Selection of toroidal shape of partially polymerized membranes

Ou-Yang Zhong-can
Institute of Theoretical Physics, P.O. Box 2735, Beijing 100080, China
(Received 13 July 1992; revised manuscript received 23 September 1992)

The theroretical prediction of the existence of Clifford toroidal vesicles by Ou-Yang [Phys. Rev. A **41**, 4517 (1990)] has been confirmed by the experiment of Mutz and Bensimon [Phys. Rev. A **43**, 4525 (1991)]. However, the few nonaxisymmetric toroidal vesicles observed by Fourcade, Mutz, and Bensimon [Phys. Rev. Lett. **68**, 2551 (1992)] raise the question of why the formation of nonaxisymmetric toroidal vesicles is so rare in comparison with that of axisymmetric ones. In this report we are going to show that for a solution of the shape equation of membranes, the spontaneous curvature c_0 of nonaxisymmetric Dupin cyclide has to be zero, while axisymmetric Clifford solution has no such restriction as shown by Ou-Yang [Phys. Rev. A **41**, 4517 (1990)].

PACS number(s): 82.70.−y

Vesicles with toroidal topology are predicted to be stable for the Helfrich free-energy model [1], and have been observed in partially polymerized phospholipid membranes by Mutz and Bensimon (MB) [2]. Furthermore, calculations and experiments agree on the specific toroidal shape , the so-called Clifford torus, an axisymmetric torus such that the ratio of its generating circles is $\sqrt{2}$. Recently, in an interesting Letter [3], Fourcade, Mutz, and Bensimon (FMB) report a richer observation of toroidal vesicles of diacetylenic phospolipids. Together with the earlier findings by MB [2] the observation supports recent theoretical predictions [1,4,5]. However, their observation is not completely understood. For example, Seifert [4] has questioned the quantitative analysis in terms of the model for fluid membranes discussed by MB [2] on two points: (1) One has to suppose that the essential effect of the polymerization results in an effective spontaneous curvature c_0. (2) If c_0 is large and negative, the shape of lowest bending energy is no longer the Clifford torus but rather a nonaxisymmetric shape. If the corresponding spontaneous curvature is small and distributed about zero, one would expect that apart from the Clifford torus also nonaxisymmetric shapes occur. In other words, MB's finding [2] seems to have happened only in rare cases. From the experimental side, contrarily, MB [2] and FMB [3] only observed a specific family of shapes, the Clifford torus, and, *less often*, the branch of nonaxisymmetric Dupin cyclides generated by its conformal transformations. To explain this problem, FMB has proposed a plausible explanation by assuming that the partially polymerized vesicles are permeable on short time scale, so in the early stage of their formation, they can settle at the absolute minimum of the bending energy, i.e., the Clifford torus or its conformal transformation. But, the question of [4] is still not completely settled, especially, on the selection between axisymmetric and nonaxisymmetric toroids. In this report we are going to show that the selection biased towards Clifford torus observed by MB and FMB is consistent with an exact solution of the shape equation of vesicles.

Indeed, if we suppose the spontaneous curvature c_0 to be zero the above selection problem arises as commented in [4]. It is, due to Willmore [6], long known that when $c_0 = 0$ the Clifford torus and its inversion with respect to any point possess the same shape energy. This degeneracy should thus lead to the remarkable result: the nonaxisymmetric Dupin cyclides will be more often observed than the Clifford one, that is, just contradictory to observation. In order to solve the dilemma we look into the difference of the properties for the two types of torus. If the essential effect of the polymerization should be an introduction of an effective nonvanishing c_0 then the dilemma would be solved. In [1] it has been shown that the Clifford torus is an equilibrium shape irrespective of whether c_0 is equal to zero or not. Now the question is what about the nonaxisymmetric torus?

