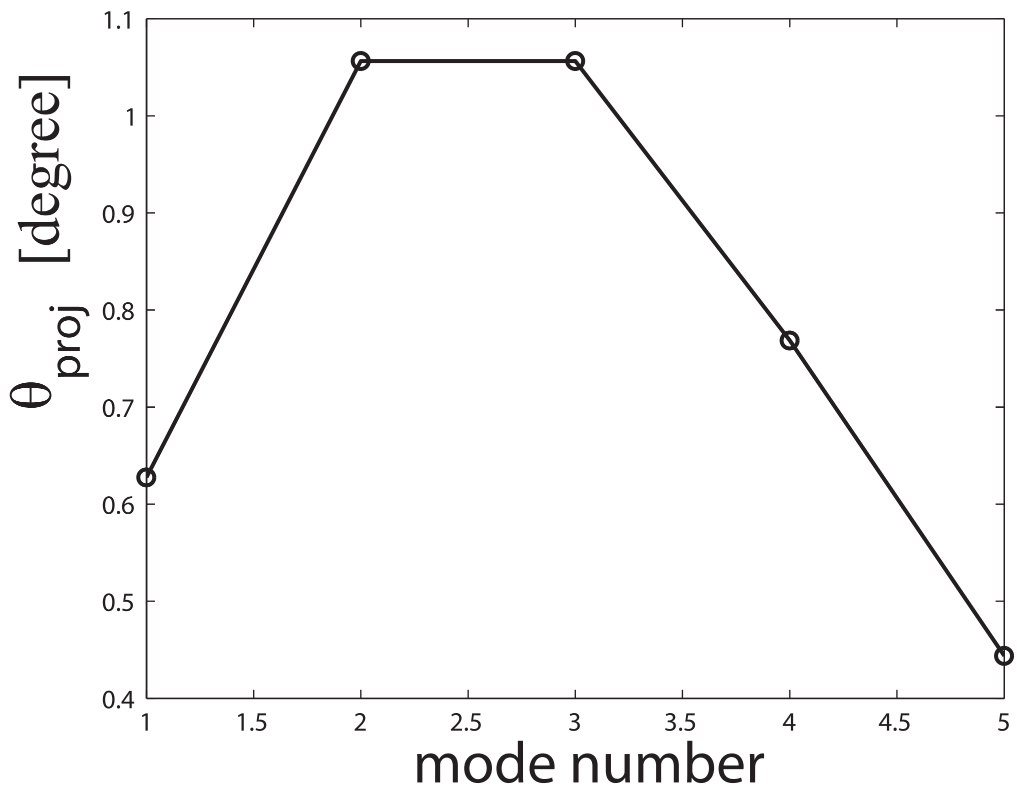

FIG. S1.1: Angle between the eigenmode of a curvature-induced membrane and the corresponding mode of a membrane under zero intrinsic curvature. In generating these plots, we have employed $L = 250$ nm, $x_0 = 60$ nm and $\sigma = 0$ N/m.

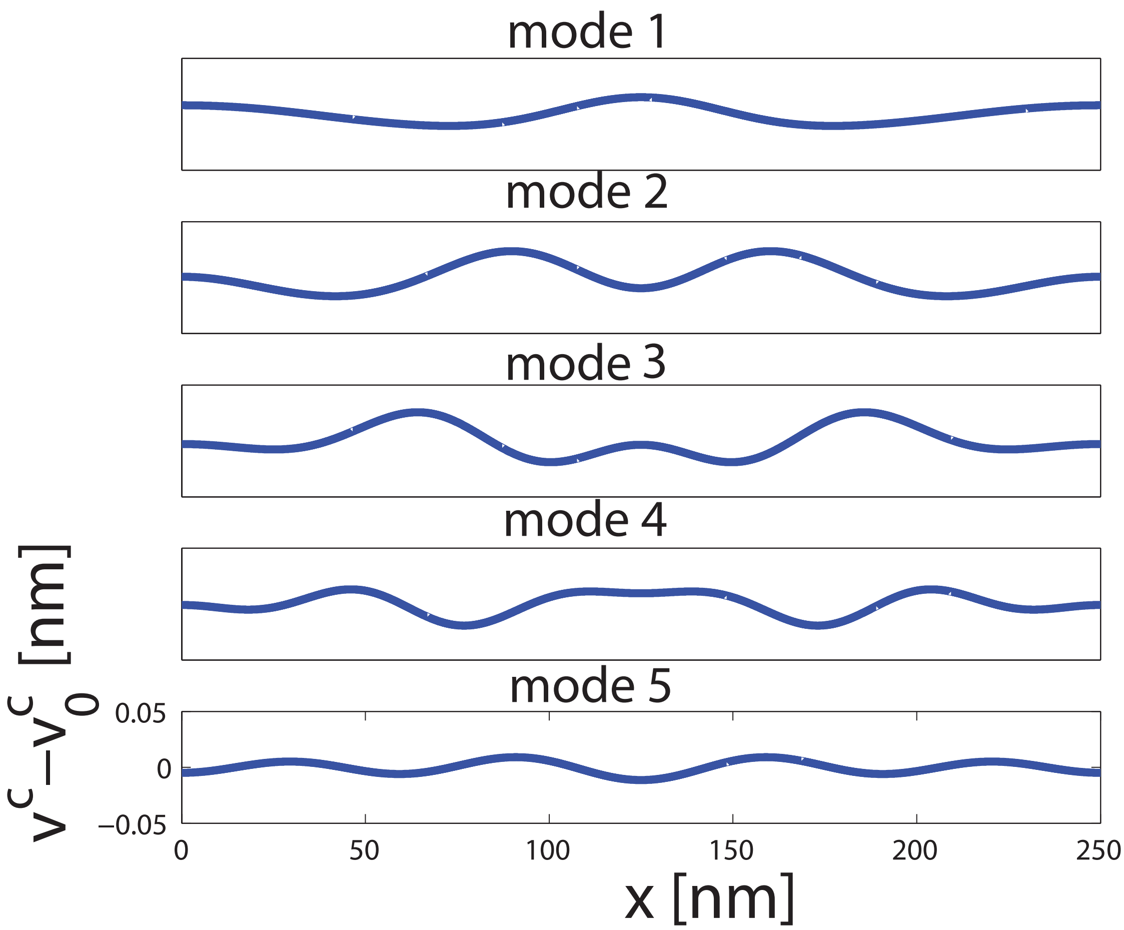

FIG. S1.2: Membrane height difference for a unit displacement along each eigenmode v_0^c and v^c.

[1] L. C. L. Lin and F. L. H. Brown, Phys. Rev. E 72, 011910 (2005).

[2] G. B. Arfken, Mathematical methods for physicists (Academic Press, Orlando, 1985).