Journal of Statistical Physics, Vol. 88, Nos. 1/2, 1997

Crumpling Self-Avoiding Surfaces

E. J. Janse van Rensburg[1]

Received April 9, 1996; final January 14, 1997

Self-avoiding plaquette surfaces with a folding or bending fugacity are believed to undergo a "crumpling" transition from a flaccid phase with branched polymer characteristics (corresponding to surfaces with a high degree of folding), to a "smooth" phase (corresponding to surfaces faith a low degree of folding). I develop rigorous techniques in order to bound the free energy of this model. In particular, the limiting free energy is proven to be positive for all positive values of the folding fugacity. In addition, the existence of a non-analyticity in the limiting free energy of a (nontrivial) subclass of surfaces is proven. This implies the existence of a phase transition in this model, which I conjecture to be from a "flaccid" to a "smooth" phase.

KEY WORDS: Plaquette surfaces; fugacity; flaccid phase; smooth phase; limiting free energy; phase transition.

1. INTRODUCTION

The study of lipid bilayers, microemulsions, membranes and vesicles in chemistry and biology has inspired several models of self-avoiding surfaces in statistical mechanics.[1,2,3] A particularly interesting model is the plaquette surface model, which is a model of self-avoiding surfaces in the hypercubic lattice, constructed by glueing unit squares (plaquettes) at their edges into a surface.[1,2] These surfaces are often weighted with respect to their volume or with respect to their curvature (or both), and are usually called *vesicles.*

The behaviour of vesicles in the absence of bending rigidity is thought to be well understood. In particular, if the pressure difference between the

[1] Department of Mathematics and Statistics, York University, North York, Ontario, M3J 1P3, Canada; e-mail: rensburg@mathstat.yorku.ca.

0022-4715/97/0700-0177$12.50/0

interior and the exterior of the vesicle is negative or zero, then the vesicle deflates, and is said to be in the "flaccid regime."[1,4,5,6] In this case, numerical work suggests that the vesicle behaves as a branched polymer, in the sense that all scaling exponents are those of lattice animals or trees[7] (see also refs. 8–12). On the other hand, a positive pressure difference between the interior and exterior of the vesicle inflates it into an "expanded regime"; the volume of the vesicle scales as the $\frac{3}{2}$-power of its area[1] (see also ref. 13).

These phases can be described as follows: Let $v_n(m)$ be the number of vesicles of area n and volume m per lattice site in the 3 dimensional lattice. The generating function for this model is

$$V(x, y) = \sum_{n, m} v_n(m)\, x^n y^m \tag{1.1}$$

where x and y are fugacities conjugate to the area and volume respectively. For every fixed y, the radius of convergence of $V(x, y)$, $x_c^P(y)$, describes a curve of non-analyticities in $V(x, y)$ in the (x, y)-plane, each associated with a critical limit of (1.1). Fisher *et al* (1991)[13] have shown that for $0 < x < x_c^P(1)$ and $y \to 1^-$, there is a line of essential singularities corresponding to first order transitions to the "expanded" or "inflated" phase (these are also called "droplet singularities", see refs. 14–16. On the other hand, if $y < 1$ and $x \nearrow x_c^P(y)$, then the vesicle collapses to a "flaccid phase" resembling a branched polymer. It is known that the phase boundary here behaves as $x_c^P(y) \sim y^{-1/4}$.[13] These lines of singularities are separated by a multicritical point at $y = 1$ and $x = x_c^P(1)$ in the phase diagram, which exhibits itself as a non-analyticity in $x_c^P(y)$. At this point vesicles of the same area are uniformly weighted in (1.1) and they resemble branched polymers (see ref. 7 for numerical evidence supporting this scenario).

In this paper I consider a model of self-avoiding surfaces with bending rigidity (or a curvature energy). The limiting phase of surfaces with high rigidity is believed to be a phase of smooth surfaces (such as disk-like or cube-like surfaces) which may be reminiscent of the inflated phase encountered in the case of positive osmotic pressure in vesicles (see ref. 2 for numerical evidence). In addition, there is evidence that at low rigidity one recovers a crumpled phase resembling branched polymers. It is conjectured that a multicritical point separates these phases. At this point the inflated regime goes though a "crumpling transition" to the "crumpled phase." There is some dispute in the literature whether this is a critical point, or whether there is a cross-over from one regime to the other, without critical behaviour, see for example refs. 2, 5, and 17.

In this paper I apply rigorous methods to surfaces with bending rigidity. My objective is to establish rigorous results describing the limiting free energy of surfaces with a bending rigidity. In Section 2 the combinatorial properties of self-avoiding surfaces with a prescribed number of folds or bends are studied. In particular, if $s_n(\leqslant \varepsilon n)$ is the number of surfaces with area n and at most εn folds, then $\lim_{n\to\infty}(\log s_n(\leqslant \varepsilon n)/n = \log \beta_0(\varepsilon)$ exists and is concave and continuous for $\varepsilon \in [0, 2]$. In Section 3 I consider the statistical mechanics of surfaces with bending rigidity. A fugacity conjugate to the number of folds is introduced and I prove the existence of a limiting free energy. The most interesting result in this section is a bound which shows that the limiting free energy is non-zero for every finite value of the folding fugacity, even though numerical simulations seem to indicate that it approaches very close to zero for finite values of the folding fugacity.[2] In Section 4 I consider a restricted model of surfaces. In this model small, isolated, "excitations" are suppressed. I prove that there is a transition at a finite value of the folding fugacity in this model. I conclude with some remarks in Section 5.

2. COMBINATORIAL PROPERTIES OF SURFACES

In this section the combinatorial behaviour of surfaces with a prescribed number of folds is studied (see 2.1 for definitions). I prove bounds on the number of surfaces, and these establish bounds in the asymptotic limit of increasing area. For related methods, see refs. 1, 13, 18–24.

2.1. Definitions

A *plaquette* is a unit square with corners (*vertices*) which have integer coordinates in the cubic lattice. The boundary of a plaquette is the union of 4 unit line segments called *edges*. The ε-neighbourhood of a vertex v is the open ball $B_\varepsilon(v) \in \mathscr{R}^3$ with centre v and radius ε. Let S be a non-empty set of plaquettes; then the vertex $v \in S$ is occupied <u>once</u> if the set $(S - v) \cap B_\varepsilon(v)$ is connected for arbitrarily small $\varepsilon > 0$. S is a *lattice surface* (1) if every vertex $v \in S$ is occupied once, (2) if every edge in S is incident with at most 2 plaquettes and (3) if S is connected. The *boundary* of S is the set of all edges incident with exactly one plaquette. If the boundary of S is empty, then S is *closed.* I shall focus the discussion on closed surfaces with the topology of a sphere: In other words, if S is composed of n plaquettes and contains v vertices, then by Euler's theorem, $v - n = 2$. A *fold* in S is an edge incident with 2 plaquettes which are at right angles to each other. The *volume* of a closed surface is the number of unit cubes in its interior. If a surface is composed of n plaquettes, then its *area* is n. Note that the maximum number of folds in a closed surface of area n is $2n$, since every plaquette is incident with 4 edges, and each edge is incident with 2 plaquettes.

The *bottom* and *top* plaquettes of a closed surface are the first and last plaquettes in an increasing lexicographic ordering of the plaquettes by the coordinates of their barycentres. If $\hat{\imath}$ is the first direction in the cubic lattice, then it can be shown that the top and bottom plaquettes are normal to $\hat{\imath}$.[18] [2]

2.2. Upper Bounds

Two surfaces are distinct if they cannot be made identical by a translation in the cubic lattice. Let $s_n(l)$, where $0 \leqslant l \leqslant 2n$, be the number of distinct surfaces with n plaquettes and l folds. Let $s_n = \sum_{l \geqslant 0} s_n(l)$ be the number of surfaces with area n.

An upper bound on s_n can be found as follows: I construct all possible closed surfaces by recursively adding plaquettes to a boundary. Put down a plaquette in the cubic lattice in one of three possible orientations, and label this plaquette with 1. Order the edges of this plaquette lexicographically with the coordinates of their midpoints, and append plaquette 2 to the lexicographic least labeled edge of plaquette 1, and then continue with the remaining edges of plaquette 1 by adding plaquettes 3, 4 and 5 in lexicographic order. Once j plaquettes have been added, let i be the smallest label such that plaquette i has an edge not paired with another plaquette. Order the unpaired edges of i lexicographically and append plaquettes $j+1, j+2,...,$ to these in order. Repeat this process until n plaquettes have been labeled. At each step in the construction there is a unique edge to append the next plaquette, which may be added in one of three possible orientations. Thus, the maximum number of surfaces that one may construct in this way is 3^n. (Note that only a subclass of surfaces, which includes all closed surfaces, can be constructed in this way. This bound is cited in ref. 25 and is originally due to Ginibre *et al.*[26]).

