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Comment on ‘‘Theory for the bending anisotropy of lipid membranes and tubule formation’’

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We point out that the mechanism proposed by Chen [Phys. Rev. E **59**, 6192 (1999)] for the stabilization of tubular vesicles via bending anisotropy and edge tension cannot be applied to symmetric bilayers. Other possible mechanisms are reviewed, with special emphasis on one involving a nematic order of the surfactant polar heads.

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In a recent paper [1], Chen has discussed a mechanism for the stabilization of tubular vesicles, based on the anisotropy of the bending rigidity due to the tilt of the lipid molecules in the $L_{\beta'}$ phase and on the presence of a spontaneous curvature. This model is applied to symmetric bilayers, on the basis of an argument according to which the removal of the edge energy of a flat bilayer patch, obtained by forming a closed vesicle, can be represented by the introduction of a spontaneous curvature. Explicitly, the integral

$$\mathcal{C}=\tau \oint_{\Sigma} d\sigma(c_1+c_2), \tag{1}$$

where Σ represents the vesicle surface, whose local principal curvatures are c_1 and c_2 [2], would give the edge energy of the corresponding flat membrane with line tension τ.

This equivalence does not hold since the integral $\mathcal{C}$ is not independent of the shape assumed by the vesicle. For example, it is easy to establish that

$$\mathcal{C}=\tau\frac{\alpha+4}{\sqrt{\alpha+2}}\sqrt{2\pi S} \tag{2}$$

for a tubular vesicle with area S, closed by two spherical caps, where α is the ratio of the length of the cylindrical part to its radius. The sphere corresponds to the limit case $\alpha=0$.

The intrinsic length $\ell=\kappa/\tau$, where κ is the bending rigidity, does not define a spontaneous curvature radius, but sets instead the *minimal* size of the vesicles (neglecting entropic effects). When the bilayer patch is smaller than $\approx\ell$, it would rather remain flat with an open boundary than close up to form a vesicle. Nothing prevents, however, the formation of large, closed vesicles with an arbitrarily small curvature [3]. On the other hand, if a spontaneous curvature term were intrinsically associated to the edge tension, experiments would show quite monodisperse distributions of vesicle sizes, specific to the molecular nature of the bilayer, which is clearly not the case.

In a closed vesicle, the anisotropy of the bending rigidity does not allow one to define a characteristic length in the absence of a spontaneous curvature. Therefore it cannot explain by itself the stability of tubules with a well-defined radius independent of their area.

While the mechanism proposed by Chen cannot explain the stability of tubules in symmetric bilayers, it can be relevant for asymmetric ones. Tubules can be stabilized in *symmetric* bilayers with in-plane anisotropy if the anisotropy directions in the two monolayers are unlocked [4,5]. This can take place, e.g., if a difference is allowed in the tilt direction between the two monolayers of a $L_{\beta'}$ membrane [4] or in the presence of in-plane nematic or paranematic order of the surfactant polar heads [5,6], when the corresponding monolayer directors are unlocked.

For definiteness, we reproduce here the reasoning of Ref. [6]. Consider a curved monolayer made of surfactants that have elongated polar heads possessing an in-plane nematic order $\mathsf{Q}=S(\mathbf{nn}-\frac{1}{2}\mathsf{I})$. We assume that the nematic director $\mathbf{n}$ lies along one of the principal directions of curvature in the tangent plane. The curvature tensor takes then the form $\mathsf{K}=c_\parallel\mathbf{nn}+c_\perp\mathbf{n}_\perp\mathbf{n}_\perp$, with $\mathbf{n}_\perp$ a unit vector orthogonal to $\mathbf{n}$. The most general form of the monolayer's curvature energy compatible with the symmetry is then

$$f(c_\parallel,c_\perp)=\frac{\kappa}{4}(c_\parallel+c_\perp)^2+\frac{\bar{\kappa}}{2}c_\parallel c_\perp+\alpha(c_\parallel+c_\perp)+\frac{1}{2}\beta S(c_\parallel-c_\perp). \tag{3}$$

For the sake of simplicity we have neglected the anisotropy of κ and $\bar{\kappa}$.

We now consider a curved *bilayer* made of two such monolayers. We assume first that the directors of the two monolayers are orthogonal to each other. The free energy F of the bilayer is obtained by adding $f(c_\parallel,c_\perp)$ and $f(-c_\perp,-c_\parallel)$, and is given by

$$F=\frac{\kappa}{2}(c_\parallel+c_\perp)^2+\bar{\kappa}c_\parallel c_\perp+\beta S(c_\parallel-c_\perp). \tag{4}$$

An obvious uniform solution minimizing Eq. (4) is a tube. Setting $c_\parallel=0$ and $c_\perp=c$, we obtain

$$F_{\text{tube}}=\frac{\kappa}{2}c^2-\beta S c, \tag{5}$$

the minimum of which is obtained for

$$c_{\text{tube}}=\frac{\beta S}{\kappa}. \tag{6}$$

It is easy to check that this configuration yields the minimum free energy also in the presence of a (not too large) interaction tending to align the nematic order parameters of the two monolayers.

In order to estimate the parameter β we introduce the quantities $c_{\parallel}^{0}$ and $c_{\perp}^{0}$, representing the two spontaneous principal curvatures of the *monolayer* for a perfect ordering of the polar heads. By minimizing Eq. (3) with $S=1$ one easily obtains

$$c_{\parallel}^{0}+c_{\perp}^{0}=-\frac{4\alpha}{2\kappa+\bar{\kappa}}, \tag{7}$$

$$c_{\parallel}^{0}-c_{\perp}^{0}=\frac{2\beta}{\bar{\kappa}}. \tag{8}$$

One obtains therefore $\beta=\frac{1}{2}\bar{\kappa}(c_{\parallel}^{0}-c_{\perp}^{0})$. Assuming $\bar{\kappa}\simeq 2\kappa$, the estimates $c_{\parallel}^{0}\simeq(100\ \text{Å})^{-1}$, $c_{\perp}^{0}\simeq(120\ \text{Å})^{-1}$, and $S\simeq 0.3$, for dimeric surfactants with long chains, yield $1/c_{\text{tube}}\simeq 2000\ \text{Å}$, in agreement with the optically visible tubules reported in Ref. [7].

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