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Excluded volume and its relation to the onset of percolation

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The general relationship between the percolation threshold of systems of various objects and the excluded volume associated with these objects is discussed. In particular, we derive the average excluded area and the average excluded volume associated with two- and three-dimensional randomly oriented objects. The results yield predictions for the dependencies, of the percolation critical concentration of various kinds of "sticks," on the stick aspect ratio and the anisotropy of the stick orientation distribution. Comparison of the present results with available Monte Carlo data shows that the percolation threshold of the sticks is described by the above dependencies. On the other hand, the numerical values of the excluded area and the excluded volume are not dimensional invariants as suggested in the literature, but rather depend on the randomness of the stick orientations. The usefulness of the present results for percolation-threshold problems in the continuum is discussed. In particular, it is shown that the excluded area and the excluded volume give the number of bonds per object B_c when the objects are all the same size. In the case where there is a distribution of object sizes, the proper average of the excluded area or volume is a dimensional invariant while B_c is not.

I. INTRODUCTION

Thirteen years ago it was shown by Scher and Zallen[1] that the fractional area s_c and the fractional volume τ_c associated with the onset of percolation are, to a reasonable accuracy, dimensional invariants for all lattices. They obtained their result by inscribing circles or spheres about lattice sites with radii of half the nearest-neighbor distance. Multiplying the computed results for the site-percolation critical occupation probability p_c by the filling factor of the lattices, they have determined s_c and τ_c. In this case of hard-core circles and spheres (neighboring circles or spheres, touching at one point) they found that $s_c=0.44$ and $\tau_c=0.16$. These results were found to be "universal" with an accuracy of a few percent. When a random system of hard-core spheres was considered, the critical fractional volume found[2] was somewhat larger, $\tau_c=0.18$. Similar "universal" behaviors were found in continuum problems for which soft-core (interpenetrating) circles and spheres had been considered. For these cases it is by now well established[3,4] that $s_c=0.68$ and $\tau_c=0.29$. These values are also consistent with a total critical area $N_c a$ of 1.10 ± 0.05, and a total critical volume $N_c v$ of 0.35 ± 0.02, of the occupying circles and spheres. Here N_c is the critical concentration of the circles of given area a (spheres of given volume v), which are randomly distributed in a unit square (cube) and for which $a \ll 1$ ($v \ll 1$). These values have been derived[4–8] by determining the critical radius, the critical average number of bonds per site, or the area s_c (volume τ_c). As was shown by Pike and Seager[4] these are all consistent and yield the $N_c a$ and $N_c v$ values given above.

Following the above universal values of $N_c a$ and $N_c v$, one can also consider the universal values of the corresponding total excluded area A_{ex} and total excluded volume V_{ex}. The excluded area (volume) of an object is defined as the area (volume) around an object into which the center of another similar object is not allowed to enter if overlapping of the two objects is to be avoided.[9] The total excluded area (volume) is this area (volume) multiplied by N_c. It is trivial that for circles, $A_{\text{ex}}=4N_c a$, while for spheres, $V_{\text{ex}}=8N_c v$. Skal and Shklovskii[8] have shown that the "universality" applies also to systems of other regular objects (e.g., cubes and ellipsoids). Their study, however, was limited to the cases in which all the objects are aligned parallel to each other. One notes that in all these cases, the shapes of the excluded volumes are the same as those of the objects of which the percolation system is made. As for the spheres[4,5] for which $V_{\text{ex}}=2.8$, they found a value of $V_{\text{ex}}\approx 3$ for all the objects considered in their work.

There has recently been considerable interest in the percolation properties of systems made of nonspherical objects (which can be described as "sticks") that have random orientations in space. Examples include polymers,[10] fiber-enhanced polymers,[11–14] and patterns of fractures in rocks.[15] So far, however, only two-dimensional Monte Carlo simulations of randomly aligned zero-width sticks have been reported.[4,14,15] As shown by Onsager[9] in a different context, the excluded volume for an elongated object is very different in shape from the actual object and it depends (unlike the above-mentioned cases) on the relative orientation of the objects. Hence, for a statistical distribution of orientations one can only define an *average* excluded area $\langle A \rangle$ such that the total excluded area $\langle A_{ex} \rangle$ is given by

$$\langle A_{ex} \rangle = \langle A \rangle N_c \ , \tag{1a}$$

and similarly for the average excluded volumes:

$$\langle V_{ex} \rangle = \langle V \rangle N_c \ . \tag{1b}$$

Using these concepts we would like to test a generalized hypothesis of Scher and Zallen, i.e., whether the quantities $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ are good enough for the prediction of percolation thresholds. To do this we calculate $\langle A \rangle$ and $\langle V \rangle$ for line segments and narrow strips in two dimensions and for cylindrical rods in three dimensions. In the averaging procedure we consider both isotropic and anisotropic angular distributions. This is done in Sec. II. In Sec. III we compare the results with available Monte Carlo data. Following this comparison it is concluded that there are two kinds of system invariants. These are discussed in Sec. IV.

II. CALCULATION OF EXCLUDED AREA AND VOLUME

In this section we derive first the excluded area of a system of widthless sticks because the large amount of data available for this system allows one to make a detailed comparison between the excluded-area theory and the Monte Carlo computations. We proceed then with the case of finite-width two-dimensional sticks (e.g., rectangles) and we conclude by considering the case of a three-dimensional stick (capped-cylinder) system.

A. Excluded area of the widthless stick

Let us consider a stick of length L which makes an angle θ_i with respect to a given direction in the plane. Let another stick make an angle θ_j with the same given direction. As can easily be seen from Fig. 1 the excluded area is simply the area of the parallelogram,

$$L^2 \sin(\theta_i - \theta_j) \ . \tag{2}$$

This is the excluded area for two given sticks. For an ensemble of sticks we must average over all possible orientations of the sticks by considering the distribution function of the angles $P(\theta_i)$. Since generalization to various distributions will become apparent from the following discussion, we start from the simple uniform distribution in which the angles between the sticks and the predetermined direction are randomly distributed within the interval[14]

$$-\theta_\mu \le \theta_i, \theta_j \le \theta_\mu \ , \tag{3}$$

where $\theta_\mu \le \pi/2$. The isotropic case is given by $\theta_\mu = \pi/2$, and the smaller the θ_μ, the more anisotropic the system. As was shown[14] previously, the macroscopic anisotropy of a system of N sticks can be defined as

$$P_{\parallel}/P_{\perp} = \sum_{i=1}^{N} |\cos\theta_i| \Big/ \sum_{i=1}^{N} |\sin\theta_i| \ . \tag{4}$$

