From the Plateau problem to periodic minimal surfaces
in lipids, surfactants and diblock copolymers

by

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Abstract

The novel method is presented for generating periodic surfaces. Such periodic
surfaces appear in all systems which are characterized by internal interfaces and
which additionally exhibit ordering. One example is the system of the AB diblock
copolymers, where the internal interfaces are formed by the chemical bonds between
the A and B blocks. In this system at least two bicontinuous phases are formed:
the ordered bicontinuous double diamond phase and gyroid phase. In these phases
the ordered domains of A monomers and B monomers are separated by the periodic
interface of same symmetry as the phases themselves. Here we present the novel
method for the generation of such periodic surfaces based on the simple Landau-
Ginzburg model of microemulsions. We test the method on four known minimal
periodic surfaces (P,D,G and I-WP), find two new surfaces of cubic symmetry,
show how to obtain periodic surfaces of high genus and n-tuply-continuous phases
((n > 2) So far only bicontinuous (n= 2) phases have been known. We point that the
Landau model used here should be generic for all systems characterized by internal
interfaces, including the diblock copolymer systems.
I. Introduction

Triply periodic minimal surfaces are paradigm of surfaces in cubic bicontinuous phases formed by biological molecules (lipids) and surfactants in aqueous solutions\(^1\-\text{3})\), silicate membranes macrostructures\(^4\,5\), zero potential surfaces in ionic crystals\(^6\), and domain interfaces in diblock copolymer structures\(^7\text{ab}\)). They can be used in the description of structures of carbon\(^8\) \(\text{C}_{60}\), blue phases of thermotropic liquid crystals\(^9\), blue phases of DNA\(^10\), and possibly cellular structures in the earliest stages of embryogenesis\(^11\) and chloroplast structures within plant cells\(^12\). Here we present the novel method for their generation based on the simple Landau-Ginzburg model of microemulsions\(^13\). We test the method on four known minimal surfaces (P,D,G and I-WP, Table 1. and Fig.1), find two new surfaces of cubic symmetry (Figs.(2,3) and Table 1), show how to obtain periodic surfaces of high genus (Fig.4) and \(n\)-tuply-continuous phases (\(n>2\)) (quadruply-continuous gyroid structure is shown in Fig.5). So far only bicontinuous \((n=2)\) phases have been known.

Most biological molecules (e.g. lipids) have both polar and nonpolar segments and therefore they self-assemble, in water solutions, in bilayers. These bilayers can further organize in periodic one, two or three dimensional structures, similarly to soaps and detergents, as was explicitly demonstrated by Luzzati et al.\(^1\) using x-ray technique. Although the symmetry of these phases can in principle be obtained from the x-ray or neutron scattering it is not clear how to obtain their geometry and topology from experimental studies alone. That is why the periodic minimal surfaces\(^14\-\text{16}) have served as paradigm of the periodic biological (or surfactant) surfaces formed in solutions. Minimal surface is the surface of zero average curvature at every point. The fact that such surfaces can form three dimensional periodic structures was discovered by Schwarz in XIX century. There are two ways of gener-
ating such surfaces: either from the definition of the average curvature\textsuperscript{15,16}) or from the Weierstrass parametrization\textsuperscript{17,18}). Both methods are related to the geometry of the problem and not to the physics of soaps, detergents or biological molecules in solutions. Here we fill the gap in our understanding of the connection between physical interfaces and geometrical models by presenting a simple physical model of microemulsions\textsuperscript{13}) and showing explicitly that physical interfaces in the model are periodic minimal surfaces. As a by-product of our research we have obtained a new method of generating periodic minimal surfaces. We have tested the method on four known surfaces i.e. P,D Schwarz and G (Fig.1),I-WP Schoen surfaces and proved its efficiency by generating new surfaces of cubic symmetry (Table 1 and Figs.2,3). The knowledge of new types of surfaces can be useful in indentifying them in real systems. In the lipid-water system six cubic phases have been found, out of which only four have been determined unambiguously\textsuperscript{3}).

The structure shown in Fig.5 is of special interest. It shows three periodic surfaces of nonpositive Gaussian curvature partitioning the space into four continuous disjoint subvolumes, making a periodic quadruply continuous structure. In all the examples shown in Figs. (1-4) a single surface partitions the space into two subvolumes and consequently the emerging structures are only bicontinuous. The idea of quadruple continuity or in general n-tuple continuity (n> 2) has not been previously considered.