Let σ be a closed surface. Label the bottom plaquette of σ with 1, and order its edges lexicographically. The plaquette incident with the least edge gets label 2, and the plaquette incident with the next least edge gets label 3, and so on. If j plaquettes have been labeled, and i is the least label so that plaquette i is incident with an unlabeled plaquette, then order the edges of i incident with unlabeled plaquettes lexicographically, and label the plaquette incident with the least edge with $j+1$, etc. This gives a canonical labeling for the plaquettes of σ, and gives the order whereby one

[2] The unit vectors in $\mathscr{Z}^3$ will be taken as $\hat{\imath}$, $\hat{\jmath}$ and $\hat{k}$. The *first* direction will be $\hat{\imath}$, the *second* $\hat{\jmath}$ and the *third* $\hat{k}$. A lexicographic ordering of points in $\mathscr{Z}^3$ will be with respect to these lattice directions in order, unless explicitly stated otherwise.

can add plaquettes starting at the bottom plaquette to construct σ as above. Hence

$$s_n \leqslant 3^n \tag{2.1}$$

I can find a bound on $s_n(l)$ by using the same process as above: If i is the smallest label with an unpaired edge, let $j+1$ be the next plaquette to be added incident to i. There is a choice between (1) adding plaquette $j+1$ at right-angles to i (in one of 2 ways), creating a fold, or (2) adding plaquette $j+1$ with the same orientation as i. This process is repeated at most n times, and one can create a fold at k of these in $\binom{n}{k}$ ways. The maximum number of ways that a surface can be created in this way is $\binom{n}{k} 2^k$. But note that a surface with l folds can be created by choosing $k \leqslant l$ from the n plaquettes for creating a fold. Thus

$$s_n(l) \leqslant \sum_{k \leqslant 1} \binom{n}{k} 2^k \tag{2.2}$$

Observe that this bound equals 3^n if $l = n$, thus the best bound on $s_n(l)$ if $l \geqslant n$ is given by (2.1).

2.3. Concatenation and the Limiting Behaviour of s_n

Suppose that σ and τ are two closed surfaces, and let the top plaquette of σ be t_σ and the bottom plaquette of τ be b_τ. Translate σ such that $t_\sigma + \hat{\imath} = b_\tau$, (the top and bottom plaquettes are always normal to the unit vector $\hat{\imath}$). A new surface can be constructed from τ and σ by deleting t_σ and b_τ, and then adding 4 plaquettes as illustrated in Fig. 1 to connect σ and τ into a closed surface $\sigma \oplus \tau$. If σ has area n and τ has area m, then $\sigma \oplus \tau$ has area $n + m + 2$. Note that at least 2, and at most 4, of the edges of any bottom or top plaquette are folds. Some of these may disappear when the concatenation is performed, and new folds may be created.

A case analysis shows that the total number of folds may change by 0, ± 2 or ± 4 when two closed surfaces are concatenated. (There are 9 cases in total, since the number of folds on the top and bottom plaquettes may take values 2, 3 or 4. For example, if the number of folds in the edges of t_σ is 2, and in b_τ is 3, then the concatenation removes these folds, but create 2 new folds in the edges of t_σ and 1 new fold in the edges of b_τ. Together with the folds in the 4 new plaquettes, this gives a change of $+2$ in the total number of folds. The other cases can be checked similarly.) This construction implies that the number of surfaces are supermultiplicative: If a closed surface with area n and k folds, is concatenated with a closed surface

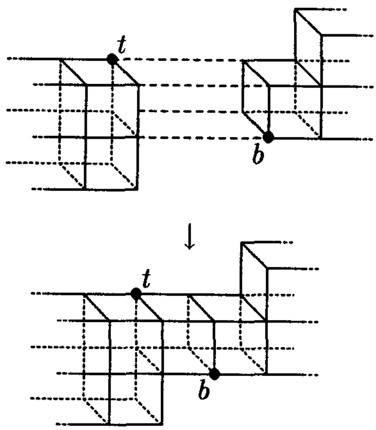

Fig. 1. Concatenation of two surfaces.

with area m and l folds, then a unique closed surface, with area $n+m+2$ and at least $k+l-4$ or at most $k+l+4$ folds, is the result. Hence

$$s_n(k)\,s_m(l) \leqslant \sum_{j=-2}^{2} s_{n+m+2}(k+l+2j) \tag{2.3}$$

$$s_n s_m \leqslant s_{n+m+2} \tag{2.4}$$

Since s_n is bounded by an exponential in (2.1) there exists a constant $\beta_0 > 0$ such that

$$\log \beta_0 = \lim_{n \to \infty} \frac{1}{n} \log s_n \tag{2.5}$$

and moreover, $s_n \leqslant \beta_0^{n+2}$.[27, 28] β_0 is called a *growth constant*, and by (2.1) $\beta_0 \leqslant 3$ (see ref. 25). A numerical estimate for the growth constant is $\beta_0 = 1.733 \pm 0.006$.[8–10]

2.4. Crumpled Surfaces

The number of folds in a surface is the *degree of crumpling* of that surface. Let $s_n(\leqslant l) = \sum_{k \leqslant l} s_n(k)$ be the number of surfaces with *at most l* folds. Then $s_n(\leqslant \varepsilon n)$ is the number of surfaces with at most εn folds, where ε can take values in (0,2]. Lower bounds on $s_n(\leqslant \varepsilon n)$ can be obtained for infinitely many values of n in the following proposition:

Proposition 1. (1) For any $m \geqslant 1$ and every $\varepsilon \in (0, 2]$, there exists a finite positive integer $q_0(\varepsilon)$ such that for every fixed $q > q_0(\varepsilon)$,

$$s_{6mq^2-2(m-1)}(\leqslant \sigma[6mq^2 - 2(m-1)]) \geqslant 4^{m-1}$$

(2) For every positive $\varepsilon < \frac{3}{2}$

$$s_{6q^2+4\lfloor \varepsilon q^2 \rfloor}(\leqslant 12(q + \varepsilon q^2)) \geqslant \binom{6\lfloor (q-2)^2/4 \rfloor}{\lfloor \varepsilon q^2 \rfloor}$$

Proof. (1) I construct a family of surfaces of area $n = (6mq^2 - 2(m-1))$, and with fewer than εn folds. A q-cube is a surface of area $6q^2$, and with the geometry of a cube with side-length q in 3 dimensions. The degree of folding in a q-cube is $12q$. A top plaquette t^{++} and a bottom plaquette b^{++} of a q-cube can be found by a lexicographic ordering of the barycentres of the plaquettes with respect to the directions $(\hat{\imath}, \hat{\jmath}, \hat{k})$. Similarly, if the lexicographic ordering is done with respect to the directions $(\hat{\imath}, -\hat{\jmath}, -\hat{k})$, then the top and bottom plaquettes t^{--} and b^{--} are found instead. Lastly, if the ordering is done with respect to $(\hat{\imath}, \hat{\jmath}, -\hat{k})$, then the top and bottom plaquettes are t^{+-} and b^{+-}, and if the ordering is done with respect to $(\hat{\imath}, -\hat{\jmath}, \hat{k})$, then t^{-+} and b^{-+} are the top and bottom plaquettes. These top plaquettes (and the bottom plaquettes) are distinct if $q > 1$. Two q-cubes can be "stringed together" (if $q > 1$) by *identifying* either t^{++} on the first with b^{++} on the second, or t^{--} on the first with b^{--} on the second. Alternatively, one can identify t^{+-} with b^{+-}, or t^{-+} with b^{-+}. If m such q-cubes are stringed together, then there are 4^{m-1} possible conformations. Each identification deletes two plaquettes from the q-cubes, but the number of folds are preserved. The total area is $6mq^2 - 2(m-1)$, and the total degree of folding is $12mq$. Thus, $s_{6mq^2-2(m-1)}(12mq) \geqslant 4^{m-1}$. Now increase q (if necessary), until $\varepsilon > 12q/6q^2 - 2$. Since $12q/6q^2 - 2 \geqslant 12mq/6mq^2 - 2(m-1)$ for any $m \geqslant 1$, this value of q is sufficient (put $q_0(\varepsilon) = \lceil 1/\varepsilon + \sqrt{1/\varepsilon^2 + 1/2} \rceil$).

