New Solutions to the Helfrich Variation Problem for the Shapes of Lipid Bilayer Vesicles: Beyond Delaunay's Surfaces

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Surfaces of revolution with constant mean curvature, i.e., catenoids, unduloids, nodoids, circular cylinders, and spheres, are called Delaunay's surfaces. All these surfaces are found to be solutions of the Helfrich variation problem which is the determination of the equilibrium shapes of lipid bilayer vesicles. It is shown that two kinds of surfaces of revolution *not* with constant mean curvature are also rigorous solutions of the same Helfrich variation problem. The solutions are similar to unduloids and nodoids, and degenerate to spheres, circular cylinders, and tori in certain limiting cases.

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Minimal surfaces and surfaces with constant mean curvatures have become famous and challenging problems in differential geometry since the study of soap bubbles by Plateau [1] and of the shape of air/liquid interfaces in a capillary tube by Young [2] and Laplace [3] in the middle of the nineteenth century. Even nowadays, there are still interesting problems in this field. For instance, many new problems of surfaces arise from the shapes of vesicles that are formed by lipid bilayers in aqueous solution, which are simple models for biological membranes and cells. The theoretical approach for the determination of the equilibrium shapes is based on the elasticity of lipid bilayers proposed by Helfrich [4], and is called the Helfrich variation problem. In this approach, the shape free energy is given by

$$F = \frac{1}{2} k \oint (c_1 + c_2 - c_0)^2 dA + \bar{k} \oint c_1 c_2 \, dA + \Delta p \int dV + \lambda \oint dA , \quad (1)$$

where k is the bending rigidity, $\bar{k}$ the Gaussian curvature modulus, c_1 and c_2 the two principal curvatures, c_0 the spontaneous curvature, dA the surface area, dV the volume element, $\Delta p = p_o - p_i$ the osmotic pressure difference between outer (p_o) and inner media (p_i), and λ the tensile stress. Δp and λ are also regarded as the Lagrange multipliers which take account of the constraints of constant volume and area, depending on the situation.

The expression for the shape of the vesicle at mechanical equilibrium has been derived from the first variation of F with respect to the normal to the vesicle surface using general rules of differential geometry and imposing the closed surface condition [5]. The expression is called the general shape equation and is given by

$$\Delta p - 2\lambda H + k(2H + c_0)(2H^2 - 2K - c_0 H) + 2k\nabla^2 H = 0 , \quad (2)$$

where H $[=-(c_1 + c_2)/2]$ and K $(=c_1 c_2)$ are the mean and Gaussian curvatures, respectively, and ∇^2 is the Laplace-Beltrami operator. Most studies of the Helfrich variation problem have so far been based on the numerical calculation of the shape equation for vesicles with special geometries such as axisymmetry [67]. However, general shapes have not been analytically examined yet, except for a sphere, a cylinder, and a Clifford torus and its conformal transformations [8]. In the case of $\Delta p/\lambda = -c_0$, Eq. (2) is satisfied when the surface has constant mean curvature $H = -c_0/2$; the surface is a minimal surface if $c_0 = 0$. In 1841 Delaunay [9] showed that the surfaces of revolution with constant mean curvature in Euclidean space are catenoids ($H = 0$), unduloids, nodoids, circular cylinders, and spheres. These surfaces are called Delaunay's surfaces and are the solution of Eq. (2), as shown below.

In this Letter, we report new solutions of the Helfrich variation problem [Eq. (2)], which are similar to Delaunay's surfaces but *not* with constant mean curvatures. Two kinds of surfaces of revolution similar to unduloids and nodoids are found as new rigorous solutions of Eq. (2). The solutions become spheres, cylinders, and tori in certain limiting cases.