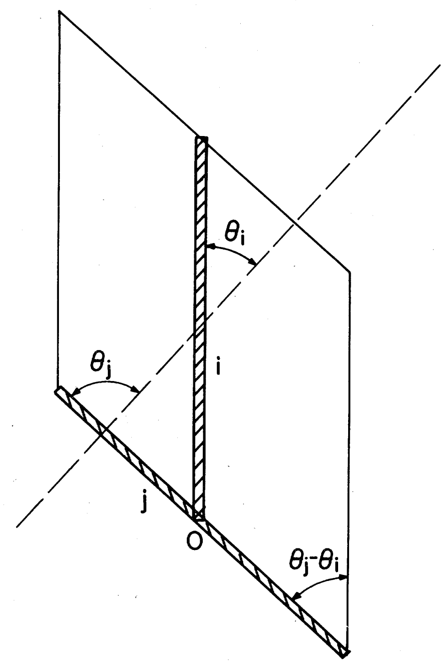

FIG. 1. Two "widthless" sticks (shaded area) and their corresponding excluded area. This area is the parallelogram which is obtained by following the center 0 of the stick j as it travels around the stick i while being parallel to itself and touching stick i at a single point.

It can easily be shown[14] that for large N and a random distribution of the stick orientations, this anisotropy is just

$$P_{\parallel}/P_{\perp} = \cot(\theta_\mu/2) \ . \tag{5}$$

For the uniform distribution of angles we must consider all possible angles θ_i and θ_j and their corresponding uniform probability

$$P(\theta_i) = 1/2\theta_\mu \tag{6}$$

in the interval $2\theta_\mu$. Hence the averaged excluded area is

$$\langle A \rangle = L^2 \int_{-\pi/2}^{\pi/2} \int_{-\pi/2}^{\pi/2} \sin|\theta_i - \theta_j| P(\theta_i) P(\theta_j) \times d\theta_i d\theta_j \ . \tag{7}$$

Substituting the distribution (6) in Eq. (7) yields the average excluded area

$$\langle A \rangle = (L/2\theta_\mu)^2 [4\theta_\mu - 2\sin(2\theta_\mu)] \ . \tag{8}$$

For the isotropic case, $\theta_\mu = \pi/2$, the excluded area will then be

$$\langle A \rangle = (2/\pi) L^2 \ . \tag{9}$$

If the assumption of the dimensional invariance of the excluded area is correct, i.e., if $\langle A_{ex} \rangle = A_{ex}$, we can derive the desired information for the stick system on the basis of the information available, for example, for the circle system.[4] For the isotropic case we can find by using Eqs. (1a) and (9) that the critical stick length (for a system of N_c sticks), L_{ci}, is

$$L_{ci} = [(\pi/2)(A_{ex}/N_c)]^{1/2} \ . \tag{10}$$

This result is in agreement with the empirical criterion found by Monte Carlo simulation[4] which states that $N_c L_{ci}^2 = \text{const}$. For the dependence of the percolation threshold on the anisotropy of the system, a weaker condition, i.e., that $\langle A_{ex} \rangle$ is invariant for the stick system, is sufficient. Using Eqs. (8) and (9), this yields

$$L_c / L_{ci} = (2\theta_\mu) / \{ \pi [2\theta_\mu - \sin(2\theta_\mu)] \}^{1/2} . \tag{11}$$

In Ref. 14 we have concluded from other considerations that

$$\begin{aligned} L_c / L_{ci} &= 1/(\sin\theta_\mu)^{1/2} \\ &= \{ \tfrac{1}{2} [(P_\parallel / P_\perp) + 1/(P_\parallel / P_\perp)] \}^{1/2} . \end{aligned} \tag{12}$$

As will be discussed in Sec. III, predictions (11) and (12) are practically the same, indicating that the above weaker condition is fulfilled. In Eqs. (10)—(12) we have derived L_{ci} and L_c / L_{ci} for a system of a given concentration of sticks, since this was the system discussed in the more detailed Monte Carlo studies.[4,14] If one considers a system where the stick length L is given, and the variable is the critical stick density N_c, one immediately obtains from Eq. (10) that, in the isotropic case,

$$N_{ci} = (\pi / 2L^2) A_{ex} . \tag{13}$$

For the anisotropic case we obtain by using Eqs. (1a), (8), and (12) that

$$N_c / N_{ci} = \tfrac{1}{2} [(P_\parallel / P_\perp) + 1/(P_\parallel / P_\perp)] . \tag{14}$$

Another, simple, example of the utilization of the same method for widthless sticks is for a system in which the sticks can be either horizontal or vertical. In fact, this is essentially a highly-correlated lattice problem where the lattice unit length (or the mesh used) is much smaller than the stick length. Here the anisotropy is introduced by having M sticks in the vertical direction for every stick in the horizontal direction. Using the definition given by Eq. (4), the macroscopic anisotropy of this system is simply M. For calculating the average excluded area we must proceed by using Eq. (7). The probability function in the present case however, is

$$P(\theta_i) = [\delta(\theta_i) + M\delta(\theta_i - \pi/2)] / (M+1) . \tag{15}$$

One obtains then that

$$\langle A \rangle = 2ML^2 / (1+M)^2 . \tag{16}$$

In the isotropic ($M=1$) case the critical stick length is $L_{ci} = \sqrt{2\langle A \rangle}$ and for the anisotropic case

$$L_c = L_{ci} [(M+1)^2 / 4M]^{1/2} . \tag{17}$$

Hence, as in the previous random case, the dependence of the percolation threshold on the system's anisotropy is in a form which can be readily compared with Monte Carlo results (see Sec. III).

B. The excluded area of a stick with a finite width

Similar to the procedure carried out in Sec. II A for widthless sticks, we derive here the excluded area for "sticks" having a length L and a width W. We discuss such rectangles since Monte Carlo computations are available for squares. Generalization of these computations to rectangles or other regular two-dimensional objects is quite easy.

