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Mathematical Models of Triple Junctions

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Abstract. Including triple junctions in rigorous mathematical models had very limited success until about 25 years ago. This paper surveys the mathematical results on triple junctions since then. Anisotropic surface energies, zero triple line energy, positive triple line energy, and negative triple line energy are all considered, in both sharp and diffuse interfaces. In particular, the problems with negative line junction energies in sharp interface theory are presented. Both equilibrium and growth models are reviewed, in 2 and 3 dimensions.

Keywords: survey, mathematical models, positive triple junction line tension, negative triple junction line tension, polycrystalline grain growth, motion by weighted mean curvature, triple junctions

1. Introduction

Plateau [1] observed that soap films and soap bubble clusters were smooth surfaces except for the singular sets where three pieces of surface came together at equal (120°) angles along smooth curves, and isolated points where four such triple junction curves met, at equal ($\arccos(-1/3)$) angles, bringing together six pieces of surface. However, it took until 1976 before a mathematical model of soap bubbles and soap films was published and Plateau's observations were able to be proved as theorems.

The object of this paper is to say what the difficulties were that made it take so long to produce a good model and to survey the mathematical results proved since then. Results on anisotropic surface free energies such as could occur in polycrystalline materials will be reviewed as well as those for the isotropic case of the area integrand. Triple junction line energies will be considered, both positive and negative, with an emphasis on the mathematical difficulties posed by negative junction energies. Finally, the results of trying to model some growth laws will be reviewed.

Elastic energies and the motion of dislocations will not be considered in this paper. Similarly, atomistic calculations and effects will be omitted. The object is to see what results can be derived purely from surface energy and line energy effects.

2. Soap Films and Bubbles

It is not very difficult to prove that the only cones (i.e., surfaces consisting of rays from the origin) which can possibly be area-minimizing are those whose intersection with the unit sphere are great circle segments meeting three at a time at equal (120°) angles. Deformations of the surface inside the sphere would decrease surface area otherwise. And it is not too difficult to classify all such nets on the sphere [2] (although the first person who tried it missed some [3]) and to prove that the cones over all but three of them are not area-minimizing [4]. The three remaining are a single great circle, whose cone is a plane, three half circles, whose cone is the standard straight triple junction, and six segments forming a curved version of the edges of a regular tetrahedron. It is also not difficult to show that three cones are then the only possible tangent cones (resulting from translating a point to the origin and then successively expanding the surface by a factor $1/r_i$ for some sequence of r_i going to zero) for an area minimizing surface.

What is difficult is to show that an area-minimizing configuration must consist of smooth pieces of surface that are smooth enough at their edges that they meet along smooth curves and smoothly at isolated points. Attempts to impose this as an *a priori* condition failed, because existence of solutions could not then be proved.

Even uniqueness of tangent cones is an issue, until proven otherwise. (Does the particular sequence of r_i going to zero that is chosen above matter?)

Jesse Douglas earned one of the first two Fields medals, in 1933, by proving that there exist minimal surfaces (without the possibility of triple junctions) having as boundary the smooth image of a circle [5]. The Fields medal has often been called the mathematician's equivalent of a Nobel Prize. But it was not until 1976 that triple junctions and other soap-film singularities were successfully modeled. The theory of what were (infelicitously) called "(F, ϵ, δ) minimal sets with boundary in B" was developed by Fred Almgren to be able to handle surfaces with singularities such as occur in soap bubbles. The theory of such surfaces was published by Almgren in 1976 [6]. Almost simultaneously, this author published the proof that the singular structure of any such 2-dimensional surface in R^3 was just what Plateau observed [4].