(2) On the other handed a 1-cube can be "fused" on the outside of a q-cube by identifying a plaquette on the 1-cube with a plaquette in the q-cube, and then deleting the plaquette (as illustrated in Fig. 2). Perform this construction by selecting l plaquettes disjoint with folds and with each other in the q-cube. This can be done in at least $\binom{6\lfloor (q-2)^2/4 \rfloor}{l}$ ways. By counting the number of folds and plaquettes, the resulting surfaces have degree of folding $12q + 12l$, and area $6q^2 + 4l$. Thus, $s_{6q^2+4l}(\leqslant (12(q+l))) \geqslant \binom{6\lfloor (q-2)^2/4 \rfloor}{l}$. Now put $l = \lfloor \varepsilon q^2 \rfloor$, and the result follows. ∎

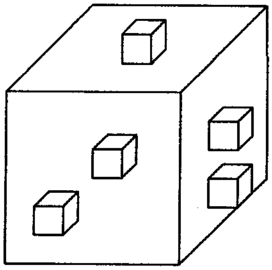

Fig. 2. A q-cube with 1-cubes fused to its outer surface.

The construction of concatenating surfaces preceding and leading to Eq. (2.3) shows that

$$s_n(\leqslant k)\, s_m(\leqslant l) \leqslant \sum_{j=-2}^{2} s_{n+m+2}(\leqslant (k+l+2j)) \tag{2.6}$$

The maximum is obtained if $j=2$ in the sum above; this gives the inequality

$$s_n(\leqslant k)\, s_m(\leqslant l) \leqslant 5 s_{n+m+2}(\leqslant (k+l+4)) \tag{2.7}$$

If $k=\varepsilon n$ and $l=\delta m$, then (2.7) becomes

$$s_n(\leqslant \varepsilon n)\, s_m(\leqslant \delta m) \leqslant 5 s_{n+m+2}(\leqslant (\varepsilon n+\delta m+4)) \tag{2.8}$$

and by putting $\varepsilon=\delta$ it shows that $\frac{1}{5} s_{n+2}(\leqslant (\varepsilon n-4))$ is a supermultiplicative function of the kind considered by Wilker and Whittington (1979).[28] Together with the bound in (2.1) this gives:

Proposition 2. There exists a function $\beta_0(\varepsilon)$ defined for every $\varepsilon \in (0, 2]$ such that

$$\lim_{n\to\infty} \frac{1}{n} \log s_n(\leqslant \varepsilon n) = \log \beta_0(\varepsilon)$$

If we let $n=m$ in Eq. (2.8), then

$$s_n(\leqslant \varepsilon n)\, s_n(\leqslant \delta n) \leqslant 5 s_{2n+2}(\leqslant (\varepsilon+\delta)\, n+2) \tag{2.9}$$

Take the logarithm of (2.9), divide by n, and let $n \to \infty$; this proves that

$$\log \beta_0(\varepsilon) + \log \beta_0(\delta) \leqslant 2 \log \beta_0 \left(\frac{(\varepsilon + \delta)}{2} \right) \tag{2.10}$$

Thus $\log \beta_0(\varepsilon)$ is a concave function of ε, and is continuous in $(0, 2)$. $\beta_0(\varepsilon)$ is defined for $\varepsilon \in (0, 2]$, but its domain can be extended to $[0, 2]$ by defining $\beta_0(0) = \lim_{\varepsilon \to 0^+} \beta_0(\varepsilon)$. The following inequality is proven in ref. 21, and it will be useful in the proof of Proposition 3: Let q be a finite positive real number, then if $0 \leqslant \gamma \leqslant q/q+1$,

$$\sum_{k=0}^{\lfloor \gamma n \rfloor} \binom{n}{k} q^k \leqslant \left[\frac{q^\gamma}{\gamma^\gamma (1-\gamma)^{1-\gamma}} \right]^n \tag{2.11}$$

The following proposition is a summary of the properties of $\beta_0(\varepsilon)$:

Proposition 3. $\log \beta_0(\varepsilon)$ is a concave function of ε, and is continuous for $\varepsilon \in [0, 2]$. Moreover, $\beta_0(2) = \beta_0$ and $\lim_{\varepsilon \to 0^+} \beta_0(\varepsilon) = \beta_0(0) = 1$.

Proof. By Eq. (2.10), $\log \beta_0(\varepsilon)$ is concave in $(0, 2]$, and thus continuous in $(0, 2)$. To prove continuity at $\varepsilon = 2$, note that Eqs. (2.8), (2.9) and (2.10) are valid even if $\varepsilon \geqslant 2$. Thus $\beta_0(\varepsilon)$ is continuous at $\varepsilon = 2$ (and obviously, $\beta_0(2) = \beta_0$). Thus $\beta_0(\varepsilon)$ is continuous in $(0, 2]$. It remains to be shown that $\lim_{\varepsilon \to 0^+} \beta_0(\varepsilon) = 1$. The limit exists, since $\beta_0(\varepsilon)$ is a monotone function in $(0, 2]$. By Proposition 1(1), for every $\varepsilon > 0$ there exists a fixed number $q > 0$ such that $s_n(\leqslant \varepsilon n) \geqslant 4^{(n-2)/(6q^2-2)} > 1$ for infinitely many values of n. Thus $\lim_{\varepsilon \to 0^+} \beta_0(\varepsilon) \geqslant 1$. Otherwise, use Eq. (2.2): $s_n(\leqslant \varepsilon n) \leqslant \sum_{l \leqslant \varepsilon n} \sum_{k \leqslant l} \binom{n}{k} 2^k$, then for small ε ($\varepsilon \leqslant \frac{1}{2}$ is sufficient), the sum over k can be bounded from above by $(l+1)\binom{n}{l} 2^l$, since the maximum is obtained by putting $k = l$. Thus $s_n(\leqslant \varepsilon n) \leqslant \sum_{l \leqslant \varepsilon n} (l+1)\binom{n}{l} 2^l \leqslant (\varepsilon n + 1) \sum_{l \leqslant \varepsilon n} \binom{n}{l} 2^l$. The sum over l can be bounded from above by using (2.11):

$$s_n(\leqslant \varepsilon n) \leqslant (\varepsilon n + 1) \left[\frac{2^\varepsilon}{\varepsilon^\varepsilon (1-\varepsilon)^{1-\varepsilon}} \right]^n$$

provided that $0 \leqslant \varepsilon \leqslant \frac{2}{3}$, so that $\beta_0(\varepsilon) \leqslant 2^\varepsilon / \varepsilon^\varepsilon (1-\varepsilon)^{1-\varepsilon}$. As one takes $\varepsilon \to 0^+$, the result is $\lim_{\varepsilon \to 0^+} \beta_0(\varepsilon) \leqslant 1$. Now define $\beta_0(0) = 1 = \lim_{\varepsilon \to 0^+} \beta_0(\varepsilon)$. This proves the proposition. ∎

Remarks. By Proposition 1, $\beta_0(2) > \beta_0(0) = 1$. Since $\beta_0(\varepsilon)$ is continuous and non-decreasing for $\varepsilon \in [0, 2]$, there exists an $\varepsilon_c \in (0, 2]$ such that $\beta_0(\varepsilon) = \beta_0(2)$ $\forall \varepsilon \geqslant \varepsilon_c$. I plot the expected behaviour of $\beta_0(\varepsilon)$ in Fig. 3. Bounds on $\beta_0(\varepsilon)$ for small ε can be derived from Proposition 1(2), and from the proof of

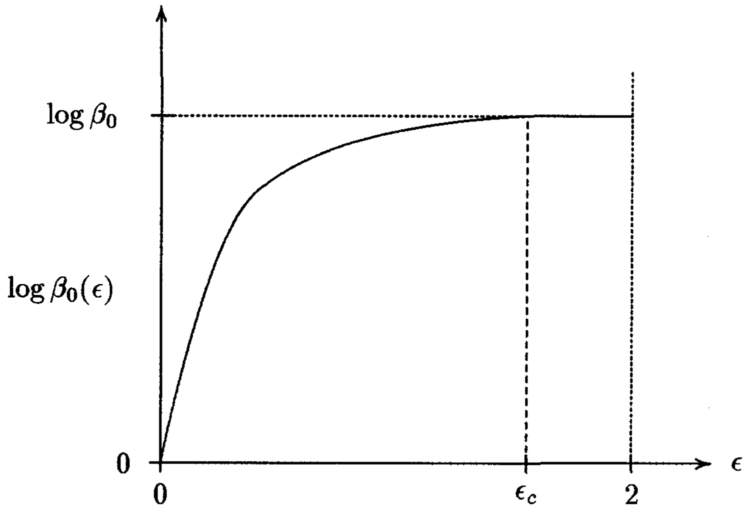

Fig. 3. The expected behaviour of $\log \beta_0(\varepsilon)$.