Let us consider then two sticks (rectangles), the angle between which is $\theta = \theta_i - \theta_j$. The excluded area can be obtained simply by moving one stick around the other and registering the center of the moving stick. In Fig. 2 we show a result of such a procedure. The shaded area represents the stationary stick and the curve is the path of the center of the other stick as it is moved around the first stick. The area within the curve is the excluded area. A quick calculation shows that this excluded area is given by

$$\begin{aligned} &(L\sin\theta + W + W\cos\theta)(L + W\sin\theta + L\cos\theta) \\ &\qquad - (L^2 + W^2)\sin\theta\cos\theta . \end{aligned} \tag{18}$$

Application of the uniform distribution [see Eq. (6)] and Eq. (7) yields then the average excluded area:

$$\begin{aligned} \langle A \rangle = {} & 2WL [1 + (1/2\theta_\mu^2)(1 - \cos 2\theta_\mu)] \\ & + (L^2 + W^2)(4\theta_\mu - 2\sin 2\theta_\mu)/(4\theta_\mu^2) . \end{aligned} \tag{19}$$

This result can readily be simplified for the square ($L = W$) isotropic ($\theta_\mu = \pi/2$) case yielding

$$\langle A \rangle = 2L^2 [1 + 2/\pi + (2/\pi)^2] . \tag{20}$$

Another two-dimensional finite-width stick is the "capped" rectangle stick. This object is useful because it can be extrapolated to a circle. In addition, the derivation of the excluded area of this object indicates how to handle the three-dimensional problem (see Sec. III C). We assume now a rectangle of length L, width W, and caps of radius $W/2$ at its ends. As in Fig. 2, we show in Fig. 3 the capped rectangle and the excluded area which is formed around it. One can readily find that the excluded area for these two sticks, which have an angle θ between them, is

$$4WL + \pi W^2 + L^2 \sin\theta . \tag{21}$$

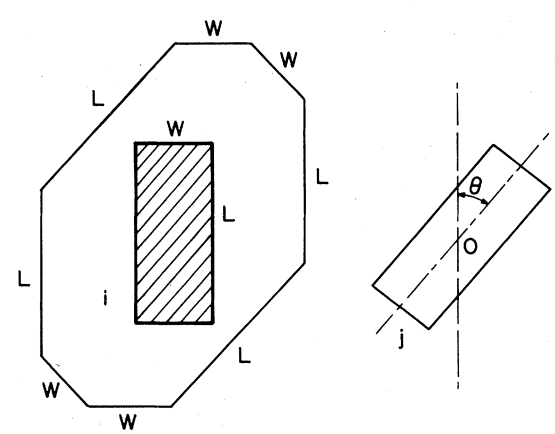

FIG. 2. Two sticks of length L and width W, the angle between which is θ. The excluded area is obtained by following the center 0 as stick j travels around stick i while touching it at least at one point.

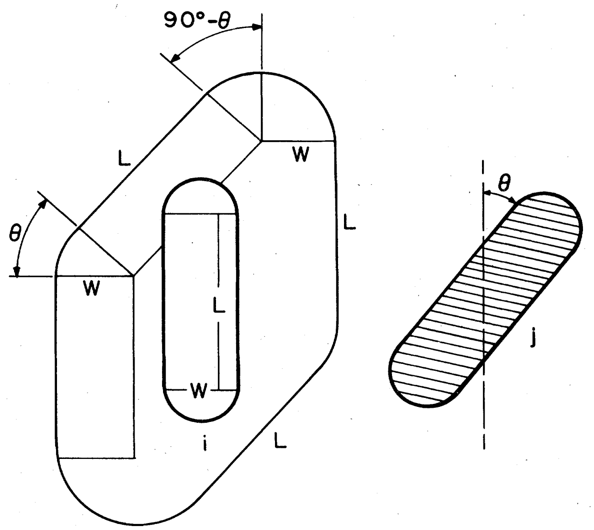

FIG. 3. This configuration is the same as that of Fig. 2 except that the sticks are capped rectangles. The length of the sticks is L, their width is W and the radius of the caps is $W/2$.

Application of the integration procedure, used in Eq. (7) for the uniform random-orientation-distribution case, now yields

$$\langle A \rangle = 4WL + \pi W^2 + (L/2\theta_\mu)^2[4\theta_\mu - 2\sin(2\theta_\mu)] \ . \quad (22)$$

As can be appreciated by comparing Eq. (19) with Eq. (22), the angular dependence is simpler in the latter case. This enables a simpler comparison with computational data. Also apparent is the fact that when the stick is reduced to a circle ($L/W \to 0$) we recover the excluded area of the circle. Results (19) and (22) can be used for the determination of the dependence of L_c on the aspect ratio L/W and on the macroscopic anisotropy of the system. This by expressing L_c in terms of A_{ex} or $\langle A_{ex} \rangle$ as was done for the widthless-stick case [Eqs. (10) and (11)].

C. Excluded volume of a stick

In three dimensions we must consider two elongated objects, the axes of which are determined by their spherical coordinates θ_j, θ_i, ϕ_i, and ϕ_j. We can derive the excluded volume for sticks which are shaped as a capped cylinder by an argument similar to the one used in two dimensions. Let γ be the angle between the axes of the two cylinders in the three-dimensional space. All we have to do is to move stick j around stick i, keeping stick j parallel to itself, so that the two sticks just touch each other. Considering sticks which are capped cylinders yields an excluded volume which is a capped parallelepiped. In a plane which is parallel to the capped parallelepiped, the projection of the excluded volume is the capped parallelogram shown in Fig. 3. In the present three-dimensional case we then obtain, by moving one capped cylinder (of length L and radius $W/2$) around the other, a capped parallelepiped which is $2W$ wide. As can be appreciated from Fig. 3, the parallelepiped is capped by four half-cylinders, of radius W and length L (instead of rectangles in two dimensions), and by four spherical sectors (rather than circular sectors in two dimensions) which add up to a full sphere. Correspondingly, the excluded volume of the capped cylinder is

$$(4\pi/3)W^3 + 2\pi W^2 L + 2WL^2 \sin\gamma \ . \quad (23)$$

To get the value of the average excluded volume $\langle V \rangle$ for the randomly oriented system, one must average $\sin\gamma$ over all possible solid angles of stick i and stick j. The full expression for this average is given in Appendix A. Here, we simply write the averaged excluded volume of the randomly oriented system as

$$\langle V \rangle = (4\pi/3)W^3 + 2\pi W^2 L + 2WL^2 \langle \sin\gamma \rangle_\mu \ , \quad (24)$$

where $\langle \sin\gamma \rangle_\mu$ is the above-mentioned average when θ_i and θ_j are confined to an angle of $2\theta_\mu$ around the z axis of the system. We note in passing that for the isotropic case of $\theta_\mu = \pi/2$ one finds that $\langle \sin\gamma \rangle_\mu = \pi/4$. Another point to note is that for the all-parallel stick system, $\sin\gamma \equiv 0$ and the excluded volume is (as for spheres) 8 times the true volume of the cylinders from which the system is made. We can conclude, on the basis of these results and the results obtained for other objects,[5,8] that the excluded volume in the all-parallel-object systems is expected to be a dimensional invariant. To illustrate the all-parallel case, which is visually simpler than the $\theta_\mu > 0$ case, we show in Fig. 4 the excluded area of two parallel

