Mapping between a self-attracting self-avoiding surface and a percolation hull in three dimensions

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I show that for a certain range in its parameter space, a self-attracting self-avoiding surface can be mapped onto the hull of a percolation cluster in three dimensions. At a particular point on the boundary of the low-temperature phase, the self-attracting self-avoiding surface can be mapped onto a percolation hull at threshold.

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At high temperatures, a polymer chain in a good solvent behaves essentially as if it were a self-avoiding walk (SAW): Its mean radius of gyration grows as $\langle R \rangle \sim N^{\nu}$, where ν has the same value as for the SAW and N is the molecular weight [1]. As the temperature T is reduced, however, the short-ranged van der Waals attraction between monomers becomes increasingly important. At a certain temperature T_{θ}, the chain collapses. The exponent ν is $1/d$ in d dimensions for all $T < T_{\theta}$. The theta point $T = T_{\theta}$ is a tricritical point [1].

Recently, there has been a flurry of interest in the properties of random self-avoiding surfaces [2–9]. A self-avoiding surface (SAS) is a connected set of elementary plaquettes on a regular lattice with the property that each lattice bond belongs to at most two plaquettes. SAS's are natural generalizations of the self-avoiding walk problem, and they arise in high-temperature expansions of lattice gauge theories [10].

In this paper, we shall study a self-attracting self-avoiding surface (SASAS). The SASAS collapse transition that occurs as the temperature is reduced is a natural analog of the theta point in linear polymers.

Self-attracting self-avoiding surfaces have not been studied previously. However, simulations of self-attracting "tethered surfaces" have been performed [11,12]. A tethered surface consists of a regular array of hard spheres tied together by flexible strings [13,14]. The length of the strings is taken to be short enough that the surface is self-avoiding. Tethered surfaces are expected to be a good model of polymer membranes at high temperatures and in good solvents. However, in addition to the hard-core repulsion between monomers, there is a longer-ranged van der Waals attraction between monomers in a real polymer membrane. This attractive interaction becomes increasingly important as the temperature T is reduced and, at low temperatures, the membrane collapses. In the low-temperature phase, the membrane has fractal dimension 3. Recently, Abraham and Kardar [11] and Liu and Plischke [12] studied the nature of this collapse transition. The results of Liu and Plischke are consistent with recent experimental work on exfoliated sheets of graphite oxide in aqueous suspension [15,16].

In attempting to make progress on the SASAS problem, it is natural to begin by mimicking the approaches that have been most useful in elucidating the nature the collapse transition of a polymer chain in two dimensions (2D). Perhaps the most important tool used in the theory of the theta polymers in 2D has been the mapping introduced by Coniglio *et al.* [17]. These workers showed that at its collapse transition, a self-attracting polymer ring on the hexagonal lattice can be mapped onto the hull of a percolation cluster at threshold. Since it has been proven that the radius of gyration exponent ν is $\frac{4}{7}$ for the perimeter of a percolation cluster at threshold in 2D [18], the value of the size exponent at the tricritical point ν_t must also be $\frac{4}{7}$. Similar approaches to the collapse transition of a polymer chain in 2D have been developed by Duplantier and Saleur [19] and Bradley [20,21].

In this paper, I demonstrate that for a range of parameter values, a certain self-attracting self-avoiding surface can be mapped onto the hull of a percolation cluster in three dimensions (3D). This mapping is a generalization of the mapping of Coniglio *et al.* to SASAS's embedded in a 3D lattice [22]. I also show that at a particular point on the boundary of its low-temperature phase, the SASAS is equivalent to a percolation hull at threshold. Finally, the phase diagram of the SASAS is discussed.

Our SASAS is defined on the lattice dual to the face-centered cubic (fcc) lattice. The Wigner-Seitz (WS) cell of the fcc lattice is a rhombic dodecahedron (Fig. 1). The WS (or dual) lattice is constructed by packing rhombic dodecahedra to fill space. A site of the original lattice resides at the center of each WS cell. Each of the twelve faces of a WS cell are identical rhombi, and each of these rhombi bisect bonds between nearest-neighbor (NN) pairs of sites in the original fcc lattice. For brevity, we will refer to a face of a WS cell as a "plaquette." Note that each bond in the WS lattice is shared by three plaquettes.

Our SASAS's are made up of occupied plaquettes. A set of occupied plaquettes on the WS lattice will be called connected if, given two plaquettes π and π' in the set, there exists a sequence of occupied plaquettes $\pi, \pi_1, \ldots, \pi_n, \pi'$ with the property that each consecutive pair of plaquettes has a common edge. A set of occupied plaquettes on the WS lattice will be called a closed self-avoiding surface if the set is connected and if each bond in the WS lattice is shared by either zero or two occupied plaquettes. If a face of a WS cell is occupied, we will say that the fcc lattice site at the center of the cell is "adjacent" to the occupied plaquette. Similarly, we say that this fcc lattice site is adjacent to the SASAS itself.