Proposition 3: By Proposition 1(2) $\lim_{q\to\infty} 1/6q^2 \log s_{6q^2+4\lfloor \varepsilon q^2\rfloor}(\leqslant 12(q+\varepsilon q^2)) \geqslant \lim_{q\to\infty} 1/6q^2 \log\binom{6\lfloor (q-2)^2/4\rfloor}{\lfloor \varepsilon q^2\rfloor}$ implies that (provided that $\varepsilon < \frac{3}{2}$)

$$[\beta_0(2\varepsilon)]^2 \geqslant \left[\beta_0\left(\frac{2\varepsilon}{1+2/3\varepsilon}\right)\right]^{1+2/3\varepsilon} \geqslant \frac{1/4^{1/4}}{\varepsilon/6^{\varepsilon/6}(1/4-\varepsilon/6)^{(1/4-\varepsilon/6)}}$$

Taken together with the upper bound derived in the proof of Proposition 3,

$$\left[\frac{27}{\varepsilon^\varepsilon(3-\varepsilon)^{(3-\varepsilon)}}\right]^{1/24} \leqslant \beta_0(\varepsilon) \leqslant \frac{2^\varepsilon}{\varepsilon^\varepsilon(1-\varepsilon)^{1-\varepsilon}} \tag{2.12}$$

where the lower bound is valid if $\varepsilon \leqslant 2$, and the upper bound is valid if $\varepsilon < \frac{1}{2}$. An immediate consequence of these bounds is that (by the squeeze theorem for limits)

$$\left.\frac{d^+\beta_0(\varepsilon)}{d\varepsilon}\right|_{\varepsilon=0} = \lim_{\varepsilon\to 0^+} \frac{\beta_0(\varepsilon)-\beta_0(0)}{\varepsilon} = \infty \tag{2.13}$$

Thus the graph of $\log \beta_0(\varepsilon)$ in Fig. 3 approaches the point $(0, 0)$ with infinite gradient.

3. THE STATISTICAL MECHANICS OF CRUMPLING SELF-AVOIDING SURFACES

A model of "crumpling surfaces" is defined by the introduction of a "folding fugacity" z conjugate to the degree of folding. This is the model which was considered by Whittington,[1] and Orlandini *et al.* (1995),[2] and one may consider it to be a discrete approximation to surfaces with a curvature energy. The main results in this section are a set of bounds on the free energy; these are derived using the results in Section 2. In particular, I prove that the free energy is always positive, for any finite value of the fugacity. This settles a question raised in the numerical study by Orlandini *et al.* (1995)[2] which suggests that the free energy approaches very close to zero for finite values of the fugacity.

3.1. Free Energies

The thermodynamic behaviour of a self-avoiding surface is completely described by its free energy. Let

$$G_n(z) = \sum_{l=0}^{2n} s_n(l)\, z^l \tag{3.1}$$

be the generating function of surfaces of area n, with fugacity z conjugate to the degree of folding in the surfaces. (This is the *canonical* partition function or the "folding generating function"). The free energy per plaquette, $F_n(z)$, is defined by

$$F_n(z) = \frac{1}{n} \log G_n(z) \tag{3.2}$$

The existence of a limiting free energy (as $n \to \infty$) was proven by Whittington;[1] this is a direct consequence of the generalised supermultiplicative relation $\sum_{k=0}^{l} s_n(l-k)\, s_m(k) \leqslant \sum_{j=-2}^{2} s_{n+m+2}(l+2j)$, which one obtains in the same way as (2.6).

Proposition 4 (Whittington, 1993[1]). There exists a function $\mathscr{F}(z)$ for every $z \in [0, \infty)$ such that

$$\mathscr{F}(z) = \lim_{n \to \infty} F_n(z) = \lim_{n \to \infty} \frac{1}{n} \log G_n(z)$$

Moreover $G_n(z) \leqslant (\sum_{j=-2}^{2} z^{2j})[\mathscr{F}(z)]^{n+2}$, and $\mathscr{F}(z)$ is a convex function of $\log z$ and therefore continuous for $z \in (0, \infty)$.

I will also show that $\mathcal{F}(0^+) = 0$ at the end of this section. A second generating function for this model is the so-called "grand canonical partition function," which is defined as

$$G(x, z) = \sum_{n=0}^{\infty} \sum_{l=0}^{2n} s_n(l)\, z^l x^n = \sum_{n=0}^{\infty} G_n(z)\, x^n \tag{3.3}$$

If one compares (3.2) and Proposition 4 with (3.3), then $e^{-\mathcal{F}(z)} = x_c(z)$ is the radius of convergence of the infinite series in x defining $G(x, z)$. Thus, one merely needs to study $x_c(z)$ in order to derive properties of $\mathcal{F}(z)$. Under some circumstances this will prove more suitable than working directly with the limiting free energy, but in other cases. I shall work directly with the limiting free energy $\mathcal{F}(z)$.

3.2. Bounds on $\mathcal{F}(z)$ if $z \geqslant 1$

An upper bound is obtained with relative ease: Note that $G_n(z) = \sum_l s_n(l)\, z^l \leqslant s_n \sum_{l=0}^{2n} z^l \leqslant 2n s_n z^{2n}$ since $z \geqslant 1$. By taking the logarithm of this inequality, dividing by n and letting $n \to \infty$, (and using (2.5))

$$\begin{aligned} &\mathcal{F}(z) \leqslant \log \beta_0 + 2 \log z, \qquad \text{provided that } z \geqslant 1 \\ &\mathcal{F}(1) = \log \beta_0 \end{aligned} \tag{3.4}$$

A lower bound is more difficult. In this case one can construct a subset of surfaces by using "blocks" such as illustrated in Fig. 4.

The block in Fig. 4 has width equal to 5, area $n = 78$ and degree of folding $l = 2n = 156$. In general, if the width of a block is p (where p is odd), then it has area $3(p^2 + 1)$, and degree of folding $6(p^2 + 1)$. One can join these blocks together, as illustrated in Fig. 5, by identifying rightmost and

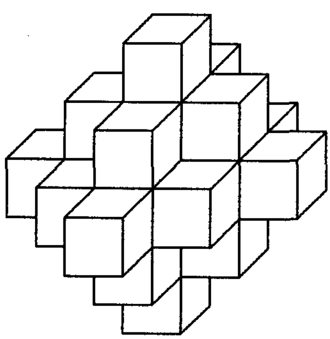

Fig. 4. A block with area $n = 78$ and $l = 2n = 156$ folds.

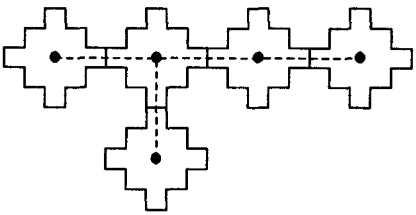

Fig. 5. A cross-section of a site tree generated by glueing together the blocks in Fig. 4. The vertices are separated by 5 units.

leftmost plaquettes. The number of ways that this can be done is related to the number of site trees in a sublattice of $\mathscr{Z}^3$ as follows: Let the midpoint of each block be a vertex in a site tree on the dual lattice of $\mathscr{Z}^3$, and let two vertices be adjacent if the corresponding blocks are joined. The length of the edge between these vertices is p, and the number of different ways in which q blocks can be joined into a surface is t_q, the number of site trees in $\mathscr{Z}^3$ with q vertices. Each time that two blocks are joined, 2 plaquettes are lost, as well as 8 folds. Thus, in a surface built from q blocks, the area is $n = 3(p^2+1)\,q - 2(q-1) = (3p^2+1)\,q+2$ and degree of folding $l = 6(p^2+1)\,q - 8(q-1) = (6p^2-2)\,q+8$. Hence, $G_{(3p^2+1)\,q+2}(z) \geqslant t_q z^{(6p^2-2)\,q+8}$. Fix p and let $q \to \infty$. Then $n \to \infty$,

$$\mathscr{F}(z) \geqslant \lim_{q\to\infty} \left(\frac{1}{(3p^2+1)\,q+2} \log t_q + \frac{(6p^2-2)\,q+8}{(3p^2+1)\,q+2} \log z \right) \tag{3.5}$$