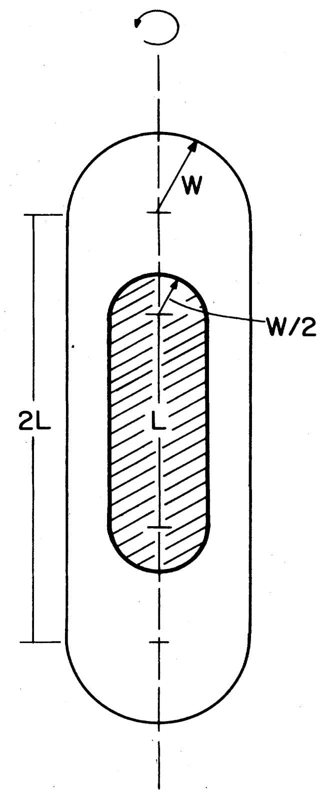

FIG. 4. Capped rectangle and the corresponding excluded area which is obtained with $\theta = 0$. In three dimensions the stick is a capped cylinder and so is the excluded volume. Both the stick and the excluded volume are obtained by rotating the two-dimensional figure around the axis shown.

capped rectangles. For the three-dimensional case, one must consider the body of rotation which is obtained by rotating the capped rectangle and its excluded area.

The result (24) deserves some discussion. While for spheres and parallel objects the excluded volume is just the object's volume multiplied by a constant, the excluded volume of the capped cylinder [Eq. (24)] is not proportional to its volume $[(4\pi/3)(W/2)^3+\pi(W/2)^2L]$. Hence, the criterion of a constant total occupied volume is not compatible with the criterion of a constant total excluded volume [Eq. (24)] for the general random case. To be more specific, let us consider the dependence of the critical concentration on W and L in the $L \gg W$ case. A true volume criterion[1,4] would give

$$N_c \propto 1/W^2L \ , \tag{25}$$

while according to the excluded volume (or the Onsager[9]) prediction

$$N_c \propto 1/L^2W \ . \tag{26}$$

Which of these relations is the "correct" one can be tested by computer experiments (see Sec. III).

Another question that arises involves the coefficient of proportionality in relations such as (26). Following the work on soft-core spheres[4,5] and the work of Skal and Shklovskii[8] for other soft-core objects, one would be tempted to assume the same excluded volume for all systems. This would mean that the value $(4\pi/3)W^3N_c \approx 3$ will be an invariant not only for parallel objects but also for randomly aligned objects. For example, in the isotropic case of long sticks this would mean that $2WL^2(\pi/4)N_c \approx 3$. As will be mentioned in Sec. III and discussed in Sec. IV, this is not the case. The excluded volume is found to determine the critical behavior but the numerical value is not an invariant beyond the all-parallel-object case.

III. COMPARISON OF PREDICTIONS WITH AVAILABLE DATA

Before proceeding with the comparison of the results obtained in Sec. II with Monte Carlo results reported in the literature, it is important to note that the excluded-area and excluded-volume arguments are not exact. This is unlike the truly universal values of critical exponents. The dimensional invariance cannot be expected to be better than 10% as can be concluded from the results of Scher and Zallen[1] and the results of Skal and Shklovskii.[8] Our interest in this paper is to develop general expressions for excluded areas and excluded volumes. The comparison must consist then of two steps. First, to find whether the predicted dependence of the percolation threshold on the object shape and the ensemble anisotropy is in agreement with the data, and second, to find whether there is agreement between the predicted and the "experimentally" determined numerical values. If the assumption of a universal excluded area (volume) is correct, the agreement between the general expressions and the Monte Carlo results should be within the 10% accuracy. Much larger disagreements indicate a limitation of the excluded-area (volume) argument.

Let us start with a comparison of the system of widthless sticks with a system of circles. Pike and Seager[4] found from a Monte Carlo study that the critical radius of a soft-core circle, in a system of N circles in a unit square, is $r_c=1.058r_s$ (where r_s is defined as $1/\sqrt{\pi N}$). This result is well established[5] within 5%. The critical excluded area of this circle is $A_{\text{ex}}=4\pi r_c^2N$. As we have seen above [Eq. (9)], the excluded area of the widthless stick in the isotropic case is $\langle A\rangle=(2/\pi)L_{ci}^2$. Using the Monte Carlo results[4] of $r_c=1.06r_s$ for circles and $L_{ci}=4.2r_s$ for sticks, we find that while the $\langle A_{\text{ex}}\rangle$ associated with the sticks is 3.57 the A_{ex} associated with the circles is 4.48. Computing L_{ci} from the latter value and the assumption $\langle A\rangle=A$, yield the value $L_{ci}=4.7r_s$. This is in contrast with the above well-established value[4,14] $L_{ci}=4.2r_s$. As will be suggested in Sec. IV this discrepancy is not accidental and is beyond the accuracy mentioned above. On the other hand, the dependence on the anisotropy, $P_{\parallel}/P_{\perp}[=\sin\theta_\mu/(1-\cos\theta_\mu)]$, as given by Eq. (11) is within the accuracy of the available Monte Carlo data.[14] In Fig. 5 we show that the predictions given by Eq. (11) and Eq. (12) are practically the same. The prediction in Eq. (12) was obtained from a topological argument which assumes a representative stick that makes an angle $\theta_\mu/2$ with the axis of anisotropy.[14] The proximity of the two results suggests that the dependence on the system parameters is obeyed more closely than the numerical value of $\langle A_{\text{ex}}\rangle$. (This point is exhibited clearly by the results mentioned below for three dimensions.) An independent Monte Carlo study[16] has also confirmed the prediction of Eq. (14). Again there is a full agreement within the accuracy of the Monte Carlo data. Monte Carlo computations have also been carried out[16] for the horizontal-vertical stick system. Again, the dependence given by Eq. (17) has been confirmed with an accuracy similar to that associated with the confirmation of Eqs.