It is worthwhile to give the definition of (F, ϵ, δ) minimal sets, since they precisely capture the idea of stable force balance. The main idea is that for such sets, deformations with small support cannot decrease surface energy by a fraction much different from one. The F refers to the surface free energy function; nowadays, such a function is commonly called γ in materials science, σ in physics, and Φ in mathematics. The ϵ refers to a function $\epsilon(r)$ which goes to zero as r goes to zero. For soap films and bubbles, it is appropriate to use $\epsilon(r) = Cr$ for some constant C. And δ refers to the size of the support of the deformation. The definition is now as follows: for any point p in a surface, let ϕ be any Lipschitz function taking the ball of radius r around p into itself. It is not required to be one-to-one (it can smash parts of the surface together), but we do require $r < \delta$; ϕ being Lipschitz means that it has a bound to the rate at which it stretches space (that is, there exists some L such that $|\phi(p_1) - \phi(p_2)| \leq L\,|p_1 - p_2|$ for all points p_1, p_2). A set S is (F, ϵ, δ) minimal if for any p in S and any such ϕ, the energy of S (as measured by F) within that ball is never more than $(1 + \epsilon(r))$ times the energy of $\phi(S)$ within that ball. If S were actually minimizing, then the factor would be 1 and not $1+\epsilon(r)$; the extra leeway (which goes to zero as the ball gets smaller) is to allow things like the roundness of soap bubbles. The limit imposed by δ is to prevent one from squashing a whole giant bubble down to a point, stretching a thin outer layer beyond the bubble to maintain continuity and the Lipschitz property of the function.

The theorems are then that clusters with prescribed volumes for the various bubbles exist as (F, ϵ, δ) minimal sets [6], that under certain conditions on F, *any* (F, ϵ, δ) minimal set with boundary is composed of Hölder-continuously differentiable pieces of surface except for a singular set of zero area [6], and that when F just measures surface area and the surfaces are 2 dimensional in R^3, this singular set is as Plateau described [4].

One of the important features of this definition, and of the theorems, is that they apply to a much broader class of surfaces than soap bubble froths. The definition captures the idea of *local* (because of the δ) stable force-balance (because in the limit as the radius r goes down to zero, energy cannot be decreased). Tangent cones to such surfaces are always minimizing, even if the surfaces themselves are not.

The difficulty of working with (F, ϵ, δ) minimal surfaces is that there is so little structure known ahead of time. Even when F measures area, the only easy step was to determine the structure of all possible tangent cones. The difficult part was accomplished by proving a type of "epiperimetric inequality" similar to the one proved by Reifenburg in his earlier on area-minimizing surfaces (without any triple junctions) in higher dimensions and co-dimensions [7, 8]. An article by Robert Kanigel [9] described part of this proof for non-experts. Simpler versions of several steps in the proof of this epiperimetric inequality were later proved by Brian White, and extended to isotropic surface free energies that are different on different interfaces [10, 11]. Also, the results were extended to fluids in containers with specified energies and hence contact angles [12].

Hoffman and Cahn [13] defined ξ vectors and developed a condition involving them which was necessary for a multi-junction to be part of a minimizing surface (more precisely, an (F, ϵ, δ) minimal set). Lawlor and Morgan [14] later proved that that condition was also sufficient for the tangent cone to separate regions of space with the least possible surface free energy. See the commentary on the Cahn-Hoffman papers [15] in Cahn's Selected Works.

There are no results known to the author on the smoothness of singularities for anisotropic surface free energies, except in the case where those energies are almost trivially related to area [16]. Therefore the key mathematical result that links the structure of minimizing tangent cones to the structure of (F, ϵ, δ) minimal sets is completely absent.

Computing (approximately) the shapes of surfaces with triple and point junctions has been made possible by Ken Brakke's remarkable computer program called the *Surface Evolver* [17]. This program computes

minima by evolving initial configurations toward lower total energy. Using the program effectively on complex geometries requires some training; for example, the program makes topological changes in surfaces only via instruction by the user. Nevertheless, *Surface Evolver* has been used to compute complicated geometries. And for curves in the two-dimensional plane with triple junctions, *Surface Evolver* makes topological changes quite easily with no user intervention. It is freely available from www.geom.umn.edu or www.susqu.edu/facstaff/b/brakke.