The number of site trees in three dimensions is known to be $t_q = \Lambda_s^{q+o(q)}$ (see ref. 29 for a proof of this in 2 dimensions; this can be readily adapted to 3 dimensions), where $\Lambda_s \geqslant 3$ is the growth constant of site trees. (This bound is seen by noting that the number of self-avoiding walks, with steps only in the positive lattice directions, grows as 3^n). Consequently, from (3.5):

$$\mathscr{F}(z) \geqslant \frac{1}{3p^2+1} \log \Lambda_s + \frac{6p^2-2}{3p^2+1} \log z, \qquad \text{for odd } p \geqslant 1 \text{ provided that } z \geqslant 1 \tag{3.6}$$

If $p=1$ in (3.6), then $\mathscr{F}(z) \geqslant \log 3/4 + \log z$, and if $p \to \infty$ in (3.6), then $\mathscr{F}(z) \geqslant 2 \log z$.[3] No improvement is gained from other values of p; one

[3] Note that $p=1$ corresponds to a "deflation" of the surfaces, and that the surfaces "inflate" as $p \to \infty$. The bound $(\log 3/4) + \log z$ can be improved with better lower bounds on Λ_s.

can show that all the lines defined by $y = (\log 3/3p^2 + 1) + ((6p^2 - 2)/(3p^2 + 1)) \log z$ intersect in the point $(\log 3/4, 2 \log 3/4)$. Thus

$$\mathscr{F}(z) \geqslant \begin{cases} \dfrac{\log 3}{4} + \log z, & \text{if} \quad 0 \leqslant \log z \leqslant \dfrac{\log 3}{4} \\ 2 \log z, & \text{if} \quad \log z \geqslant \dfrac{\log 3}{4} \end{cases} \tag{3.7}$$

3.3. Bounds on $\mathscr{F}(z)$ if $z \leqslant 1$

Since $G_n(z)$ is a monotonic increasing function with z, (3.5) implies that

$$\mathscr{F}(z) \leqslant \mathscr{F}(1) = \log \beta_0 \qquad \forall z \leqslant 1 \tag{3.8}$$

Consider $x_c(z)$, the radius of convergence of the generating function $G(x, z)$ (Eq. (3.3)). The following proposition gives upper bounds on $x_c(z)$:

Proposition 5. For every $z \in [0, 1]$:

$$x_c(z) \leqslant \min\{1, \beta_0^{-1} z^{-2}\}$$

and thus $x_c(z) < 1$ if $\sqrt{\beta_0}^{-1} \leqslant z \leqslant 1$.

Proof. Note that $G(x, z) \geqslant \sum_n \sum_{l \geqslant 0}^{\varepsilon n} s_n(l)\, z^l x^n \geqslant \sum_n \sum_{l \geqslant 0}^{\varepsilon n} s_n(l)\, z^{\varepsilon n} x^n \geqslant \sum_n s_n(\leqslant \varepsilon n)\, z^{\varepsilon n} x^n$. But by Proposition 2, $s_n(\leqslant \varepsilon n) = [\beta_0(\varepsilon)]^{n + o(n)}$, so $G(x, z) \geqslant \sum_n [\beta_0(\varepsilon)]^{n + o(n)} z^{\varepsilon n} x^n$. The radius of convergence of the last series is an upper bound on $x_c(z)$, thus

$$x_c(z) \leqslant [\beta_0(\varepsilon)]^{-1} z^{-\varepsilon}$$

Now take $\varepsilon \to 0$ to obtain $x_c(z) \leqslant 1$. Alternatively, bound $G(x, z)$ from below as follows: $G(x, z) \geqslant \sum_n \sum_{l \geqslant 0}^{2n} s_n(l)\, z^{2n} x^n$, since $z \leqslant 1$. Thus $G(x, z) \geqslant \sum_n s_n z^{2n} x^n$, and the radius of convergence of this last series is likewise an upper bound on $x_c(z)$. By Eq. (2.5)

$$x_c(z) \leqslant \beta_0^{-1} z^{-2}$$

This bound is better that the first if $\sqrt{\beta_0}^{1} \leqslant z \leqslant 1$, otherwise the first is better. ∎

Does $x_c(z)$ ever become equal to 1 for <u>finite</u> z? Since it is a monotonic non-increasing function this will imply that it (and thus $\mathscr{F}(z)$) has a non-analyticity. This non-analyticity would signal a phase transition between the "flaccid" (branched polymer) regime, and a phase of "smooth" surfaces (where the generating function $G(x, z)$ is dominated by surfaces with low degree of folding). This in fact does not happen: I prove in Proposition 6 that $x_c(z) < 1$ for all z. This does not rule out a non-analyticity in $\mathscr{F}(z)$. The proof for this depends on the lower bound in equation (2.12), which was derived by considering the surfaces illustrated in Fig. 2.

Proposition 6. For every positive $z \leqslant 1$, $x_c(z) \leqslant e^{-z^{24}/8e} < 1$.

Proof. Let $z \leqslant 1$. Observe that $G(x, z) \geqslant \sum_{n=0}^{\infty} \sum_{l \leqslant \varepsilon n} s_n(l) z^l x^n \geqslant \sum_{n=0}^{\infty} s_n(\leqslant \varepsilon n) z^{\varepsilon n} x^n = \sum_{n=0}^{\infty} [\beta_0(\varepsilon)]^{n+o(n)} z^{\varepsilon n} x^n$. Thus (by using the bound in (2.12))

$$G(x, z) \geqslant \sum_{n=0}^{\infty} \left[\frac{27}{\varepsilon^{\varepsilon}(3-\varepsilon)^{(3-\varepsilon)}}\right]^{n/24+o(n)} z^{\varepsilon n} x^n$$

if $\varepsilon \leqslant 2$. The factor in the square brackets above is bounded from below by $3/\varepsilon$. Hence

$$x_c(z) \leqslant \left(\frac{\varepsilon/3}{z^{24}}\right)^{\varepsilon/24} < 1 \qquad \text{if} \quad \varepsilon < 3z^{24}$$

Thus, for every positive $z \leqslant 1$ there exists such an $\varepsilon > 0$. The minimum upper bound is derived by taking $d/d\varepsilon(\varepsilon/24 \log(\varepsilon/3/z^{24})) = \frac{1}{24} + \frac{1}{24} \log(\varepsilon/3/z^{24}) = 0$. This gives the upper bound $e^{-z^{24}/8e}$ on $x_c(z)$, for $z \in [0, 1]$. ∎

The bounds in Propositions 5 and 6 translate into lower bounds on $\mathscr{F}(z)$.

$$\mathscr{F}(z) \geqslant \begin{cases} \log \beta_0 + 2 \log z & \text{if} \quad -0.275 \cdots \leqslant \log z \leqslant 0 \\ \dfrac{z^{24}}{8e} & \text{if} \quad \log z \leqslant -0.275 \cdots \end{cases} \tag{3.9}$$

The two lower bounds on $\mathscr{F}(z)$ are equal if $\log z \approx -0.275$; if $\log z$ is larger than this number, then the first bound is better, otherwise the second bound is better.

A lower bound on $x_c(z)$ is found by writing $G(x, z)$ as the sum of two terms which will be bounded from above:

$$\begin{aligned} G(x, z) &= \sum_n \sum_{l=0}^{2n} s_n(l) z^l x^n = \sum_n \sum_{l \leqslant \varepsilon n} s_n(l) z^l x^n + \sum_n \sum_{l > \varepsilon n} s_n(l) z^l x^n \\ &\leqslant \sum_n \sum_{l \leqslant \varepsilon n} \left(\sum_{k \leqslant l} \binom{n}{k} 2^k\right) z^l x^n + \sum_n \sum_{l > \varepsilon n} s_n(l) z^l x^n \end{aligned} \tag{3.10}$$

where (2.2) was used, and where I will put $\varepsilon = \frac{1}{2}$. The following bound will prove useful:

Lemma 7. Let $n > 1$ and l be integers such that $0 < l < n$. Then

$$\frac{n^n}{l^l(n-l)^{(n-l)}} \leqslant \frac{72\sqrt{2\pi}}{110}\sqrt{n}\binom{n}{l}$$