It is noteworthy to look into the similarity between membranes of amphiphilic molecular bilayer and molecular layers of smectic liquid crystals in the *Sm-A* phase. First of all, both kinds of molecular layers are of constant thickness [7,8]. Secondly, in the molecular bilayer, the long amphiphilic molecules stay perpendicular to the membrane surface, and the director of the smectic liquid crystal is also perpendicular to the smectic molecular layer [7,8]. These geometrical similarities between amphiphilic membranes and smectic liquid crystals lead us to believe that the shape of toroidal vesicles is closely related to the texture of smectic liquid crystals. Mathematically, the focal conic texture of smectic liquid crystals [9] comes from the formation of a series of Dupin cyclides by the smectic molecular layers. Thus we would expect that Dupin cyclide should be a solution of the equilibrium shape equation of membranes [10]:

$$\Delta p - 2\lambda H + k(2H + c_0)(2H^2 - 2K - c_0 H) + 2k\nabla^2 H = 0\,, \tag{1}$$

where the Lagrange multipliers Δp and λ take account of the constraints of constant volume and area, respectively, k is the elastic constant, H is the mean curvature, K is the Gaussian curvature, and ∇^2 denotes the Bertrami-Laplace differential operator.

Using usual Cartesian coordinates, Forsyth [11] gives two equivalent implicit equations for a nonaxisymmetric Dupin cyclide shown in Fig. 1,

$$(x^2+y^2+z^2-\mu^2+b^2)^2=4(ax-c\mu)^2+4b^2y^2\ , \tag{2}$$

$$(x^2+y^2+z^2-\mu^2-b^2)^2=4(cx-a\mu)^2-4b^2z^2\ , \tag{3}$$

where the parametric constants a, b, c, and μ are imposed by the constraints of $a^2=b^2+c^2$ and $0<c<\mu<a$. The parametric form of curvature coordinates (p. 326 of [11]) and the first and second fundamental forms (p. 327) are also involved in the early investigation. The parametric form is

$$\begin{aligned} x&=[\mu(c-a\cos\theta\cos\psi)+b^2\cos\theta]/(a-c\cos\theta\cos\psi)\ ,\\ y&=b\sin\theta(a-\mu\cos\psi)/(a-c\cos\theta\cos\psi)\ ,\\ z&=b\sin\psi(c\cos\theta-\mu)/(a-c\cos\theta\cos\psi)\ , \end{aligned} \tag{4}$$

where θ and ψ are two parameters of curvature coordinates. The constants a, b, c relate two conics involved in a geometric definition of the surface, namely the ellipse

$$(x/a)^2+(y/b)^2=1\ ,\quad z=0\ , \tag{5}$$

and the hyperbola

$$(x/c)^2-(z/b)^2=1\ ,\quad y=0\ , \tag{6}$$

lying in perpendicular planes. Each conic is required to pass through the foci of other, which imposes the additional constraint

$$a^2=b^2+c^2\ . \tag{7}$$

The cyclide then can be seen as an envelope of a sphere of variable radius moving so that its center lies on one of the conics while the other conic is intersected in a fixed point. The additional constant μ is determined by the choice of intersection point and, therefore, closely relates the radius of the moving sphere to the position of its center.

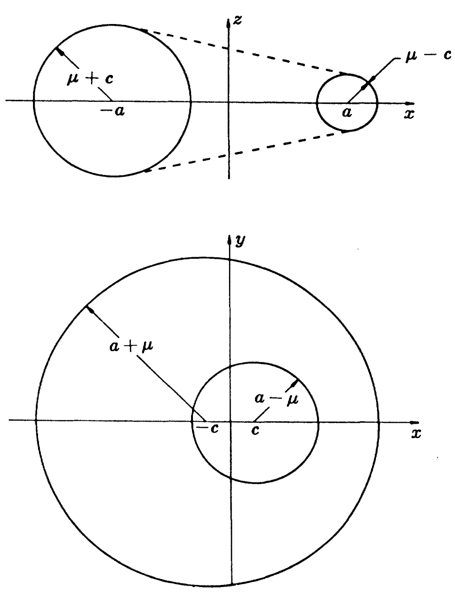

FIG. 1. Cross section of a Dupin cyclide in its planes of symmetry.