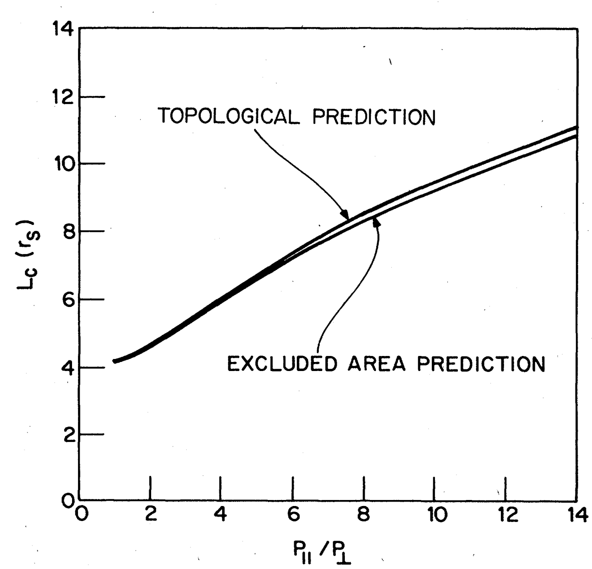

FIG. 5. Dependence of the critical length of a widthless stick on the macroscopic orientational anisotropy of the system. This dependence was calculated using the prediction of Ref. 14 [Eq. (12)] and the excluded-area prediction of Eq. (11).

(11), (12), and (14).

For the finite-width sticks not much data are available and the only comparison which can be made is with results obtained for parallel squares.[4,5] For this case $\theta_\mu=0$ and Eq. (19) reduces to $\langle A \rangle = 4L_c^2$. Since $A=4a$ for circles, one can check whether the excluded-area argument, $L_c^2=\pi r_c^2$, holds. As was pointed out already by Pike and Seager,[4] this relation is indeed correct since it is in good agreement (8%) with the Monte Carlo results.[17] Considering the above examples of two-dimensional sticks we may conclude then, that within the discussed accuracies, the excluded area is a universal invariant as far as the dependence of the threshold on the system parameters is concerned. It is also a numerical invariant for parallel objects but it does not appear to be a numerical invariant for randomly aligned objects. As we shall see below, these conclusions become firm when the three-dimensional system is considered.

Turning to the three-dimensional case we recall that the important predictions of our excluded volume result (found in Sec. II C) are that the dependence of $1/N_c$ on W will change from linear to cubic with increasing W, that the dependence on L will change from linear to quadratic with increasing L, and that N_c will be inversely proportional to $\langle \sin\gamma \rangle_\mu$. The confirmation of these predictions by Monte Carlo results[16] shows that the excluded volume and not the occupied volume of the object (which is proportional to W^2L) is the quantity which determines the percolation threshold. (It is only for parallel objects that the two arguments coincide.) Skal and Shklovskii[8] realized that the excluded volume is a fundamental dimensional invariant. However, since their work was concerned with parallel objects (the excluded volume of which is proportional to the volume of the objects) their argument could not be distinguished from a true volume argument. This is probably the reason why the distinction between the two types of volumes has not been stressed previously. Here, by taking an object, the excluded volume of which has a different shape than the object itself, we are able to show that such a distinction exists. Hence, the above-mentioned agreement between the present predictions and the Monte Carlo results[16] shows that the excluded volume is the more fundamental quantity to the extent that the determination of the percolation threshold is concerned.

While agreement was found in the dependences of the $1/N_c$ on W, L, and $\langle \sin\gamma \rangle_\mu$, there was a substantial discrepancy between the numerical values, obtained, for spheres and sticks. For example, in the Monte Carlo study[16] it was observed that for the isotropic long-stick ($\theta_\mu=\pi/2$, $L \gg W$) case

$$N_c 2WL^2(\pi/4) \approx 1.4 \ , \tag{27}$$

while for spheres we know that[5,7,16]

$$N_c(4\pi/3)W^3 \approx 2.8 \ . \tag{28}$$

While Eq. (27) is the only result available at this stage, it appears already that for randomly-aligned objects the total excluded volume needed for percolation is smaller than for the all-parallel nonelongated object system. It will be interesting to find out whether there is a common total excluded volume for the randomly aligned objects (e.g., in the isotropic case) in the $L \gg W$ limit as there is for the all-parallel objects[8] (≈ 3). At present we may conclude that there is no single constant for each dimension and that so far universal constants exist only for systems of all-parallel objects. This presumably reflects the fact that one cannot define a proper excluded volume, and the average quantity we compute apparently does not describe the system fully. It is still apparent from the above comparison that the excluded-volume criterion is very useful at least as far as the dependencies on the system parameters are concerned. In two dimensions the criterion appears, within reasonable accuracy, to be also useful for quantitative determination of the critical parameters. As will be discussed in Sec. IV, in both two and three dimensions this is an "exact" argument for the determination of the critical number of bonds per object.

In the above comparison we have discussed the effect of randomness and anisotropy of the object orientation on the excluded area (volume). Now that data are available on the effect of the stick-length distribution on the percolation threshold, one may try to apply an averaging process to this case and compare the calculated average excluded volume with these recent data. We have found that if proper averaging is applied to this case the object-size distribution does not alter the invariance of the total excluded area or volume. The considerations involved in this case are presented in Appendix B.

IV. DISCUSSION

The comparison made in Sec. III between the present results and the available data yields three principal conclusions.

(1) There is a dimensional invariance of the total excluded volume (area) for a system of all-parallel soft-core objects, i.e.,

$$\begin{aligned} &\text{(a)}\ \ \langle V_{\text{ex}} \rangle = N_c 8v \equiv C_3 \ , \\ &\text{(b)}\ \ \langle A_{\text{ex}} \rangle = N_c 4a \equiv C_2 \ , \end{aligned} \tag{29}$$

where C_3 and C_2 are constants.

(2) In a system of nonparallel objects, relations (29) are not fulfilled but $N_c\langle V \rangle$ and $N_c\langle A \rangle$ are independent of the degree of anisotropy, i.e., $\langle V_{\text{ex}} \rangle$ and $\langle A_{\text{ex}} \rangle$ are invariants for soft-core objects of a given shape.