Proof. Use the Stirling approximation to $k!$: For every $k \geqslant 1$ (see for example Buck (1965)[30]):

$$\left|\frac{k!}{k^k e^{-k}\sqrt{2\pi k}} - 1\right| \leqslant \frac{1}{11k}$$

Hence

$$\frac{11}{12}\frac{e^k}{\sqrt{2\pi k}} \leqslant \frac{k^k}{k!} \leqslant \frac{11}{10}\frac{e^k}{\sqrt{2\pi k}}$$

If these bounds on k^k are substituted in $n^n/l^l(n-l)^{(n-l)}$ for $k = n$, l and $(n-l)$, then the upper bound is proven. ∎

The second term in (3.10) is bounded as follows:

$$\sum_n \sum_{l > \lfloor n/2 \rfloor}^{2n} s_n(l)\, z^l x^n \leqslant \sum_n \left(2n - \left\lfloor \frac{n}{2} \right\rfloor\right) 3^n z^{n/2} x^n \qquad \text{by (2.1)} \qquad (3.11)$$

which is finite if $x < 1/3\sqrt{z}$. The first term requires somewhat more work; and the use of Lemma 7:

$$\begin{aligned}
&\sum_n \sum_{l=0}^{\lfloor n/2 \rfloor} \left(\sum_{k \leqslant l} \binom{n}{k} 2^k\right) z^l x^n \\
&\quad \leqslant \sum_n \sum_{l=0}^{\lfloor n/2 \rfloor} \left(\frac{2^{l/n}}{(l/n)^{l/n}(1-l/n)^{(1-l/n)}}\right)^n z^l x^n, \qquad \text{since } l \leqslant \frac{n}{2}, \text{ and by (2.11)} \\
&\quad = \sum_n \sum_{l=0}^{\lfloor n/2 \rfloor} \frac{n^n}{l^l(n-l)^{(n-l)}} (2z)^l x^n \\
&\quad \leqslant \sum_n \frac{72\sqrt{2\pi}}{110}\sqrt{n} \sum_{l=0}^{\lfloor n/2 \rfloor} \binom{n}{l} (2z)^l x^n, \qquad \text{by Lemma 7} \\
&\quad \leqslant \frac{72\sqrt{2\pi}}{110} \sum_n \sqrt{n}\,(1+2z)^n x^n \qquad (3.12)
\end{aligned}$$

which is finite if $x < 1/1 + 2z$. Thus, by comparing (3.11) and (3.12), the following lower bound on $x_c(z)$ is obtained:

Proposition 8. $x_c(z)$ is bounded from below in $[0, 1]$ as

$$x_c(z) \geqslant \begin{cases} \dfrac{1}{3\sqrt{z}}, & \text{if} \quad z \in \left[\dfrac{1}{4}, 1\right] \\ \dfrac{1}{1+2z}, & \text{if} \quad z \in \left[0, \dfrac{1}{4}\right] \end{cases}$$

Hence;

$$\mathscr{F}(z) \leqslant \begin{cases} \log 3 + \frac{1}{2} \log z, & \text{if} \quad \frac{1}{4} \leqslant z \leqslant 1 \\ \log(1+2z), & \text{if} \quad 0 \leqslant z \leqslant \frac{1}{4} \end{cases} \tag{3.13}$$

3.4. Remarks

The bounds derived in this section are plotted in Fig. 6 against $\log z$. Note that $\mathscr{F}(z)$ is asymptotic to $C_0 + 2 \log z$ if $z > 1$, where C_0 is a constant between 0 and $\log \beta_0 \approx 0.550$. For $\log z < 0$, it is asymptotic to 0. These results have implications for the average degree of folding in the $n \to \infty$ limit: This is defined as $\lim_{n \to \infty} \langle l \rangle / n = \lim_{n \to \infty} d/(d \log z)(1/n) \log G_n(z)$ (if this limit exists). Since $(1/n) \log G_n(z)$ is a sequence of convex functions (Proposition 4), and the limit of this sequence is a convex function $\mathscr{F}(z)$, it follows that $(d^-/d \log z)\, \mathscr{F}(z) \leqslant \liminf_{n \to \infty} (d^-/d \log z)(1/n) \log G_n(z) \leqslant \limsup_{n \to \infty} (d^+/d \log z)(1/n) \log G_n(z) \leqslant (d^+/d \log z)\, \mathscr{F}(z)$.[31] With increasing z, the left- and right-derivatives of $\mathscr{F}(z)$ approach 2, and the average degree of folding is 2 in the limit. Similarly, if z approaches 0, then the average degree of folding is 0 in the limit. (In other words, I can only show that the limit defining the average degree of folding exists if $z = 0$ or if $z \to \infty$.) Numerical simulations [2] suggest that $x_c(z) (= e^{-\mathscr{F}(z)})$ approaches very close to 1 for modest values of z. In particular, $x_c(0.635) = 0.987 \pm 0.010$. This is slightly smaller than the upper bound proved in Proposition 6, which predicts that $x_c(0.635) \leqslant 0.99999915\ldots$. This upper bound is due to inflated surfaces (those in Fig. 2) with isolated folds exploring a large smooth surface area. In this regime, it seems that the limiting theory is one of inflated surfaces, even though a limiting theory of "disk-like" surfaces was suggested by Orlandini *et al.* (1995).[2] For larger values of z the upper bound on $x_c(z)$ is given by (3.7), and these are due to the "tree-like" surfaces as in Fig. 5. Here it seems likely that the limiting theory is a theory of branched polymers. However, there is a qualification which must be taken into account: The bound for large values of z is

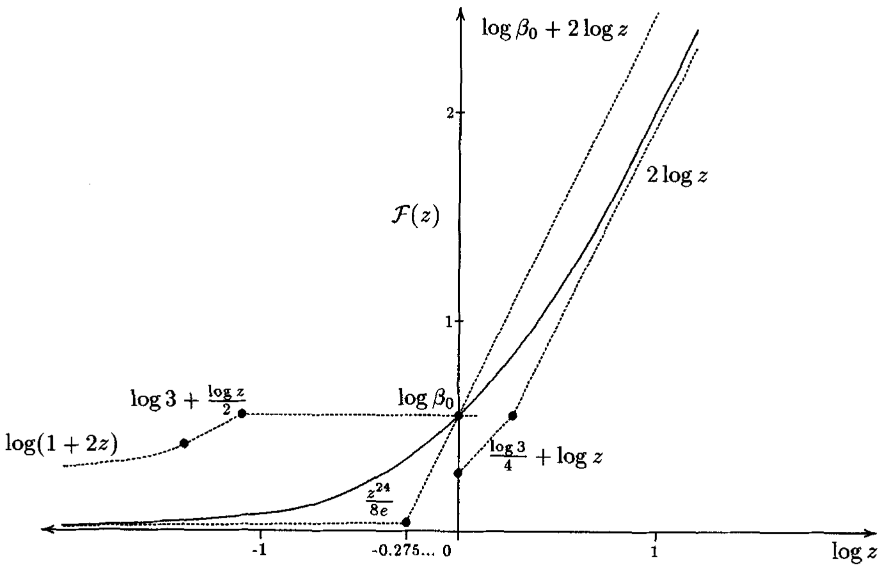

Fig. 6. $\mathcal{F}(z)$ as a function of $\log z$.

derived by taking $p \to \infty$ in (3.6), thus "inflating" the surfaces which are schematically illustrated in Fig. 5 (this is only done after the $n \to \infty$ limit is already taken). This inflation has no effect on the branched polymer character of the limiting theory. (Thus, the limiting theory, for large z, could be described as one of inflated, branched surfaces.) It is conjectured that these phases are separated by a multicritical point. The nature of the singularities on $G(x, z)$ (as $x \nearrow x_c(z)$) is not clear: A collapse to smooth surfaces at small values of z (as $x \nearrow x_c(z)$) may be expected to be a first order transition (in analogy with the results obtained for 2 dimensional vesicles by Fisher *et al.* 1991).[13] on the other hand, the collapse (for larger values of z) to the branched polymer phase is conjectured to be continuous. If this is indeed the case, then the multicritical point separating the two limiting phases is *tricritical* as suggested by Orlandini *et al.* (1995).[2] Lastly, note that $\lim_{z \to 0^+} \mathcal{F}(z) = 0$, and thus $\mathcal{F}(z)$ is continuous for $z \in [0, \infty)$, as promised in the first line after Proposition 4.