A diffuse interface between two regions is typically modeled by an order parameter u which is defined on R^3 and takes values in the real numbers. The energy of the interface in the isotropic case is usually taken to be $\int_{R^3}(\frac{\epsilon}{2}|\nabla u|^2 + \frac{1}{\epsilon}F(u)\,dV$, where F is a double well potential. One typical F to use is $F_1(u) = (1-u^2)^2/2$. Another is $F_2(u) = (1-u^2)/2$ if $|u| \le 1$ and $F_2(u) = \infty$ if $|u| > 1$. Anisotropic surface free energies result by replacing $|\nabla u|^2$ by $\Gamma^2(\nabla u)$ where Γ satisfies $\Gamma(\mathbf{p}) = |\mathbf{p}|\Gamma(\frac{\mathbf{p}}{|\mathbf{p}|})$. A flat interface with normal $\mathbf{n}$ where u goes from -1 as $\mathbf{x}\cdot\mathbf{n}$ goes to $-\infty$ to $+1$ as $\mathbf{x}\cdot\mathbf{n}$ goes to ∞ and that minimizes E has, when $F = F_1$, the equilibrium profile of $u = \tanh(\frac{\mathbf{x}\cdot\mathbf{n}}{\epsilon\Gamma(\mathbf{n})})$. When $F = F_2$, it is $u = \sin(\frac{\mathbf{x}\cdot\mathbf{n}}{\epsilon\Gamma(\mathbf{n})})$ if $|\mathbf{x}\cdot\mathbf{n}| \le \epsilon\pi/2$, $u = 1$ if $\mathbf{x}\cdot\mathbf{n} > \epsilon\pi/2$ and $u = -1$ otherwise. The surface energy per unit area of the sharp interface that this diffuse interface models is $\frac{4}{3}\Gamma(\mathbf{n})$ when using $F = F_1$ and $\frac{\pi}{2}\Gamma(\mathbf{n})$ when using $F = F_2$. There are many sources where this is derived; one such source is [18].

There are several options to model triple junctions in diffuse interfaces. One is to have multiple order parameters, one for each regions, with these usually required to take nonnegative values and to add to 1. (In the case of only two order parameters, u_1 would correspond to $(1+u)/2$ and u_2 to $1-u_1 = (1-u)/2$.) Another is to have just two variables u_1 and u_2 but to have multiple wells in the graph of F, each well corresponding to a different phase (the "jello mold" approach). This author does not know of any studies of equilibrium shapes of soap bubble clusters using such a method.

A different diffuse interface approach is to use a level-set method for a function u with little inherent physical meaning; this approach is often used for moving surfaces by their mean curvature, since one seeks viscosity solutions in a Hamilton-Jacobi formulation and all level sets then move by their mean curvature. This approach has been extended to model triple junction equilibrium and motion in [19]; published computations are in R^2 but it is asserted that the method could easily be extended to R^3. Again, this author knows of no studies of equilibrium shapes of soap bubble clusters using such a method.

3. Triple Junction Line Energies

In [20], Frank Morgan and this author considered what happens to the standard collection of soap film singularities if in addition to surface free energy there is a positive free energy per unit length of triple junctions. The least length of a set of points connecting the four corners of a regular tetrahedron is given by a set of five line segments, one from each corner plus one in the middle, with each endpoint of the middle segment being a standard 120° triple point where it meets segments from two of the corners. This suggests that the surface which has the least sum for the length of the junctions and the surface energy might have the tetrahedral singularities (where four triple junctions meet at a point) replaced by configurations that insert a small middle segment along which four surfaces meet. Indeed, we proved that such a configuration has less total energy than the tetrahedral singularity. Morgan has since done further mathematical work along this line [21, 22].