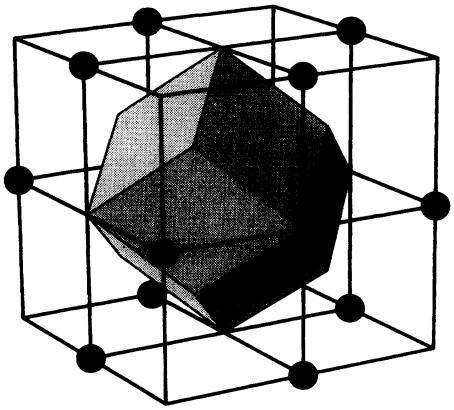

FIG. 1. A Wigner-Seitz cell for the fcc lattice. The sites of the fcc lattice are shown using solid circles. The shaded solid is the Wigner-Seitz cell. The fcc lattice site that resides at the center of this rhombic dodecahedron is not shown.

Consider the equilibrium statistical mechanics of a closed self-avoiding surface with N plaquettes on the WS lattice. For simplicity, we take one of the plaquettes in the surface to be held fixed. We shall permit the genus of the surface to assume any value. To assign an energy to a given surface configuration C, we first color the fcc lattice sites adjacent to the surface. Sites inside the surface are colored black, while sites outside the surface are colored white. We then assign an energy $\epsilon_+ \geq 0$ to the black sites and an energy $\epsilon_- \geq 0$ to the white sites. Let the number of black sites be $n_+(C)$ and the number of white sites be $n_-(C)$. The energy of the configuration C is thus

$$E(C)=\epsilon_+ n_+(C)+\epsilon_- n_-(C) .$$

For $\epsilon_+=\epsilon_-=0$, our SASAS reduces to a closed SAS without self-attraction and with arbitrary genus. On the other hand, if either ϵ_+ or ϵ_- is positive, the SAS is self-attracting, since the configurations C with the smallest values of $n_+(C)$ and $n_-(C)$ have the lowest energies.

For the case $\epsilon_+=\epsilon_-$, our SASAS is a natural generalization of the self-attracting self-avoiding loop introduced by Coniglio *et al.* [17] from two dimensions to three. Now consider our SASAS in the general case in which ϵ_+ and ϵ_- differ. For definiteness, assume that $\epsilon_+ > \epsilon_-$. In this case, the inside of the SAS attracts itself more strongly than the outside does. Such an asymmetry could occur in a real vesicle if opposite sides of the membrane have different chemical composition. Note, however, that SASAS's of all possible genuses occur in our ensemble. In contrast, the topology of a vesicle is fixed.

The partition function for a surface of N plaquettes is

$$Z(x,y,N)=\sum_{C'} x^{n_+(C')} y^{n_-(C')} . \tag{1}$$

Here $\beta \equiv T^{-1}$, $x \equiv \exp(-\beta\epsilon_+)$, $y \equiv \exp(-\beta\epsilon_-)$, and the sum runs over all closed SAS configurations C'. The Boltzmann weight of the configuration C is

$$w(C,x,y,N)=x^{n_+(C)} y^{n_-(C)}/Z(x,y,N) . \tag{2}$$

We will now show that for $x+y=1$, our SASAS can be mapped onto the hull of a site percolation cluster on the fcc lattice. We begin by defining the hull for site percolation on the fcc lattice.

Following the work of Strenski, Bradley, and Debierre [23] and Bradley, Strenski, and Debierre [24], we take the hull of a percolation cluster to be a closed SAS composed of plaquettes on the WS lattice. Consider an arbitrary plaquette in the hull. By definition, one of the fcc lattice sites adjacent to this plaquette belongs to the cluster, while the other does not. Since the fcc lattice sites adjacent to the plaquette are nearest neighbors, one of these sites is occupied and the other is not. If the fcc lattice sites inside the hull and adjacent to it are occupied, the hull will be called an external percolation hull; otherwise, it is an internal percolation hull.

The hulls in site percolation on the fcc lattice are especially simple for two reasons. First, each bond in the WS lattice either does not belong to a hull or is shared by two plaquettes in a hull. This is simpler than the situation for the simple cubic lattice. Bonds in the WS lattice of the simple cubic lattice can be shared by zero, two, or four plaquettes in hulls, and as a consequence there is more than one possible definition of the hull [24]. The second reason is that one of the fcc lattice sites adjacent to a plaquette in the hull is occupied and the other is not. In contrast, the hull is more complex in site percolation on the bcc lattice, the problem studied by Strenski, Bradley, and Debierre [23] and Bradley, Strenski, and Debierre [24]. In that case, there are two types of plaquettes—hexagonal and square. Hexagonal faces bisect bonds between NN pairs of sites in the original lattice, while square faces lie between next-nearest-neighbor sites. As a result, the sites adjacent to a square plaquette in the hull may both be occupied, or one of these sites could be unoccupied.