The following simple experiment can be used for the direct visualization of a simple patch of minimal surface: Take a metal non-planar frame and immerse it in the water solution of soap. The soap bubble which forms on this frame assumes the shape that minimizes its surface free energy associated with the surface tension and consequently it forms a surface of least area. Thus these surfaces are called minimal surfaces. Such experiments can be traced back to Leonardo da Vinci\textsuperscript{19}) but in fact
the detailed studies of this type were done and published by Plateau\textsuperscript{20}) and hence later the problem of surface of least area spanning a given loop has been named the Plateau problem.

The history of physics and mathematics of minimal surfaces ran in parallel. Lagrange in 1761 (before Plateau) derived the equations for a surface of least area that is equivalent to the condition of vanishing mean curvature at every point on the surface. The representation of these surfaces in terms of harmonic functions was given by Weierstrass in 1866 and this representation has served many researchers up to date for their generations. A new qualitative insight into the mathematics of the problem was obtained by Schwarz and his student Neovius, who showed that simple patches of minimal surfaces can be put together to give smooth periodic three dimensional structures, which are called now triply periodic minimal surfaces (TPMS) or sometimes infinite periodic minimal surfaces (IPMS). They identified five phases three of which were of the cubic symmetry i.e. P, D and C(P). Plateau and Schwarz in fact entertained scientific contacts, but none of them had envisaged the role of these surfaces as physical interfaces. Rediscovery of the problem is due to Schoen\textsuperscript{14}), who identified 4 new surfaces of cubic symmetry(G, I-WP, F-RD and O,C-TO). In 1976 Scriven\textsuperscript{21}) observed that such surfaces could be used for the description of physical interfaces appearing in ternary mixtures of water, oil and surfactants. The surfactant molecules, which are the main ingredient of soaps and detergent have the ability to solubilize oil in water, two liquids which in the binary mixture at normal conditions are immiscible. This ability stems from their chemical structures; a surfactant molecule has the polar and nonpolar segments at two ends and thus is simultaneously hydrophobic and hydrophilic. The term amphiphilic (from Greek word loving both) molecule is used since one end (polar) of the molecule is well solubilized in water while the other (nonpolar) in
oil. Hence the molecule preferably stays at the interface between oil and water, forming a monolayer interface. At high concentration of surfactants the physical interface made of these molecules orders, forming periodic structures of various symmetries. In the water solution, without oil, the molecules form bilayers instead of monolayers. In 1967/68 Luzzati et al.\(^1\) observed this type of ordering in the lecithin-water and lipid-water systems. One of the phases observed by them was the phase of the same symmetry as the G Schoen minimal surface. It seems that both Schoen's and Luzzati discovery were made independently thus we shall use the name Schoen-Luzzati gyroid phase hereafter (Fig.1). In fact this phase appears to be very common in biological systems. Another example of such surfaces is found in the system of diblock copolymers, commercially important materials for the production of plastics. AB diblock copolymer consists of two macromolecules chemically bonded together. At low temperatures the system forms ordered A-rich and B-rich domains, with the points of bondage at the interface between the domains. In 1988 Thomas et al.\(^7\) observed that the PS/PI (polystyrene/polyisoprene) diblock copolymer forms a structure of the same symmetry as the D (diamond) Schwarz surface and argued on the basis of the relative volume fraction of PS and PI component that the resulting physical interface must be the surface of constant mean curvature at every point of the surface. Such surface belongs to the family of minimal surfaces\(^{14}\). Recently they have also observed the gyroid structure (Fig.1) in the same system in the weak segregation regime\(^7\).

Surfaces are ubiquitous. Even in the ionic crystals one can imagine a periodic zero potential surfaces (POPS in short), having same symmetry as the crystal\(^6\). Although, POPS do not have usually the same geometry as minimal surfaces (their mean curvature varies along the surface), nonetheless they share the same topology (genus etc) and symmetry as the latter.
This short historical survey points to the immense importance of minimal surfaces in many systems ranging from physics to biology and chemistry. We think that this is not the last word in the story of minimal surfaces and their application. Below we present a new method for their generation.