4. A CRUMPLING TRANSITION IN A RESTRICTED SET OF SURFACES

Let σ be a surface with area n and degree of folding L. The *skeleton* (or frame) of σ is the set of all edges which are folds in σ. In general, the skeleton of a surface is a set of lattice animals. Some surfaces have a connected skeleton, such as a q-cube, or the block in Fig. 4, but many surfaces have skeletons which are not connected, such as the example in Fig. 2. Let $s_n^C(l)$ be the number of surfaces with a *connected* skeleton, with area n and degree of folding l. By using the same constructions as in Section 2, one can show that

$$\lim_{n \to \infty} \frac{1}{n} \log s_n^C(\leqslant \varepsilon n) = \log \beta_0^C(\varepsilon), \qquad \text{for every} \quad \varepsilon \in [0, 2] \tag{4.1}$$

Moreover, $\log \beta_0^C(\varepsilon)$ is a concave function of ε, is continuous in $[0, 2]$, monotonic non-decreasing with ε and $\beta_0^C(0) = 1$ (this is seen from Eq. (2.12) and Proposition 1(1)). Also note that $\beta_0^C(2) \leqslant \beta_0(2)$. A lower bound on $\beta_0^C(\varepsilon)$, for small values of ε, cannot be derived from Proposition 1(2), since that involves surfaces with skeletons which are not connected. One can also improve on the upper bound in (2.12) as follows: observe that any surface with a connected skeleton and degree of folding l can be mapped into lattice animals weakly embedded in the cubic lattice, with l edges (to see this, remove all plaquettes from the surface, and just leave behind its skeleton). Suppose that α is a lattice animal with the property that every edge in α is in a planar (2 dimensional) polygon. Then one may attempt to convert a into a closed surface by filling in the planar polygons with planar sheets of plaquettes. About every edge in α there are potentially 4 possible directions for adding the sheet of plaquettes. If there are a_l animals with l edges, then this implies that

$$s_n^C(l) \leqslant 4^l a_l \tag{4.2}$$

where a_l is the number of lattice animals of size l. Consequently,

$$\beta_0^C(\varepsilon) \leqslant (4\lambda)^\varepsilon \tag{4.3}$$

where λ is the growth constant for lattice animals in the cubic lattice.[29] The function $\beta_0^C(\varepsilon)$ has many of the same properties as $\beta_0(\varepsilon)$, as noted above, but it differs in one important respect: it does not approach 1 as $\varepsilon \to 0$ with infinite slope, as implied by Eq. (2.13). In fact, it seems to approach 1 with <u>finite</u> slope as $\varepsilon \to 0$, since by (4.3) and since $\beta_0^C(\varepsilon)$ is monotonic non-decreasing

$$\begin{aligned} 0 &\leqslant \liminf_{\varepsilon \to 0^+} \frac{\log \beta_0^C(\varepsilon) - \log \beta_0^C(0)}{\varepsilon} \\ &\leqslant \limsup_{\varepsilon \to 0^+} \frac{\log \beta_0^C(\varepsilon) - \log \beta_0^C(0)}{\varepsilon} \leqslant \log(4\lambda) \end{aligned} \tag{4.4}$$

Thus, the right derivative exists and it is finite, as opposed to the situation described in Fig. 3 and Eq. (2.13).

4.1. Free energies

The generating functions for this model are:

$$G_n^C(z) = \sum_l s_n^C(l)\, z^l$$
$$G^C(x, z) = \sum_n G_n^C(z)\, x^n \tag{4.5}$$

The existence of a limiting free energy per plaquette, $\mathscr{F}^C(z)$, and its convexity and continuity follows by the same methods as in Section 3. Note that $\mathscr{F}^C(z) \leqslant \mathscr{F}(z)$, so that every upper bound on $\mathscr{F}(z)$ is an upper bound on $\mathscr{F}^C(z)$. Moreover, the lower bounds in (3.6) are also lower bounds on $\mathscr{F}^C(z)$, since the blocks in Fig. 4 and the surfaces in Fig. 5 have connected skeletons.

By Proposition 1(1) it is apparent that $\mathscr{F}^C(z) > 0$ if $z > 1$, since the class of surfaces constructed in that proposition have connected skeletons (this can also be seen from Eq. (3.6)). On the other hand, the following theorem is an immediate consequence of Theorem 10 in Section 5:

Theorem 9. $\mathscr{F}^C(z) = 0$ if $z \leqslant 1/4\lambda$.

4.2. Remarks

Let $z_C = \max\{z \mid \mathscr{F}^C(z) = 0\}$. Since $\mathscr{F}^C(z)$ is strictly positive if $z > 1$ there is a non-analyticity in $\mathscr{F}^C(z)$ at z_C. This non-analyticity signals a phase transition in this model. A schematic diagram of $x_c(z)$ is illustrated in Fig. 7.[4] The multicritical point at z_c separates two critical curves. The straight line for $z < z_c$ corresponds to surfaces with low degree of folding. If one argues as at the end of Section 2, then this is a phase of smooth surfaces, possibly inflated. On the other hand, the line of transitions for $z > z_c$ is a phase of branched polymers, (which could be inflated, as suggested by the results in Section 3). I conjecture that the multicritical point is tricritical. Lastly, observe that $(1/n) \log G_n^C(z)$ is a convex function of $\log z$ which converges to the convex function (of $\log z$) $\mathscr{F}^C(z)$ as $n \to \infty$. The "density

[4] This figure might be misleading, i have assumed that $x_c(z)$ has a continuous first derivative at z_c. This is of course not necessarily true, the derivative could be discontinuous (indicated by the dotted curve), as it was found in the case of 2 dimensional vesicles (fisher *et al.* 1991).[13]

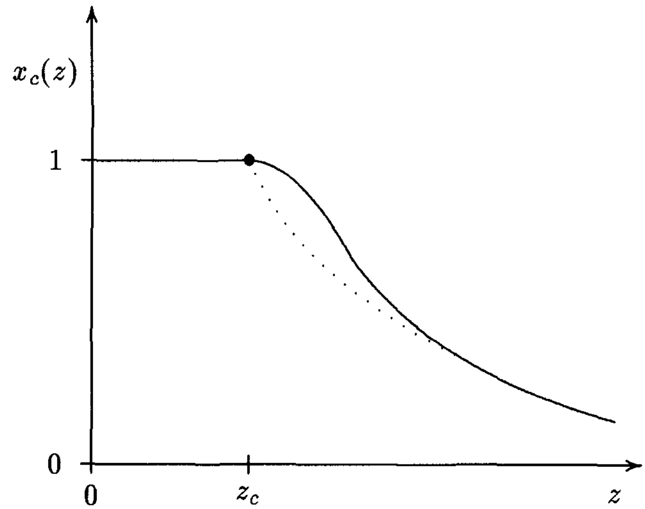

Fig. 7. The conjectured phase diagram of surfaces with connected skeletons.

of folds" can be defined as before by the derivative $\lim_{n\to\infty}\langle l\rangle/n = \lim_{n\to\infty} (d/d\log z)(1/n)\log G_n^C(z)$ (if it exists). As argued in the remarks in 3.4, it is apparent that if $z < z_c$, then $\lim_{n\to\infty}\langle l\rangle/n = 0$. On the other hand, if $z > z_c$, then $\liminf_{n\to\infty}\langle l\rangle/n > 0$.[31]

We now prove Theorem 9 by generalising it to random objects in $\mathscr{L}^3$ of arbitrary nature:

Theorem 10. Let $u_n(k)$ be the number of objects with k "defects" and "size" n, where $k \leqslant V(n)$ and $V(n) \to vn$ as $n \to \infty$ for some constant $v > 0$. Define $u_n(\leqslant k) = \sum_{l \leqslant k} u_n(l)$. Suppose that the objects can be concatenated such that the following two super-multiplicative relations are true:

$$u_n(\leqslant k)\, u_m(\leqslant l) \leqslant \sum_{a \leqslant i \leqslant b} u_{n+m+c}(\leqslant k+l+i) \tag{4.6}$$

$$\sum_{l=0}^{k} u_n(k-l)\, u_m(l) \leqslant \sum_{d \leqslant i \leqslant e} u_{n+m+f}(k+i) \tag{4.7}$$

where $\{a, b, c, d, e, f\}$ are constants. Furthermore, suppose that there exists a $k > 1$ such that $u_n(k) \leqslant K^n$. Then the following statements are true:

(1) There exists a finite, monotonic, log-concave and non-decreasing function $\psi(\varepsilon)$ on the interval $[0, v]$, such that

$$\log \psi(\varepsilon) = \lim_{n \to \infty} \frac{1}{n} \log u_n(\leqslant \varepsilon n)$$

and we define $\psi(0) = \psi(0^+)$ and $\psi(v) = \psi(v^-)$.