Consider an arbitrary closed self-avoiding surface C on the dual lattice. The number of fcc lattice sites that are adjacent to C and that are inside (outside) the surface will be denoted $n_+(C)$ $[n_-(C)]$. The probability that a given plaquette π belongs to an external percolation hull of N plaquettes is given by

$$P(p,N)=\sum_{C'} p^{n_+(C')}(1-p)^{n_-(C')} . \tag{3}$$

Here the sum runs over all closed SAS's C' with N plaquettes that contain π. If it is given that the plaquette π belongs to an external hull containing N plaquettes, the probability of a particular external hull configuration C is

$$p(C,p,N)=p^{n_+(C)}(1-p)^{n_-(C)}/P(p,N) . \tag{4}$$

Simple expressions of this kind do not apply for hulls on either the simple cubic or bcc lattices.

We are now ready to construct our mapping. Combining Eqs. (3) and (4) with Eqs. (1) and (2), we obtain

$$Z(p,1-p,N)=P(p,N) \tag{5}$$

and

$$w(C,p,1-p,N)=p(C,p,N) . \tag{6}$$

Analogous relations exist between the SASAS and inter-

nal percolation hulls, but these will not be needed and so I will not pause to discuss them.

We have shown that for $x+y=\exp(-\beta\epsilon_+)+\exp(-\beta\epsilon_-)=1$, our SASAS is equivalent to an external percolation hull for site percolation on the fcc lattice with $p=x$. Accordingly, we will refer to the line $x+y=1$ in the parameter space of the SASAS as the "percolation line."

I shall now show that at a particular point on the boundary of its collapsed low-temperature phase, the SASAS can be mapped onto the hull of a 3D percolation cluster at threshold. In our discussion, we will use the mapping just described in combination with what is known about the behavior of percolation hulls in 3D.

At $T=\infty$ our self-avoiding surfaces are not self-attracting and their genus is unrestricted. SAS's of this kind are in the same universality class as surfaces with genus zero [9]. Thus, they are in the same universality class as lattice animals.

Based on our knowledge of the behavior of self-attracting self-avoiding walks, we expect that so long as either ϵ_+ or ϵ_- is positive, a collapse transition of our SASAS will occur at a certain temperature $T_\theta>0$. (Of course, T_θ will depend on ϵ_+ and ϵ_-.) For $T\gg T_\theta$, the scaling properties of the surface should be the same as at $T=\infty$. For $T<T_\theta$, on the other hand, the SASAS collapses and it has fractal dimension $D=3$.

Recall that for $x+y=1$, the SASAS is equivalent to the hull of a percolation cluster with $p=x$. Recently, Strenski, Bradley, and Debierre [23], Bradley, Strenski, and Debierre [24], and Cao and Wong [25] carried out extensive Monte Carlo simulations of percolation hulls in 3D. For $p<p_c$ and for $p>1-p_c$, only finite percolation hulls exist. For $p_c\le p\le 1-p_c$, there is an infinitely large hull. The asymptotic fractal dimension of the infinite hull is 3 for $p_c<p<1-p_c$ [23,24,26,27]. Monte Carlo simulations yielded the estimates $D'=2.548\pm0.014$ for the fractal dimension of the infinite hull at threshold and $\sigma'=0.46\pm0.01$ for the crossover exponent [23,24]. The percolation threshold for site percolation on the fcc lattice was found to be $p_c=0.1998\pm0.0006$ using a series expansion [28].

For $p_c<p<1-p_c$, the asymptotic fractal dimension of the hull is 3. This is the fractal dimension of the SASAS in its low-temperature phase. Thus, the segment of the percolation line with $p_c<x<1-p_c$ must lie in the low-temperature phase.

I shall now argue that the segment of the percolation line with $x<p_c$ cannot lie in the low-temperature phase of the SASAS. Call this segment S. For $p<p_c$, only finite percolation hulls exist. In this regime, percolation clusters with linear dimensions large compared to the correlation length ξ are believed to have the same fractal dimension as lattice animals in 3D [29]. The fractal dimension of lattice animals in 3D is exactly 2 [30]. Lattice animal hulls have not yet been studied and their fractal dimension is unknown. It is clear, however, that the fractal dimension of the lattice animal hull cannot exceed the fractal dimension of the cluster itself, and so is less than or equal to 2. Thus, the segment S does not lie in the low-temperature phase of the SASAS.