II. The model

The simple Landau-Ginzburg model considered in this paper has been proposed by Teubner and Strey\textsuperscript{24} and Gompper and Schick\textsuperscript{13,25} based on the neutron scattering experiments performed on microemulsion (homogeneous ternary mixture of oil, water and surfactant) and later experiments and theory of their wetting properties\textsuperscript{26,27}. The Landau-Ginzburg free energy functional has the following form:

\[ F[\phi] = \int d^3r \left( |\triangle \phi|^2 + (g_2 \phi^2 - g_0) |\nabla \phi|^2 + (\phi^2 - 1)^2(\phi^2 + f_0) \right) \]

where \( \phi \), the order parameter, has the interpretation of the normalized difference between oil and water concentrations; \( g_2, g_0 \) are two positive constants and \( f_0 \) can be of either sign. The sign of the latter depends on the stability of bulk microemulsion phase (average order parameter is zero): for \( f_0 > 0 \) microemulsion is a metastable bulk phase whereas pure water phase (\( \phi = 1 \)) or pure oil phase (\( \phi = -1 \)) are stable; for \( f_0 \leq 0 \) microemulsion is stable. For \( g_0 > 2 \) the system can undergo a transition to periodically ordered phases where water rich domains and oil rich domains order similarly as in the case of the aforementioned AB diblock copolymer system\textsuperscript{7ab}. The interface between the domains corresponds to \( \phi(r) = 0 \). The only stable ordered structure is the lamellar phase, however there exists a large number of ordered metastable phases, corresponding to the local minima of the functional (1). It is the main result of this paper that in many of these metastable phases the physical interface between the domains, given by \( \phi(r) = 0 \), is a triply periodic
minimal embedded surface. We have obtained 6 cubic structures listed in Table 1 including known examples of P, D, G (Fig.1), I-WP and and two new phases shown in Figs. (2,3). The high genus surface shown in Fig.4 has nonpositive Gaussian curvature and large internal surface area, but numerical accuracy does not allow us to claim that it is a minimal surface.

In order to find the local minima of the functional we have discretized Eq(1) on the cubic lattice. Thus the functional $F[\phi(r)]$ becomes a function $F(\{\phi_{i,j,k}\})$ of $N^3$ variables, where $Nh$ is the linear dimension of the cubic lattice and $h$ is the distance between the lattice points. Each variable $\phi_{i,j,k}$ represents the value of the field $\phi(r)$ at the lattice site $S = (i,j,k)$, and the indices $i,j,k$ change from 1 to $N$. In our calculations we use $N=129$, which results in over 2 million points per unit cell. The first and second derivatives in the gradient and laplacian term of the functional (1) were calculated on the lattice according to the following formulas\textsuperscript{28)

$$g(r) \frac{\partial \phi(r)}{\partial x} \rightarrow g_{i,j,k} \frac{\phi_{i+1,j,k} - \phi_{i-1,j,k}}{2h}$$

and

$$\frac{\partial^2 \phi(r)}{\partial x^2} \rightarrow \frac{1}{12h^2} \left( -\phi_{i+2,j,k} + 16\phi_{i+1,j,k} - 30\phi_{i,j,k} + 16\phi_{i-1,j,k} - \phi_{i-2,j,k} \right) \quad (3)$$

and similar in $y$ and $z$ directions.

We impose on the field $\phi(r)$ the symmetry of the structure, we are looking for, by building up the field inside a unit cubic cell from a smaller polyhedron, replicating it by reflections. For example structures having $m3m$ space group symmetry are build of quadrirectangular tetrahedron cut out of the unit cubic cell by the planes of symmetry. The polyhedrons that we have used to construct the cubic unit cells are the same as those described by Coxeter as kaleidoscopic cells\textsuperscript{14,15}. Such a procedure enables substantial reduction of independent variables $\phi_{i,j,k}$ in the function
\( F(\{\phi_{i,j,k}\}) \). We impose on the field \( \phi(\mathbf{r}) \) the periodic boundary conditions in \( x \), \( y \) and \( z \) directions.

The topology of the structure is set up by building the field \( \phi(\mathbf{r}) \) first on a small lattice \( N = 3 \) or \( 5 \) analogically to a two component (A,B) molecular crystal. The value of the field \( \phi_{i,j,k} \) at a lattice site \( S = (i,j,k) \) is set to 1 if in the molecular crystal an atom A is in this place, if there is an atom B \( \phi_{i,j,k} \) is set to -1, if there is an empty place \( \phi_{i,j,k} \) is set to 0. Next the small lattice can be enlarged to desired size by changing the number of points from \( N \) to \( 2N - 1 \) and finding the values of \( \phi_{i,j,k} \) in new lattice sites by interpolation.

We have used the conjugate gradient method\(^{29} \) to find a minimum of the function \( F(\{\phi_{i,j,k}\}) \). It is highly unlikely, because of numerical accuracy, that a value of the field \( \phi_{i,j,k} \) at a lattice site \( S = (i,j,k) \) is zero. Therefore the points of the surface have to be localized by linear interpolation between the neighbour sites of the lattice. This approximation is legible because the field \( \phi(\mathbf{r}) \) is very smooth.