Many materials scientists, however, believe that triple junctions ought to have a *negative* energy per unit length. Gibbs [23] mentions that the apparent line tension of a triple junction is negative when "there really exists in stable equilibrium a filament of different phase from the three surrounding masses." Soap films are really composed of two films back-to-back. Rather than making sharp angles at a junctions, real soap films actually have a small amount of liquid running along their singularities. Plateau in fact called the triple junctions "liquid edges" [1]. Introducing this liquid decreases the surface energy by allowing the films to bend smoothly rather than sharply as they make that 120° angle. For a single straight wetted triple junction, the computation of energy decrease (including the bulk free energy contribution of the negative pressure of the liquid) appears in [24]. The amount that the energy is decreased is proportional to ϵ, the width of the cross section of the liquid region (which is composed of three circular arcs), and thus goes to zero as the volume goes to zero.

This idea of introducing additional liquid has been partially modeled mathematically; it is called a "wet foam." Thus one way to model negative triple junction line energies is to solve the different problem of a wet foam. One never measures the lengths of triple junctions here, nor assigns any energy to them; one only

measures volumes and the area of surfaces, which are counted with factor two (for the two interfaces with the air) where dry and with factor one (for only one interface with air) where wet. Thus it is rather indirect.

A useful tool in investigating wet foams is Brakke's *Surface Evolver*, mentioned above, which can also handle conditions such as this extra liquid. Several papers have been written on wet foams [25–27] but there does not yet exist a complete mathematical theory of them.

Directly using a negative triple junction line tension in a sharp interface theory leads to apparently unavoidable problems with existence of any surfaces in such theories. For example, onto any given surface, one could put a large number of long thin strips of extra surface, each shaped approximately as a portion of a cylinder with its axis in the long direction of the strip. Each strip would form triple junctions along its edges with the original surface, and hence each would contribute a negative constant times its length to the total energy. But there is no upper bound to the number of such skinny strips one could insert, and the surface energy contributed by of all the strips need be no more than a small fraction of the original energy of the surface. Thus one could drive the total energy towards minus infinity, just by adding lots of strips! Alternatively, one could take a given relatively straight triple junction, and in one of the three surfaces coming into the junction add a single strip with one long edge being highly wiggled and the other being along the original triple junction. One thereby has moved the triple junction over to the highly wiggled edge; on the rest of the strip, the original surface has been simply doubled up. (One could slightly relax this configuration so that the doubled surfaces separate slightly, thereby slightly decreasing the total surface area while maintaining the same new wiggly triple junction.) Thus the net added area is that of the strip, which can be as small as one likes by making it as narrow on average as one likes, whereas the total triple junction energy can be made arbitrarily negatively large by making enough wiggles.

A person might argue that these examples do not have 120° angles. There are two responses to that. One is the 120° condition is supposed to be a conclusion of a model and not a hypothesis. Another is that one can always go in very very close to the triple junction and adjust things so that the angles are 120°, without increasing area by much.

The best argument against these examples is that they require different parts of triple junctions to be very close to each other, and that for atomistic reasons this is physically impossible. But we are talking about trying to create consistent mathematical models here, and ignoring atomistics. So perhaps any mathematical theory needs an externally-imposed size constraint. It is not sufficient to impose any curvature constraint or curvature condition on the triple junction itself; again, counterexamples can be constructed, such as by having large thin circular tubes added to a surface, with extra strips along them creating more triple junctions. The curvature constraint would have to be on the surfaces themselves. All such theories appear to become very cumbersome, and it is far from clear that they could be made to be successful.

Other objections to the counterexamples given above are that they require nucleation: by looking only at what might dynamically appear, the idea is that these counterexamples might not arise. This invokes issues of metastability, and what type of perturbations are allowed. Widom [28] shows that if one uses smooth perturbations that grow in time while keeping the same shape, then a triple junction with negative line tension can be stable. But with other perturbations, it cannot be. For example, suppose one of the surfaces coming into a triple junction is locally described by $\{(x, y, 0): y \geq 0\}$. For each $t > 0$, consider the deformation of the triple line from $\{(x, 0, 0)\}$ to $\{(x, f(x), 0)\}$, with $f(x) = 0$ for $|x| > t^2$ and $f(x) = t - |x|/t$ for $|x| \leq t^2$. Remove the triangle of the original surface that is bounded by the original and the deformed triple junction, and add that triangle to each of the other two surfaces coming in to the original triple junction. The increase in area is the area of the triangle, namely t^3, and the increase in triple junction length is approximately $2t$. Clearly enough t, the net change in total energy with a negative triple junction energy is negative, for all small t. Thus this is an example of a continuous family of deformations which strictly decreases total energy as t increases. It is not, however, a perturbation of the form $tg(x)$. Yet why should we be limited to perturbations of that form?