For a vesicle with axisymmetry (i.e., a vesicle surface which is a surface of revolution), Eq. (2) is the nonlinear fourth-order ordinary differential equation for contours. If we introduce the angle ψ between the surface tangent and the plane perpendicular to the axisymmetric axis (z axis), the differential equation can be reduced to the third-order differential equation of $\psi(\rho)$ [10], where ρ is the distance from the z axis. The contour $z(\rho)$ can be obtained from a simple integration

$$z(\rho) - z(\rho') = \int_{\rho'}^{\rho} \tan\psi(\rho'') d\rho'' . \quad (3)$$

The shape equation for a vesicle with axisymmetry can be obtained by simply substituting the mean and the Gaussian curvatures of an axisymmetric surface into the general shape equation [Eq. (2)] [10], and is

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$$
\begin{aligned}
\cos^3\psi\left(\frac{d^3\psi}{d\rho^3}\right) = {} & 4\sin\psi\cos^2\psi\left(\frac{d^2\psi}{d\rho^2}\right)\left(\frac{d\psi}{d\rho}\right) - \cos\psi\left(\sin^2\psi - \frac{1}{2}\cos^2\psi\right)\left(\frac{d\psi}{d\rho}\right)^3 \\
& + \frac{7\sin\psi\cos^2\psi}{2\rho}\left(\frac{d\psi}{d\rho}\right)^2 - \frac{2\cos^3\psi}{\rho}\left(\frac{d^2\psi}{d\rho^2}\right) \\
& + \left[\frac{c_0^2}{2} - \frac{2c_0\sin\psi}{\rho} + \frac{\lambda}{k} - \frac{\sin^2\psi - 2\cos^2\psi}{2\rho^2}\right]\cos\psi\left(\frac{d\psi}{d\rho}\right) \\
& + \left[\frac{\Delta p}{k} + \frac{\lambda\sin\psi}{k\rho} + \frac{c_0^2\sin\psi}{2\rho} - \frac{\sin^3\psi + 2\sin\psi\cos^2\psi}{2\rho^3}\right]. \qquad (4)
\end{aligned}
$$

At present, this equation has been accepted as the general shape equation of axisymmtric vesicles [11–13]. Although it seems that finding rigorous solutions of such a complex differential equation is almost impossible, we do find two solutions of the differential equation.

In geometry, Delaunay's surfaces can be constructed by rolling a given conic section on a line in a plane, and rotating the trace of a focus about that line. In algebra, the surfaces are simply expressed as

$$\sin\psi(\rho) = a\rho + d\rho^{-1}, \qquad (5)$$

where the two parameters a and d determine the types of the surfaces: (i) the unduloids, $0 < ad < 1/4$, and (ii) the nodoids, $ad < 0$. The spheres and the circular cylinders correspond to the two limiting cases: when $d \to 0$ the unduloids become spheres, and when $ad \to 1/4$ the unduloids degenerate to cylinders. Here, we do not discuss catenoids described by Eq. (5) when $a = 0$ which are the only minimal surfaces of revolution, because the present Letter concerns surfaces with nonzero values of the mean curvature.

To study surfaces not with constant mean curvatures, we show the new solution of Eq. (4),

$$\sin\psi(\rho) = a\rho + b + d\rho^{-1}, \qquad (6)$$

where b is an additional parameter. This solution offers a new mathematical concept beyond Delaunay's surfaces and physical insight into various vesicle surfaces. First, second, and third differentiations of ψ with respect to ρ give

$$\frac{d\psi}{d\rho} = \frac{1}{\cos\psi}(a - d\rho^{-2}), \qquad (7)$$

$$\frac{d^2\psi}{d\rho^2} = \frac{\sin\psi}{\cos^3\psi}(a - d\rho^{-2})^2 + \frac{2d}{\cos\psi}\rho^{-3}, \qquad (8)$$

and

$$
\begin{aligned}
\frac{d^3\psi}{d\rho^3} = {} & \frac{1 + 2\sin^2\psi}{\cos^5\psi}(a - d\rho^{-2})^3 \\
& + 6d\frac{\sin\psi}{\cos^3\psi}(a - d\rho^{-2})\rho^{-3} - \frac{6d}{\cos\psi}\rho^{-4}, \qquad (9)
\end{aligned}
$$

respectively. With these three expressions, we can straightforwardly show that Eq. (6) is a rigorous solution of Eq. (4) under the conditions