(3) In a system of nonparallel objects relations (29) should be replaced by the inequalities

$$\begin{aligned} &\text{(a)}\ \ \langle V_{\text{ex}} \rangle < C_3 \ , \\ &\text{(b)}\ \ \langle A_{\text{ex}} \rangle < C_2 \ , \end{aligned} \tag{30}$$

where the deviations from Eq. (29) are larger for case (a).

It is apparent from the three conclusions that we must classify the degree of the invariance of the excluded volume (area) according to two classes: a class where there is a *dimensional invariance* [Eq. (29)] and a class where there is only a *system invariance.*

In view of the two degrees of invariance (as manifested by the existence of the above classes), the question arises whether there is still any other quantity which is a more general invariant. The only other suggestion of such a dimensional invariant is that of the average number of sites (or objects) bonded to a given site (or object) at the percolation threshold, B_c. Shante and Kirkpatrick[3] (using the fact that for site percolation on lattices B_c tends to a well-defined limit with increasing coordination number) suggested that in the continuum case B_c will be a "dimensional invariant." They believed that "the existence of this invariant permits a very powerful extension of the predictions of percolation theory to situations in which a regular lattice is no longer defined."

A close examination of the site-percolation B_c concept in the above *continuum* soft-core cases shows that this quantity is both conceptually and numerically the same as the present quantity of the total excluded volume (area). This conclusion follows from the argument that since B_c is the average number of bonded objects per given object, it is also the average number of centers of objects which enter the excluded volume of a given object. Hence, this number is the density of centers N_c (in a unit cube), times the average excluded volume of an object $\langle V \rangle$, i.e.,

$$B_c = \langle V \rangle N_c \equiv \langle V_{ex} \rangle \ . \quad (31)$$

Indeed, this relation is confirmed by the available Monte Carlo data not only for the simple cases of circles and spheres but also for the systems for which we have used our averaging procedure [Eqs. (7) and (A5)]. In Sec. III we found the following $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ values: For circles, $\langle A_{ex} \rangle = 4.48$, while Monte Carlo results[4] for B_c are between 4.48 and 4.53. For spheres, $\langle V_{ex} \rangle = 2.8$, while the Monte Carlo results[4,16] for B_c are between 2.70 and 2.92. Turning to the widthless sticks we found that $\langle A_{ex} \rangle = 3.57$ while the Monte Carlo results[4,15] show that B_c is between 3.63 and 3.7. For the three-dimensional sticks we found that $\langle V_{ex} \rangle = 1.41$, while our Monte Carlo results[16] show that $B_c = 1.49$.

Now that relation (31) has been confirmed we can conclude that B_c has the same degree of invariance as $\langle V_{ex} \rangle$ (or $\langle A_{ex} \rangle$) and that there does not seem to be a more universal quantity than the excluded volume (area). Another immediate conclusion is that in systems of randomly aligned particles, there are fewer bonded objects per given object (1.4) than in the all-parallel or spherical objects case (2.8). This is contrary to the intuitive suggestion[11] that "since the surface of an elongated particle is much larger than that of a sphere of equal volume, so numerous contacts can occur on a single fiber."

The relation (31) and the available Monte Carlo data, for systems in which the size of the objects is not a constant, enable an important consequence regarding the invariance of the excluded volume (area). The Monte Carlo data[4] have shown that for soft-core objects, widening the object-size distribution brings about a decrease in B_c. For example, it was found that for circles of variable radius $B_c = 4.01$ (instead of 4.5), and for spheres of variable radius $B_c = 2.17$ (instead of 2.8). For widthless sticks it was explicitly shown[15] (for the uniform distribution of the stick length) that B_c decreases with increasing width of the distribution. On the other hand, as shown in Appendix B, a proper averaging procedure of the excluded area and volume shows that $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ are dimensional invariants under variable distributions of the object sizes. We see then that while $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ are dimensional invariants, the B_c values are not. (See, however, Appendix B for the limits of B_c.)

The above conclusion brings up the question whether we can say that B_c is also the less "fundamental" quantity (from the invariance point of view) for a system composed of equal-size objects. The answer to this question can be gathered by examining the hard-core cases. In Table I we show data for B_c as given in the literature and the values obtained from the present discussion for A_{ex} and V_{ex}. It is seen in the table that the two quantities are identical indeed in the soft-core continuum cases, they are close in the hard-core continuum cases and they are different in the hard-core lattice cases. There is, however, a systematic behavior of the B_c values, as to be expected from the less efficient packing of the hard-core circles (spheres) in the continuum. The smaller the lattice coordination number the smaller the B_c value; the smallest B_c value in the lattices approaches the B_c value in the continuum. On the other hand, the values of A_{ex} and V_{ex} for both the continuum and the lattices appear to be roughly the same. Hence, correlations associated with the lattice structure affect B_c to a much larger degree than they affect A_{ex} and V_{ex}. We may conclude then that as far as invariance is concerned, the excluded area (volume) concept is "more universal," and the property of invariance may be considered to be "more related" to this concept than to the B_c concept.

In the above discussion one must note that the term "system" must be well defined. For example, in our capped-cylinder cases, with decreasing aspect ratio or with increasing anisotropy, the capped-cylinder system behaves as a system of spheres or all-parallel-object system rather than a randomly-aligned long-object system. Hence, in the context of the excluded volume one must characterize quantitatively the system of capped cylinders. We can do this by considering the two limits of Eq. (24). The first limit is that of parallel or spherical objects $[(4\pi/3)W^3 + 2\pi W^2 L \gg 2WL^2 \langle \sin\gamma \rangle_\mu]$ and the other limit is that of the randomly aligned long objects (the reverse inequality). For intermediate cases we know then that the value of $\langle V_{ex} \rangle$ lies between the values which correspond to the

TABLE I. Monte Carlo values of B_c, A_{ex}, and V_{ex} for circles and spheres. The results for B_c were taken from Refs. 2, 4, and 16. In the case of the continuum hard-core circles, the B_c values were obtained by extrapolating the data of Refs. 4 and 17 for the dependences of r_c (the critical circle radius) and B_c on the radius of the internal hard-core circle, r_{hc}. The arrows indicate the variation of B_c with decreasing coordination number.