On the other hand, diffuse interface theories have an effective negative triple junction energy automatically, as follows. We use the multiple-order-parameter model and assume we have three order parameters u_1, u_2, u_3 with $u_1 + u_2 + u_3 = 1$. We restrict ourselves to the isotropic situation and use an F extending F_2 to the three-order-parameter situation, so that the following is an equilibrium profile (and so are the profiles obtained by permuting the subscripts): $u_3 = 0$ everywhere, $u_1 = 0$ for $y < -\epsilon$, $u_1 = 1$ for $y > \epsilon$, $u_1 = \sin(\frac{y}{\epsilon})$

for $|y| \leq \epsilon$. Now restrict this profile to $x \geq 0$; it is the diffuse approximation to a sharp interface that is a ray from the origin along the x axis. Then rotate this profile by 120°, interchanging (u_3, u_1, u_2) for (u_1, u_2, u_3), and then by $-120°$, interchanging (u_2, u_3, u_1) for (u_1, u_2, u_3). To paste the three configurations together, one throws away the overlaps, a total of six small 30°-60°-90° triangles (two per interface) at the origin; the values coincide along the lines where they meet.

The net energy of those six small triangles is subtracted from the energy of the original three rays, thus decreasing the total energy. And this isn't even necessarily the minimum energy configuration: it is just one way to accomplish a triple junction. So the energy of the three diffuse interfaces meeting at a triple junction is less than the energy of three individual interfaces. Thus one obtains an effective negative triple junction line energy. The size of this bound on the negative triple junction energy is proportional to ϵ, with the value depending on the specific F; it goes to zero as the diffuse interface approaches its sharp interface limit.

Molecular dynamics simulations (in 3-D) also produce a negative contribution to the total free energy in the atomistic modelling of triple junctions [24].

4. Weighted Mean Curvature with Triple Junctions

The mean curvature of a surface at a point is often written as $\frac{1}{r_1} + \frac{1}{r_2}$, where r_1 and r_2 are the principle radii of curvature of the surface at that point. These radii are signed, however, with the sign depending on the orientation of the surface. If the center of curvature is on the same side as that pointed to by the orienting normal vector, then the sign is often taken as negative, and if it is on the opposite side, then the sign is often taken as positive. This choice means that the mean curvature of a sphere of radius r, oriented outward as the boundary of a ball, is $\frac{2}{r}$. The natural quantity is actually the mean curvature vector, which is the negative of the mean curvature times the orienting vector, and it does not depend on the choice of orientation. Mean curvature arises in studying equilibrium and growth shapes since it is the rate of increase of area of the surface with respect to volume changes under deformations of the surface. For an anisotropic surface free energy per unit area γ, mean curvature should be replaced by weighted mean curvature, which is the rate of increase of the surface's free enenergy with respect to volume. One way to write the weighted mean curvature of a surface which is smooth enough (at least twice differentiable) if the equilibrium shape for the surface free energy function γ has no facets or corners is as follows. Extend the function γ to all vectors by defining $\gamma(\mathbf{p}) = |\mathbf{p}|\gamma(\mathbf{p}/|\mathbf{p}|)$ for $\mathbf{p}$ nonzero (and $\gamma(0) = 0$), and assume γ is a convex function which is twice differentiable except at 0. Then the weighted mean curvature κ_γ of S at $\mathbf{x}$ is $\gamma_{11}\frac{1}{r_1} + \gamma_{22}\frac{1}{r_2}$, where γ_{ii} is the second derivative of γ in the ith principle direction of S at $\mathbf{x}$, evaluated at $\mathbf{n}(\mathbf{x})$. For a discussion of weighed mean curvature in general, including in the cases where the equilibrium shape does have corners and facets, see [29].