We now see that the point $(x,y)=(p_c,1-p_c)$ lies on the boundary of the low-temperature phase. At this point (which I shall call *Pe*), the fractal dimension of the SASAS is equal to fractal dimension of the hull of a 3D percolation cluster at criticality, D'. Moreover, the crossover exponent which describes the approach to the point *Pe* along the percolation line $x+y=1$ is equal to σ'. Similar conclusions apply to the SASAS at the point $(x,y)=(1-p_c,p_c)$.

What is the nature of the SASAS phase diagram? In the simplest scenario, the SASAS would have two phases, a high-temperature lattice animal phase, and a collapsed low-temperature phase. The parameter ϵ_-/ϵ_+ would be irrelevant, and so all points on the phase boundary would be characterized by the same critical exponents.

Although this simple scenario is appealing, it is almost certainly incorrect. For $\epsilon_+=\epsilon_-$, the SASAS collapse transition should be in the same universality class as the collapse of a self-attracting lattice animal. An exact enumeration study of the collapse of a lattice animal with a contact fugacity yielded the estimate $\phi=0.82\pm0.02$ for the crossover exponent [31]. This strongly suggests that the collapse transitions that occur at the point *Pe* and on the line $x=y$ are in different universality classes. Additional evidence for this comes from the recent conjecture that $D=2$ and $\phi=1$ at the theta point of a 3D lattice animal [9]. These exponents are far from D' and σ'.

The collapse transition of the SASAS with $\epsilon_+=\infty$ and $\epsilon_->0$ is also most likely in a different universality class than the transition at *Pe*. We begin our discussion of this transition by studying the ground states of the SASAS with $\epsilon_+>0$ and $\epsilon_-=0$. For simplicity, we consider the case in which $N=10M+2$, where M is a positive integer, but similar considerations hold for general values of N. An allowed conformation of the SASAS can be constructed as follows: We begin with a tree of M occupied sites on the *dual* fcc lattice. (A tree is a cluster of sites with no loops). The corresponding SASAS on the WS lattice consists of those plaquettes that belong to one and only one WS cell with an occupied dual lattice site at its center. It can be shown that this configuration is a ground state of the SASAS, and that all of the ground states of the SASAS can be constructed in this fashion [32].

In the limit in which $\epsilon_+\to\infty$ and $\epsilon_-=0$, only the ground state configurations of the SASAS are allowed states. Thus, for $x=0$ and $y=1$ the SASAS with $N=10M+2$ plaquettes can be mapped onto a tree of M sites on the dual lattice. We conclude that for $x=0$ and $y=1$ the SASAS has the same scaling behavior as a lattice tree. Since lattice trees are in the same universality class as lattice animals [33,34], the SASAS has the same scaling behavior as a lattice animal.

For $x=0$ and $y=1$ the SASAS can be mapped onto a lattice tree without self-attraction. More generally, the SASAS is equivalent to a self-attracting lattice tree for $x=0$ and $0\le y\le 1$. As y is reduced from 1, the attraction between the branches of the tree becomes increasingly important. The tree collapses at a critical value of y which we denote by y_c. The collapse transition of a 3D self-attracting tree has recently been studied by Gaunt and Flesia [35]. They obtained the estimate

$\phi = 0.82 \pm 0.03$ for the crossover exponent for this transition, which is far from σ'. This strongly suggests that the collapse transitions at *Pe* and the point $(x,y)=(0,y_c)$ are in different universality classes.

We have all but ruled out the simplest scenario for the phase diagram of the SASAS. What then is the nature of the phase diagram? The answer to this question must await future work. The 2D analog of our SASAS has three phases—in addition to the usual high- and low-temperature phases, there is a "branched polymer" phase at intermediate temperatures [36]. Monte Carlo or exact enumeration studies of our SASAS would be particularly illuminating, since they could readily reveal the number and kind of phases present. A particularly exciting possibility is that there is an unexpected third phase present, and that *Pe* is a tetracritical point at the confluence of the three phases.

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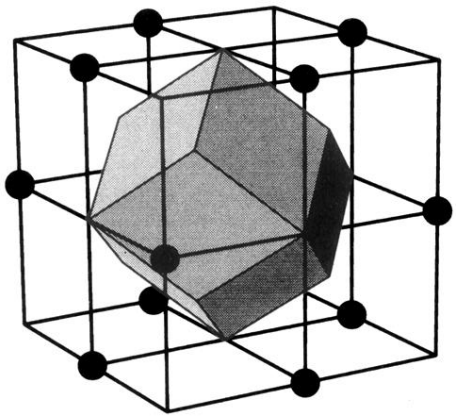

FIG. 1. A Wigner-Seitz cell for the fcc lattice. The sites of the fcc lattice are shown using solid circles. The shaded solid is the Wigner-Seitz cell. The fcc lattice site that resides at the center of this rhombic dodecahedron is not shown.