System	B_c	A_{ex} or V_{ex}
Continuum, soft-core circles	4.5	4.5
Continuum, soft-core spheres	2.8	2.8
Continuum, hard-core circles	2.0±0.2	2.2±0.4
Continuum, hard-core spheres	1.8	1.4
Lattice, hard-core circles	3→2	1.8
Lattice, hard-core spheres	2.5→1.7	1.2

two limits (2.8 and 1.4 in the above example). The transition between the two limits has been demonstrated by recent Monte Carlo computations.[16] A loose definition of the system may lead to "surprising" Monte Carlo results such as that found[18] for aspect ratios (L/W) smaller than 15. The observation was that the dependence found was $N_c \propto (L/W)^{-1}$ rather than the dependence $N_c \propto (L/W)^{-2}$ (found on composites[11] and expected from the present considerations [Eqs. (26) and (27)]). The reason for this apparent discrepancy becomes clear if one examines Eq. (24) and notes that the ratio between the last two terms is $L/4W$, i.e., that the $L/W \leq 15$ range is an intermediate region in which the $N_c \propto (L/W)^{-1}$ relation appears to be a better fit to the data. Indeed, a recent Monte Carlo study[16] has shown that for larger aspect ratios the expected dependence $N_c \propto (L/W)^{-2}$ is revealed.

Finally, let us examine the invariance associated with the percolation thresholds, in the continuum, in view of the present results. The Scher and Zallen[1] invariance for hard-core spherical objects is empirical, and there is no known *a priori* reason for it to hold as well as it does. Once such invariance relationships do hold, one would like to know how general they are and, if possible, to explain deviations from the relationships in cases where they do occur. The problem we consider here differs from all previous studies in two respects: The shapes of the actual excluded volumes are much less symmetric (e.g., capped parallelepipeds), and they have a wide spread in their sizes and orientations. We express our critical conditions in terms of an average excluded volume (area), completely disregarding the effect of the large spread of the excluded volumes and of the implied correlations. The results indicate that the average behavior of the systems where such a spread occurs is somehow more effective in producing continuum percolation paths than for systems in which no spread occurs. From our findings (Appendix B) that the longer sticks or larger circles should be given a larger weight (in producing such paths), we may conclude that the larger excluded volumes contribute to the onset of percolation to a larger extent than can be gathered from the value of their volume. Hence, the total excluded volume needed for the onset of percolation always decreases with increasing degree of randomness, as indeed confirmed by the Monte Carlo results. In view of this we believe that the decrease in the total average excluded volume with increasing degree of randomness is related much more to the replacement of the (e.g., angle-dependent) distribution of excluded volumes by its average, than to the actual shape of the excluded volume. In principle, one may confirm this by a Monte Carlo investigation of the percolation threshold for parallel but anisotropic objects with a proper shape.

In conclusion, we have found that the excluded volume is a dimensional invariant for continuum systems of objects where the only randomness is in their location in space. Increasing the degree of randomness by allowing variable orientation of the objects lowers the average excluded volume (and the corresponding percolation threshold) to a value which is system invariant. Another increase in the degree of randomness, by allowing objects of different sizes but of the same shape, does not cause a variation in the total excluded volume. On the other hand, the average number of bonds per object decreases with the increase of this kind of randomness.

Note added in proof. Using the definition (B.4) one can show rigorously that if the critical total excluded volume is given by $\alpha N_c \langle L^k \rangle^{d/k}$ where α is a constant, d is the dimensionality of the system and k is positive than it must be that $k \geq d$. For the examples considered here this means that while the averages (B3) and (B18) are plausible the averages (B2) and (B17) are not. This can be proved by considering an objects system composed of two distributions with concentrations N_1 and N_2, and averages $\langle L^k \rangle_1$ and $\langle L^k \rangle_2$, respectively. If more objects are added to the system the total excluded volume

$$V_{1,2} = \alpha (N_1 + N_2)^{1-d/k} (N_1 \langle L^k \rangle_1 + N_2 \langle L^k \rangle_2)^{d/k}$$

should increase. In order for the derivative of $V_{1,2}$ with respect to either N_1 or N_2 to be non-negative, for every possible distribution, one must have $k \geq d$.

APPENDIX A: THE AVERAGE OF $\sin\gamma$

The average of $\sin\gamma$ is the average of $|\vec{u}_i \times \vec{u}_j|$ or of $[1-(\vec{u}_i \cdot \vec{u}_j)^2]^{1/2}$ when $\vec{u}_i$ and $\vec{u}_j$ are unit vectors along the axes of the corresponding sticks. We can then define the function

$$f(\theta_i,\theta_j,\phi_i,\phi_j) = [1-(\vec{u}_i \cdot \vec{u}_j)^2]^{1/2} , \tag{A1}$$

where:

$$\vec{u}_i \cdot \vec{u}_j = \sin\theta_i \sin\theta_j \cos\phi_i \cos\phi_j + \sin\theta_i \sin\theta_j \sin\phi_i \sin\phi_j + \cos\theta_i \cos\theta_j . \tag{A2}$$

We must integrate over the proper solid angles in order to find the average of $f(\theta_i,\theta_j,\phi_i,\phi_j)$. For this purpose let us define the function

$$g_f(\theta_i) = \int_{-1}^{\cos(\pi-\theta_\mu)} d(\cos\theta_j) \int_0^{2\pi} d\phi_i \int_0^{2\pi} f(\theta_i,\theta_j,\phi_i,\phi_j) d\phi_j + \int_{\cos\theta_\mu}^{1} d(\cos\theta_j) \int_0^{2\pi} d\phi_i \int_0^{2\pi} f(\theta_i,\theta_j,\phi_i,\phi_j) d\phi_j . \tag{A3}$$

We may further define the function $g_1(\theta_i)$, which is obtained by setting $f=1$ in Eq. (A3). It is readily found that $g_1(\theta_i) = 8\pi^2(1-\cos\theta_\mu)$.

The integrals needed for the average are I_μ and ψ_μ.