$$\frac{\Delta p}{k} = -2a^2c_0, \qquad (10)$$

$$\frac{\lambda}{k} + \frac{1}{2}c_0^2 = 2ac_0, \qquad (11)$$

$$b\left(2ad + 1 - \frac{1}{2}b^2 - 2c_0d\right) = 0, \qquad (12)$$

and

$$d\left(2ad - \frac{1}{2}b^2 - c_0d\right) = 0. \qquad (13)$$

We find from Eqs. (10)–(13) that there are three limiting cases for the solutions of Eq. (6): (i) $b = d = 0$, spheres with radius $r_0 = 1/a$ [5], (ii) $d = 0$, the Clifford torus with $b = \pm\sqrt{2}$ [8], and (iii) $b = 0$, Delaunay's surfaces given by Eq. (5). In case (i), the radius of the sphere r_0 satisfies

$$\Delta p r_0^3 + 2\lambda r_0^2 - kc_0r_0(2 - c_0r_0) = 0,$$

which has been reported previously [5], with Eqs. (10) and (11). In case (ii), Eqs. (10) and (11) are identical to the relations among Δp, λ, and c_0 for the Clifford torus when $a = -1$ [8]. In the general case of $bd \neq 0$, we obtain $d = 1/c_0$ and $b = \pm\sqrt{2 + 4(a/c_0 - 1)}$ from Eqs. (12) and (13), and hence Eq. (6) becomes

$$\sin\psi(\rho) = a\rho \pm \sqrt{2 + 4\left(\frac{a}{c_0} - 1\right)} + \frac{1}{c_0}\rho^{-1}. \qquad (14)$$

Deuling and Helfrich have numerically calculated unduloids to interpret the myelin shapes of red blood cells [14]. We should note that from inspection of their calculation their results are just Delaunay's surfaces and thus do not include the surfaces described by Eq. (14).

In the following, we assume $c_0 > 0$, introduce a new parameter $\rho_m = 1/\sqrt{ac_0}$, and set $x = \rho/\rho_m$ and $\alpha = (\rho_m c_0)^{-1}$, where ρ_m is the point at which $\psi(\rho)$ has the maximum value, i.e., $d\sin\psi(\rho)/d\rho = 0$ at $\rho = \rho_m$. The assumption of $c_0 > 0$ is associated with that in the pioneering work by Deuling and Helfrich [14]. We then reduce Eq. (14) to the dimensionless form

$$\sin\psi(\rho) = \alpha(x + x^{-1}) - \sqrt{4\alpha^2 - 2}, \qquad (15)$$

where

$$\alpha \geq 3/4, \tag{16}$$

which is due to the constraint of $|\sin\psi| \leq 1$. The positive branch of Eq. (14) is inappropriate because of the same constraint. We note that Eq. (15) represents unduloidlike shapes. The representative shape is shown in the inset of Fig. 1 at $\alpha = 1.1\alpha^-$, where $\alpha^- = 3/4$. In Fig. 1, we display the half-period of the contour of the shapes at different values of α in the unit of α^-. It is evident from Fig. 1 that when $\alpha \to \alpha^-$ the unduloidlike shape becomes a circular cylinder with the radius of $x = 1$ or $\rho_0 = \rho_m = (4/3)c_0^{-1}$. It is also evident from Eq. (15) that when $\alpha = \alpha^-$ we have $x = 1$ and $\psi = \pi/2$. Ou-Yang and Helfrich have derived the relation for a circular cylinder [5]

$$\Delta p \rho_0^3 + \lambda \rho_0^2 + \frac{k}{2}(c_0^2\rho_0^2 - 1) = 0. \tag{17}$$

The present results, $\alpha = \alpha^-$ or $\rho_0 c_0 = 4/3$, $a = 1/\rho^2 c_0$, and Eqs. (10) and (11) automatically satisfy this relation. This provides a comprehensive example of the validity of the present result. In addition, it has been shown that the cylinder can be seen as a torus with extremely small ratio of the outer diameter to the width of the ring and hence Eq. (17) again holds [8].