(2) If there exists a finite constant $C > 1$ such that

$$\limsup_{\varepsilon \to 0^+} \frac{\log \psi(\varepsilon) - \log \psi(0^+)}{\varepsilon} \leqslant \log C$$

then the radius of convergence, $x_\psi(z)$, of the generating function in x, $G^\psi(x, z) = \sum_n \sum_l u_n(l)\, z^l x^n$, is a continuous function of $z \in (0, \infty)$ and $x_\psi(z) = (\psi(0^+))^{-1}$ for all $0 < z < 1/C$.

Proof. Statement (1) follows from the same line of arguments leading from Eq. (2.6) (equivalent to Eq. (4.6)) to Propositions 2 and Eq. (2.10). Continuity of $x_\psi(z)$ follows from existence and convexity and continuity of the free energy $(-\log x_\psi(z))$, starting from Eq. (4.7) and following the line of arguments in Ref. 1 leading to Proposition 4. From (1) it is apparent that $u_n(\leqslant \varepsilon n) = [\psi(\varepsilon)]^{n + o(n)}$. Since $\log \psi(\varepsilon) \leqslant \log \psi(0^+) + \varepsilon \log C$ by concavity and the bound on the limsup above, one obtains

$$\left[\psi\left(\frac{l}{n}\right)\right]^n \leqslant [\psi(0^+)\, C^{l/n}]^n = [\psi(0^+)]^n\, C^l$$

Thus

$$\begin{aligned} G^\psi(x, z) &= \sum_n \sum_{l \leqslant \lceil V(n) \rceil} u_n(l)\, z^l x^n \\ &\leqslant \sum_n \sum_{l \leqslant \lceil V(n) \rceil} [\psi(l/n)]^{o(n)}\, [\psi(0^+)]^n\, C^l z^l x^n \end{aligned}$$

Since $\psi(l/n)$ is non-decreasing, note that $\psi(l/n) \leqslant \psi(v)$. If $z < 1/C$, then the generating function is finite if $x < [\psi(0^+)]^{-1}$. Consequently, $x_\psi(z) \geqslant [\psi(0^+)]^{-1}$ if $z < 1/C$. On the other hand, if $z < 1$, then $G^\psi(x, z) \geqslant \sum_n u_n(\leqslant \lceil \delta n \rceil)\, z^{\lfloor \delta n \rfloor} x^n$, and by taking $\delta \to 0^+$, one obtains $x_\psi(z) \leqslant [\psi(0^+)]^{-1}$. Hence $x_\psi(z) = [\psi(0^+)]^{-1}$ if $0 < z < 1/C$. ∎

Theorem 10 can be applied to surfaces with connected skeletons: in that case $C = 4\lambda$ and $\psi(0^+) = 1$. These choices give Theorem 9.

5. CONCLUSIONS

In this paper I used a rigorous approach to explore the phase diagram for surfaces with a "crumpling" fugacity. In particular, I proved a variety of bounds on $\mathscr{F}(z)$; the most interesting being the fact that $\mathscr{F}(z) > 0$ for positive z. Numerical simulations of this model ref. 2 strongly suggested that $\mathscr{F}(z)$ might be identically zero for small enough z, but this possibility is now ruled out.[5] It is important to note that the results in this paper do not rule out a critical point in the phase diagram corresponding to a "crumpling transition".

In Section 4 I considered a restricted set of surfaces (those with connected skeletons). In this model it is indeed possible, through the use of a bound derived from the number of lattice animals, to prove that the free energy is zero for small enough (but positive) z. The phase diagram is given by Fig. 7, and there exists at least one non-analyticity in the corresponding limiting free energy. One cannot be sure that this non-analyticity corresponds to a "crumpling transition", but I conjecture this to be a tricritical point separating a line of first order transitions (to surfaces with low degree of folding), from a line of second order transitions (to a branched polymer phase consisting of surfaces with high degree of folding). It is not clear that this non-analyticity is "cancelled" when surfaces with disconnected skeletons are added back into the partition function. Thus, if one can prove that surfaces with disconnected skeletons only adds a term or a factor which is analytic at z_c to $\mathscr{F}^C(z)$ (to give $\mathscr{F}(z)$), then there is a non-analyticity in $\mathscr{F}(z)$ at z_c as well. This seems a very appealing (but challenging) possible course of investigation. (Of course, if adding back the surfaces with disconnected skeletons changes the value of z_c, then this argument would not work as proposed here).

ACKNOWLEDGMENTS

I had very fruitful discussions with S. G. Whittington, E. Orlandini and M. C. Tesi in the preparation of this manuscript. This research is supported by a grant from NSERC (Canada).

REFERENCES

1. G. Whittington, *J. Math. Chem.* **14**:103 (1993).
2. E. Orlandini, A. L. Stella, T. L. Einstein, M. C. Tesi, I. Beichl and F. Sullivan, *Phys. Rev. E* **53**:5800 (1996).

[5] If the free energy becomes identically zero for small enough z, then the phase diagram will have the general appearance of Fig. 7. This implies the existence of a multicritical point separating two critical lines. One can conjecture that this critical point corresponds to a "crumpling transition" in the model.

3. R. Lipowsky, *Nature* **349**:475 (1991).
4. E. Orlandini and M. C. Tesi, *Physica A* **185**:160 (1992).
5. A. Baumgartner, *Physica A* **192**:550 (1993).
6. E. Orlandini, A. L. Stella, M. C. Tesi and F. Sullivan, *Phys. Rev. E* **48**:R4203 (1993).
7. A. L. Stella, E. Orlandini, I. Beichl, F. Sullivan, M. C. Tesi and T. L. Einstein, *Phys. Rev. Lett.* **64**:3650 (1992).
8. U. Glaus, *Phys. Rev. Lett.* **56**:1996 (1986).
9. U. Glaus U., *J. Stat. Phys.* **50**:1141 (1988).
10. U. Glaus and T. L. Einstein, *J. Phys. A: Math. Gen.* **20**:L105 (1987).
11. J. O'Connell, F. Sullivan, D. Libes, E. Orlandini, M. C. Tesi, A. L. Stella and T. L. Einstein, *J. Phys. A: Math. Gen.* **24**:4619 (1991).
12. A. Baumgartner and A. Romero, *Physica A* **187**:243 (1992).
13. M. E. Fisher, A. J. Guttmann and S. G. Whittington, *J. Phys. A: Math. Gen.* **24**:3095 (1991).
14. M. E. Fisher, *Physics* **3**:255 (1967).
15. A. F. Andreev, *Sov. Phys.-JETP* **18**:1415 (1964).
16. S. N. Isakov, *Commun. Math. Phys.* **95**:427 (1984).
17. A. L. Stella, *Turkish J. Phys.* **18**:244 (1994).
18. E. J. Janse van Rensburg and S. G. Whittington, *J. Phys. A: Math. Gen.* **22**:4939 (1989).
19. E. J. Janse van Rensburg and S. G. Whittington, *J. Phys. A: Math. Gen.* **23**:2495 (1990).
20. N. Madras, C. E. Soteros, S. G. Whittington, J. L. Martin, M. F. Sykes, S. Flesia and D. S. Gaunt, *J. Phys. A: Math. Gen.* **23**:5327 (1990).
21. N. Madras, C. E. Soteros and S. G. Whittington, *J. Phys. A: Math. Gen.* **21**:4613 (1988).
22. E. J. Janse van Rensburg, *J. Phys. A: Math. Gen.* **23**:5879 (1990).
23. E. J. Janse van Rensburg, *J. Phys. A: Math. Gen.* **25**:3529 (1992).
24 E. J. Janse van Rensburg, *J. Knot Theory Ram.* **3**:365 (1994).
25. B. Durhuus, J. Fröhlich and T. Jonsson, *Nucl. Phys.* **B225[FS9]** 185 (1983).
26. J. Ginibre, A. Grossmann and D. Ruelle, *Commun. Math. Phys.* **3**:18 (1966).
27. E. Hille, *Functional Analysis and Semi-Groups* (*AMS Colloq. Publ.*) **31** (New York: AMS) (1948).
28. J. B. Wilker and S. G. Whittington, *J. Phys. A: Math. Gen.* **12**:L245 (1979).
29. D. Klarner, *Can. J. Math.* **19**:851 (1967).
30. R. Buck, *Advanced Calculus* 2nd ed. McGraw-Hill, New York 1965 (1965).
31. M. E. Fisher, *J. Math. Phys.* **6**:1643. Colloq. Publ. **31** (New York: AMS) (1965).