One of the most important properties of the Dupin cyclide is that all its lines of curvature are plane circles so that their parametric variables θ, ψ range from 0 to 2π. Using the parametric representation the coefficients of the first and the second fundamental forms given by Forsyth are

$$\begin{aligned} g_{11}&=b^2(\mu\cos\psi-a)^2/(a-c\cos\theta\cos\psi)^2\ ,\\ g_{22}&=b^2(\mu-c\cos\theta)^2/(a-c\cos\theta\cos\psi)^2\ ,\\ g_{12}&=0\ , \end{aligned} \tag{8}$$

and

$$\begin{aligned} L_{11}&=b^2\cos\psi(\mu\cos\psi-a)/(a-c\cos\theta\cos\psi)^2\ ,\\ L_{22}&=b^2(\mu-c\cos\theta)/(a-c\cos\theta\cos\psi)^2\ ,\\ L_{12}&=0\ , \end{aligned} \tag{9}$$

respectively. With Eqs. (8) and (9) we find that the Gaussian curvature K and the mean curvature H are given by

$$K=L_{11}L_{22}/g_{11}g_{22}=\cos\psi/(\mu-c\cos\theta)(\mu\cos\psi-a)\ , \tag{10}$$

and

$$H=\frac{1}{2}\left(\frac{L_{11}}{g_{11}}+\frac{L_{22}}{g_{22}}\right)=\frac{1}{2}\left(\frac{\cos\psi}{\mu\cos\psi-a}+\frac{1}{\mu-c\cos\theta}\right)\ , \tag{11}$$

respectively.

Given these we now can calculate all the terms given in the left side of Eq. (1). Before getting into the actual calculation we first simplfy the above tool formulas and equations by setting $a=1$ and $k=1$. This change in scale and in unit facilitates the mathematical expression but does not cause any loss of physical generality. Using the convenient contracted notations of $A=\cos\theta$ and $B=\cos\psi$ and letting $g=g_{11}g_{22}$ we first have

$$\begin{aligned} 2\nabla^2H&=\frac{1}{\sqrt{g}}[\partial_\theta(\sqrt{g}/g_{11})\partial_\theta+\partial_\psi(\sqrt{g}/g_{22})\partial_\psi]2H\\ &=[c^3(\mu^2-1)A^3B^3+b^2c^2\mu A^2B^3\\ &\quad+c^2(c^2-3\mu^2+2)A^2B^2-2b^2c\mu AB^2\\ &\quad+c(3\mu^2-2c^2-1)AB+b^2\mu B\\ &\quad+(c^2-\mu^2)]/[b^2(1-\mu B)^3(\mu-cA)^3]\ . \end{aligned} \tag{12}$$

Defining the new parameter $\gamma=\lambda+c_0^2/2$ and taking some algebra we obtain the sum of the other terms in the shape equation (1)

$$\Delta p-2\lambda H+(2H+c_0)(2H^2-2K-c_0H)=[(\Delta p\mu^3-\gamma\mu^2+\tfrac{1}{2})c^3A^3B^3+(2c_0\mu^2+4\gamma\mu^3-3\Delta p\mu^4-\mu)c^2A^2B^3$$
$$+(2\gamma\mu-3\Delta p\mu^2)c^3A^3B^2+(9\Delta p\mu^3-9\gamma\mu^2-4c_0\mu-\tfrac{1}{2})c^2A^2B^2$$
$$+(3\Delta p\mu-\gamma)c^3A^3B+(3\Delta p\mu^5-5\gamma\mu^4-4c_0\mu^3)cAB^3$$
$$\times(2c_0+6\gamma\mu-9\Delta p\mu^2)c^2A^2B+(8c_0\mu+12\gamma\mu^2-9\Delta p\mu^3+2)c\mu AB^2$$
$$-\Delta pc^3A^3+(2c_0\mu^4+2\gamma\mu^5-\Delta p\mu^6)B^3+(3\Delta p\mu-\gamma)c^2A^2$$
$$+(9\Delta p\mu^3-9\gamma\mu^2-4c_0\mu-\tfrac{1}{2})cAB+(3\Delta p\mu^5-5\gamma\mu^4-4c_0\mu^3)B^2$$
$$+(2\gamma\mu-3\Delta p\mu^2)cA+(2\mu c_0+4\gamma\mu^2-3\Delta p\mu^3-1)\mu B$$
$$+(\Delta p\mu^3-\gamma\mu^2+\tfrac{1}{2})]/[(1-\mu B)^3(\mu-cA)^3]\ . \qquad (13)$$