Where the surface is singular, the mean curvature is not defined. For example, if a surface is the flat surface $z = 0$ for $x < 0$ and the paraboloid $z = x^2$ for $x > 0$, then the mean curvature is not defined at the points where $x = 0$. Since the derivative is continuous, the discontinuity in curvature does not lead to an infinite driving force, as it would for the function with $z = x$ for $x > 0$. That is, the rate of change of area with volume goes to infinity for some small deformations for the second type of singularity, and does not for the first type.

Similarly, mean curvature is not traditionally defined at any triple junction. Once again, there is a distinction between the mean curvature being infinite and being discontinuous. The mean curvature is infinite if there is not force-balance, in the same way as it is for the example where $z = x$ for $x > 0$. And force-balance is exactly the concept captured by the definition of $(\gamma, \epsilon, \delta)$ minimal sets.

The most reasonable way to define the mean curvature, or the weighted mean curvature, in surfaces with triple junctions is again to look at the rate of change of area, or of total surface free energy, with volume swept out under deformations. The mathematical theory of varifolds, and the first variation of a varifold, was developed in part to define such terms [30, 31]. Where the first variation measure is singular, there is a lack of force balance; whereas the absolutely continuous part of the measure yields the (weighted) mean curvature.

5. Motion by Weighted Mean Curvature with Triple Junctions

The purpose of defining mean curvature is to determine when a surface is in equilibrium. It is also used to say how it moves, when reduction of surface free energy is part of the driving force for motion. In what follows, we do not make any additional assumptions about the physics of the motion of the triple junction;

in particular, we do not assume it has any special resistance to motion. We also assume that all triple junction line energies are zero. We consider motions that decrease bulk free energy plus surface free energy the fastest, in the L^2 sense. Such motions have the normal velocity v given by

$$v = M(\mathbf{n})(\omega - \kappa_\gamma)$$

where M denotes a normal-dependent mobility function and ω is a bulk driving force; here κ_γ denotes the weighted mean curvature as before.

A survey as of 1992 of the various methods of modeling motion by weighted mean curvature appears in [32], and that survey will not be repeated here. However, there have been several new developments since that time.

A major development is the development of a general theory of evolving surfaces by $v = M(\mathbf{n})(\omega - \kappa_\gamma)$ for arbitrary γ and compatible M, in case where there are only two types of regions, and hence no triple junctions. It is called "flat flow" because the "flat norm" of Geometric Measure Theory is used to determine the distance between configurations. The method defines approximate flows by doing a sequence of minimizations with a given time step, and then proves that there exists a limit to the approximate flows as the time step goes to zero [33, 34]. In this theory, sharp interfaces evolve through singularities such as pinch-offs without difficulty.

This theory has been partially extended to multiple regions with triple junctions by Caraballo [35], for the motion law $v = \kappa_\gamma$. A more general framework, applying to the more general law $v = M(\mathbf{n})(\omega - \kappa_\gamma)$ has been developed, and results have been proved within this framework for motion of crystalline curves in the 2-dimensional plane [36]. *Crystalline* here means that the equilibrium crystal shape for the surface free energy is a polygon.

Computations have also been made with the various diffuse interface formulations, again usually only in the 2-dimensional plane. See other papers in this volume for examples. Additionally, Novick-Cohen has studied one case of triple junction motion in which some interfaces move by mean curvature and others by surface diffusion [37].

6. Summary

Some of the mathematical issues raised by triple junctions have been surveyed, and recent mathematical progress in modelling such junctions and their motion has been outlined. The contradictions produced by trying to model negative triple junction line energies in sharp interface models have been detailed, whereas the natural way in which such negative energies appear in spread-out junctions has been noted in several contexts. But much remains to be done. In particular, little about triple junctions in mathematical models for anisotropic surfaces in R^3 has been proved, and there is much to be done if one is to successfully create a complete mathematical model using sharp interfaces in which there are to be negative triple junctions line tensions.

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