The structures of simple topology are formed for small sizes of a unit cell. The length of a unit cell, \( d \), has to be increased to obtain the structures of complex topology.

In order to calculate genera of the surfaces we use the following method. For every point \( P = (i,j,k) \) where \( i \in [1,N-1] \) , \( j \in [1,N-1] \) , \( k \in [1,N-1] \) we examine a cube formed by this point and vectors \( \mathbf{e}_1 = [h,0,0] \) , \( \mathbf{e}_2 = [0,h,0] \) , \( \mathbf{e}_3 = [0,0,h] \). If the values of the field at all vertices of this cube are not the same sign the surface \( \phi(\mathbf{r}) = 0 \) must lie inside this cube. Therefore points of the surface lie on the edges of the cube. These points form a polygon: a triangle, a tetragon, a pentagon, or a hexagon. There are no other possibilities. The edges of this polygon must lie on the faces of the cube, the vertices on the edges of the cube. Therefore each edge of the polygon belongs to two cubes and each vertex to four cubes. Thus
we can calculate the number of edges $E$, vertices $V$, and faces $F$ of the surface inside a cubic unit cell by summing $F, E, V$ in all small cubes and taking the number of vertices with a weight $1/4$ and the number of edges with a weight $1/2$. Next the Euler characteristic $\chi$ can be calculated from the formula $\chi = F + V - E$ and genera $g$ of the structures from $g = 1 - \chi/2$.

From the form of Eq(1) one can realize that indeed for some local minima of (1) the average curvature given by$^{30,31}$:

$$H = -\frac{1}{2} \nabla \left( \frac{\nabla \phi}{|\nabla \phi|} \right) = -\frac{1}{2} \frac{\Delta \phi}{|\nabla \phi|} + \frac{\nabla \phi \nabla |\nabla \phi|}{2|\nabla \phi|^2} \quad (4)$$

vanishes at every point of the $\phi(r) = 0$ surface. It follows from the second term of Eq(1) that $|\nabla \phi|$ should have the maximal value for $\phi(r) = 0$ and consequently the second term (which after a small algebra can be written as $(\partial|\nabla \phi|/\partial n)/2|\nabla \phi|$, with $\partial n$ denoting the derivative along the normal to the surface) in Eq(4) vanishes. Also for the $\phi, -\phi$ symmetry we know that $H$ averaged over the whole surface should be zero. It means that either $\Delta \phi$ is exactly zero at the surface or it changes sign. From the first term of Eq(1) it follows that the former is favored and consequently $H = 0$ at every point at the surface and hence the surface is minimal. We have checked numerically that indeed the surface $\phi(r) = 0$ coincides with $H = 0$ in all the cases listed in Table 1.

III. Summary

We have presented the novel method for the generation of periodic surfaces. The method should be useful for mathematicians working in topology and geometry of surfaces, crystallographers studying self assembling soft matter systems, physicists and biologists. The method can also find application in the design of well characterized mesoporous materials. For example in the production of such materials the internal interface in the surfactant system is used as a template for the
three dimensional polimerization of silicate. One obtains in such a way the ordered silicate pore structure characterized by the same symmetry, topology and geometry as the surfactant template.

Surfaces are also important for biology. We finish this report by giving a few examples, not commonly known, of the influence of the physical structure on the functions of biological systems. The concentrated solutions of DNA\textsuperscript{32–36}, polypeptides\textsuperscript{36} and polysaccharides\textsuperscript{36} form ordered phases. It has been also observed that DNA in bacteriophages and sperm nuclei of sepia, trout and salmon exhibit ordering\textsuperscript{34}. Moreover DNA has been shown to form blue phases\textsuperscript{10}. The activity of DNA (renaturation, transcription or replication) can be enhanced in the condensed phase\textsuperscript{34}. For example it has been observed that in the phenyl, water emulsion the renaturation rate can be increased 1000 times in comparison to the renaturation in standard conditions\textsuperscript{37}. The renaturations most likely takes place at the interface between the two phases where the local density of DNA strands can be greatly increased. Despite of the accumulation of great body of experimental results the problem of how the geometry and symmetry of surfaces can affect the biological processes remains still an open problem.

Let us at the end point out that the Landau-Ginzburg free energy which we have used in this paper should be generic for all the systems characterized by the internal interfaces. The same free energy has been obtained in the ternary mixture of A homopolymers, B homopolymers and AB diblock copolymers\textsuperscript{38}.