Substituting (12) and (13) into shape equation (1) and then letting each coefficient of the term A^mB^n, where $m,n=0,1,2,3$, equal zero we find 16 equations for the parametric constants b, c, μ, Δp, γ, and c_0. The solutions of these 16 equations fall into two categories. The first case is for $c=0$. In this case we have to require $a=b$ and $\mu=1/\sqrt{2}$ but no restriction on the value of c_0. From Fig. 1 one can find that this is the case of the axisymmetric Clifford torus discussed in [1]. The other case is for $c\neq 0$, which describes nonaxisymmetric Dupin cyclides (see Fig. 1). In this case we find harsh constraints given by

$$\Delta p=\gamma=c_0=0\ , \qquad (14)$$

and

$$\mu^2=\tfrac{1}{2}(1+c^2)\ . \qquad (15)$$

The constraint (14) is easy to see from the coefficients of A^3, A^2, A^2B, and so on in (13). The constraint (15) together with (14) can just cause all the coefficients of A^nB^m to vanish in the sum of (12) and (13). In other words, (14) and (15) are the *sufficient* and *necessary* conditions for the nonaxisymmetric Dupin cyclide as a vesicle solution.

If we restore a to its original scaling, i.e., not restrict $a=1$, (15) should read as

$$\mu^2=\tfrac{1}{2}(a^2+c^2)\ . \qquad (16)$$

From this geometric constraint we see that among the nonaxisymmetric Dupin cyclides only that branch which is generated by the conformal transformations of Clifford torus and satisfies the condition (14) may be observed, though it might be that for nonzero c_0 there exist nonaxisymmetric toroidal solutions close to these Dupin cyclides. On the other hand, as shown in [1], the Clifford torus may be observed for $c_0 \lesssim -3.9(4\pi/A)^{1/2}$, where A is the total area of the vesicle, and it is sufficiently stable in equilibrium. Seifert [4] and Fourcade [12] argued that it should be stable as soon as c_0 is negative. It is now clear that owing to the restriction (14) the nonaxisymmetric Dupin cyclide happens only in rare cases, while the Clifford torus can appear in many cases. Furthermore, direct measurement on the real nonaxisymmetric unpolymerized fluid toroidal vesicle given in Fig. 1 of [3] confirms the validity of (16).

In conclusion, the observation on the selection of toroidal shape of partially polymerized membranes by FMB can be explained in terms of the theoretical framework of the fluid membranes modeled by Helfrich and others. This selection in shape is also consistent with a rich history of surface problems in geometry, e.g., the famous plateau problems (soap bubble problem) characterized by H=const. For this there is only a unique solution of vesicle shape, the sphere.

The author wishes to thank Professor Yu-Chang Hsieh for his help with the manuscript. Thanks are also due FMB and U. Seift for sending copies of work prior to publication. This work was supported partially by the National Natural Science Foundation of China and Grant No. LWTZ-1298 of the Chinese Academy of Science.

[1] Ou-Yang Zhong-can, Phys. Rev. A **41**, 4517 (1990).
[2] M. Mutz and D. Bensimon, Phys. Rev. A **43**, 4525 (1991).
[3] B. Fourcade, M. Mutz, and D. Bensimon, Phys. Rev. Lett. **68**, 2551 (1992).
[4] U. Seifert, Phys. Rev. Lett. **66**, 2404 (1990).
[5] W. B. Duplantier, Physica A **168**, 179 (1990).
[6] T. J. Willmore, *Total Curvature in Riemanian Geometry* (Horwood, Chicester, 1980).
[7] Y. Boligand, J. Phys. (France) **33**, 525 (1972).
[8] W. Helfrich, Z. Naturforsch. **28c**, 693 (1973).
[9] W. Bragg, Nature (London) **133**, 445 (1934).
[10] Ou-Yang Zhong-can and W. Helfrich, Phys. Rev. Lett. **59**, 2486 (1987).
[11] A. R. Forsyth, *Lectures on Differential Geometry of Curves and Surface* (Cambridge University Press, Cambridge, 1912).
[12] B. Fourcade (unpublished).