Equation (15) does not give surfaces similar to nodoids which are included in Delaunay's surfaces expressed by Eq. (5) with $H = -c_0/2$. This does not mean that the nodoidlike solutions with $H \neq$ const are not the solutions of Eq. (4). Indeed, we can show that Eq. (4) also has a solution similar to nodoids by using our recent result [13],

$$\sin\psi = c_0 \rho \ln(\rho/\rho_B), \tag{18}$$

where ρ_B is an integration constant. In Ref. [13], we have proved that Eq. (18) is a rigorous solution of Eq. (4) under the condition $\Delta p = \lambda = 0$ and have shown that Eq. (18) represents a circular biconcave discoid, the shape of the red blood cell, for $c_0 < 0$. We point out here that, for $c_0 > 0$ and $\rho_B c_0 > e$, Eq. (18) represents the nodoidlike shapes shown in the inset of Fig. 2 , which are different from the nodoids of Delaunay's surfaces whose nodules are inscribed onto the inner surface [9]. In Fig. 2, we show the half-period of the nodoidlike shapes for various values of β, where $x = \rho/\rho_B$ and $\beta = \rho_B c_0$. We find from the figure that the nodoidlike shape degenerates to a torus with a generating ring that is not a right circle. For $\beta \to \infty$ the torus has an infinitesimal generating oblate circle. We note that this surface is not a circular cylinder, but a tape, as predicted previously [5]. It has been proven that a torus with a right circular generating ring is a solution of Eq. (2) only when the ratio of radii of the generating circles is $1/\sqrt{2}$ or 0, i.e., the torus is a Clifford torus or a circular cylinder, respectively [8].

In summary, we have shown that Delaunay's surfaces with constant mean curvature, Eq. (5), are solutions of the Helfrich variation problem in the case of a vesicle with axisymmetry. We have developed new surfaces beyond Delaunay's surfaces; we have shown that the surfaces of revolution *not* with constant mean curvature, described by Eqs. (6) and (18), are solutions of the same problem. These solutions are surfaces similar to unduloids and nodoids of Delaunay's surfaces. In addition, the solutions become spheres, circular cylinders, and tori in certain limiting cases. We note that the surfaces of revolution with $H \neq$ const, which include Delaunay's surfaces as a special case, are of mathematical as well as physical importance. We hope that the surfaces predicted in this Letter, especially the nodoidlike shapes in Fig. 2, will be

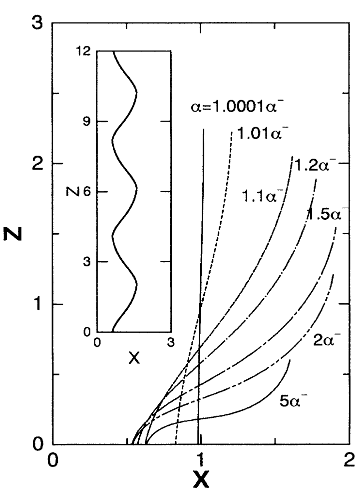

FIG. 1. Half-period of unduloidlike shapes at different values of α in the unit of α^- (= 3/4). Only one quadrant is shown. The inset illustrates three periods of the unduloidlike shape with $\alpha = 1.1\alpha-$. The axis of symmetry is the z axis.

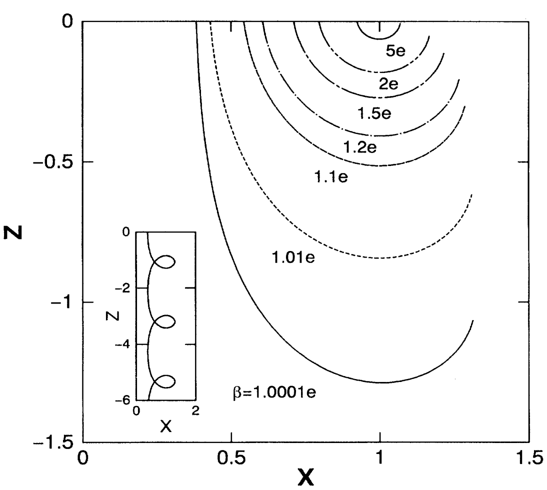

FIG. 2. Half-period of nodoidlike shapes for $c_0 > 0$ at different values of β in the unit of e, where $\ln(e) = 1$. Only one quadrant is shown. The inset shows three periods of the nodoidlike shape with $\beta = 1.0001e$. The axis of symmetry is the z axis.

experimentally demonstrated soon, as Plateau realized all Delaunay's surfaces using soap film experiments.

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