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Table Caption

Table 1. The minimal surfaces obtained using our method. In the second column the symmetry is given. We have assumed that two sides of the surface are equivalent. The volume fraction, \( V/V_{tot} \) (third column) is the volume at one side of the surface, \( V \), divided by the total volume of the unit cell, \( V_{tot} = d^3 \), where \( d \) is the dimensionless linear size of the unit cell. The error of the calculation of the volume fraction has been smaller than 0.001 for most structures. The surface area, \( S \) (fourth column) is divided by \( V^{2/3} = d^2 \) i.e. is calculated per side of the unit cubic cell. The genus, \( g \), has been calculated from the formula \( 1 - \chi/2 \), where \( \chi \) is the Euler characteristic per unit cell. All these data are model independent. The model dependent parameter is the size of the unit cell \( d \) and therefore is not listed in the Table; here we find for P surface \( d = 7.88 \), for D, \( d = 12.56 \), for G \( d = 10.08 \) for I-WP \( d = 11.78 \) for BFY (“butterfly”) \( d = 16.70 \) and for CPD \( d = 15.12 \) (obtained for \( g_0 = 3 \), \( f_0 = 0 \) and \( g_2 = 4\sqrt{1+f_0+g_0+0.01} \)). All these structures were obtained for many different values of \( g_2 \), \( g_0 \) and \( f_0 \). In all cases the parameters shown in the Table were the same, although \( d \) was different. For P and G surfaces the worst error of the average curvature was 0.01 (typical Gaussian curvature was -0.3), but at most points the average curvature was smaller than 0.005. In the D and I-WP surface the error was smaller than 0.015, whereas for the more complicated structures (BFY and CPD) it was smaller than 0.05. Because of the errors we cannot claim with certainty that BFY and CPD phases are minimal. In the Table we also give the exact values known from literature (if available) for comparison (shown in parenthesis with the reference number). The calculations were performed on the workstation IBM RS/6000 3BT and the figures were prepared using Data Explorer.
Figure Captions

Fig.1 The Schoen-Luzzatti gyroid, G, structure. One unit cell is shown. The white surface (gyroid minimal surface) located at $\phi(r) = 0$ (see Eq(1)) corresponds to the surface dividing oil ($\phi < 0$) and water ($\phi > 0$) channels into disjoint subvolumes. This phase has been recently observed in the diblock copolymer system\textsuperscript{7b}.

Fig.2 The new (presumably) minimal surface BFY (Table 1). Legend as in Fig.1. (a) $1/8$ of the unit cell. (b) One unit cell.

Fig.3 The new (presumably) minimal surface CPD (Table 1). Legend as in Fig.2 (a) $1/8$ of the unit cell. (b) One unit cell.

Fig.4 The new structure of cubic symmetry (same as D surface Table 1.) and very high genus $g = 73$. The legend as in Fig.1. The surface area $S/V^{2/3} = 8.25 \pm 0.05$, $V/d^3 = 0.5$, $d = 28.88$ (generated at the same values of $g_0 f_0$ and $g_2$ as in Table 1). The surface has nonpositive Gaussian curvature at every point of the surface. It is possible that it is also a minimal surface. (a) $1/8$ of the unit cell. (b) One unit cell.

Fig.5 The quadruply continuous cubic structure of the gyroid symmetry (Table 1). Here there are four disjoint subvolumes (two ”water” channels and two ”oil” channels) separated by three surfaces. The middle surface is the G minimal surface (see Fig.1), whereas the other two are not. All the surfaces have nonpositive Gaussian curvature at every point. Each of the three surfaces has the same genus per unit cell as the G phase (Table 1). The surface area $S/V^{2/3} = 7.55 \pm 0.05$, $V/d^3 = 0.5$, $d = 26.28$ ($g_0 f_0$ and $g_2$ same as in Table 1). (a) $1/8$ of the unit cell. (b) One unit cell.
| Name | Symmetry | Volume Fraction | Surface Area | Genus |
|------|----------|-----------------|--------------|-------|
| P    | Im$\bar{3}$m | 0.5 | 2.3453 | 3 |
|      |          |                 | [2.3451068]$^{(14,15)}$[3]$^{(14,15)}$ | |
| D    | Pn$\bar{3}$m | 0.5 | 3.8387 | 9 |
|      |          |                 | [3.8377862]$^{(15)}$ | |
| I-WP | Im$\bar{3}$m | 0.533 | 3.4640 | 7 |
|      |          |                 | [3.4646016]$^{(18)}$ | |
| G    | Ia$\bar{3}$d | 0.5 | 3.0919 | 5 |
| CPD  | Im$\bar{3}$m | 0.515 | 4.3588 | 14 |
| BFY  | Im$\bar{3}$m | 0.5 | 4.9641 | 19 |