The first integral is given by

$$I_\mu=\int_{-1}^{\cos(\pi-\theta_\mu)} g_f(\theta_i)d(\cos\theta_i) + \int_{\cos\theta_\mu}^{1} g_f(\theta_i)d(\cos\theta_i) \ , \quad (A4)$$

while ψ_μ is obtained by substituting $g_f(\theta_i)$ by $g_1(\theta_i)$. The latter substitution yields that $\psi_\mu=16\pi^2(1-\cos\theta_\mu)^2$. Hence, the *general* average of $\sin\gamma$ is given by

$$\langle \sin\gamma \rangle_\mu = I_\mu/\psi_\mu \ . \quad (A5)$$

The integral I_μ is too complicated for a general analytic result to be derived. We may obtain, however, a lower bound by considering the two-dimensional average $\langle \sin\theta \rangle_\mu$ given by Eq. (8). The reasoning behind this approximation is that one may consider one stick with its direction fixed in space, e.g., $\theta_j=0$, then calculate the excluded volume it makes with a stick which makes an angle θ_i with it, and finally perform a three-dimensional average over all possible θ_j axes. Of course, this procedure neglects some of the solid angles which are formed by the possible combinations of θ_i, θ_j, ϕ_i, and ϕ_j. If we compare, however, the values that we have derived numerically for I_μ/ψ_μ and those derived from Eq. (8) we see that this approximation is quite good and it yields the empirical relation

$$\langle \sin\gamma \rangle_\mu \approx 1.25 \langle \sin\theta \rangle_\mu \quad (A6)$$

in the interesting regime of anisotropies. For example, for $\theta_\mu=0$ both (A5) and (8) yield $\langle \sin\gamma \rangle_\mu=0$. For $\theta_\mu=\pi/6$, (A5) yields 0.44 while (8) yields 0.35. For $\theta_\mu=\pi/4$ the corresponding values found are 0.60 and 0.46, and for $\theta_\mu=\pi/2$ the corresponding values are 0.78 and 0.64. We further note that in the isotropic, $\theta_\mu=\pi/2$ case an analytic solution has been found,[9] and it is

$$\langle \sin\gamma \rangle_\mu = \pi/4 \ . \quad (A7)$$

It is worth noting that the numerical integration is quite tedious for the evaluation of I_μ. A much easier numerical method is to simply make a Monte Carlo average by taking a large number of random four-number sets $(\cos\theta_i, \cos\theta_j, \phi_i, \phi_j)$ and computing (A2) for all these sets. We have found that with 10 000 sets the accuracy is good to the third digit (e.g., 0.784 for $\pi/4$).

APPENDIX B: AVERAGES OF LENGTH DISTRIBUTIONS

In all the calculations of Sec. II we have assumed that all the sticks in the ensemble have the same size. We can easily extend these calculations to cases where the stick lengths are distributed in a given form. Here we consider only the cases for which Monte Carlo data is available, i.e., for ensembles of widthless sticks in which the stick-length distribution [or fiber-length distribution[14] (FLD)] is independent of the stick-orientation distribution [or fiber-orientation distribution[14] (FOD)]. A more rigorous and detailed account of this problem is planned to be discussed elsewhere.

Returning to the definition of $\langle A \rangle$ [Eq. (7)] with the intention of carrying out an average over a distribution of lengths, we encounter a problem since the generalization of

$$\langle A \rangle = L^2 \langle \sin|\theta_i-\theta_j| \rangle \quad (B1)$$

can be either

$$\langle A \rangle = \langle L \rangle^2 \langle \sin|\theta_i-\theta_j| \rangle \quad (B2)$$

or

$$\langle A \rangle = \langle L^2 \rangle \langle \sin|\theta_i-\theta_j| \rangle \ , \quad (B3)$$

where

$$\langle L^n \rangle = \int L^n P(L) dL \ , \quad (B4)$$

and $P(L)$ is the stick-length distribution function.

The average (B2) has the merit of following the simple construction used to derive Eqs. (1) and (B1) from the construction shown in Fig. 1. Furthermore, the average number of bonds per object is expected to be associated with the area defined by the two intersecting objects [see Eq. (31)]:

$$B_c = N_c \langle A \rangle \ . \quad (B5)$$

In contrast, Eq. (B3) does have a "self"-square associated with it, the geometrical meaning of which is less transparent than that of Eq. (B2).

On the other hand, (B3) is favorable from the Scher-Zallen—type approach where the self-area of the object rather than its "interaction" area is considered. Another point in favor of (B3) (or similar higher moments of L) is the expectation and the confirmation that the larger sticks determine the percolation threshold (while, for example, in a broad distribution with many small sticks the smaller sticks are unimportant). Another difficulty with Eq. (B2) is that for some distributions $\langle L \rangle$ is independent of the width of the distribution, in contrast with the expected importance of influence of the larger sticks. Two such distributions for which Monte Carlo computations have been carried out are the normal distribution[14]

$$P(L) = (2\pi\sigma^2)^{-1/2} \exp[-(L-L_M)^2/2\sigma^2] \ , \quad (B6)$$

where L_M is the mean and 2σ is the width, and the uniform distribution[15]

$$P(L) = 1/2f \ , \quad (B7)$$

where L is confined to the interval $L_M-f \le L \le L_M+f$. L_M is the mean and f ($\le L_M$) is the width of the distribution. On the other hand, the second moment $\langle L^2 \rangle$ depends on the width yielding

$$\langle L^2 \rangle = L_M^2 + \sigma^2 \quad (B8)$$

for the normal distribution, and

$$\langle L^2 \rangle = L_M^2 + f^2/3 \quad (B9)$$

for the uniform distribution.

Let us examine the Monte Carlo results reported in the literature which may reveal the applicability of (B2) or (B3) for the determination of the percolation threshold, i.e., which of the $\langle A \rangle$'s fulfills the relation of invariance

$$N_c \langle A \rangle = C \ , \tag{B10}$$

where C is a constant. Since the normal distribution (B6) has been applied previously[14] to a narrow-distribution case ($\sigma = L_M/4.2$), the average L_M obtained (in a sample of fixed number of sticks N) at the threshold, while being somewhat lower than $L_c = 4.2r_s$ (see Sec. III), is within "experimental" accuracy in agreement with both (B2) and (B3). The case of the uniform distribution on the other hand, which was considered in the literature[15] for various values of f, has shown clearly agreement with (B3) and disagreement with (B2). The Monte Carlo results[15] have also clearly shown (unlike the case of equal size objects) that the relation (B5) [or (31)] is invalid when there is a length distribution of the sticks. It was further found that B_c is not distribution independent, indicating, as we have suggested in Sec. IV, that an excluded-area—type average is a more fundamental quantity (from the invariance point of view) than the average number of bonds. On the other hand, from the Monte Carlo study[15] and from other data to be mentioned below it appears that the B_c values are bounded by the values suggested by the total areas $N_c\langle A \rangle_2$ and $N_c \langle A \rangle_3$, where the subscripts refer to averages according to (B2) and (B3), respectively. Hence

$$N_c \langle A \rangle_2 < B_c < N_c \langle A \rangle_3 \ . \tag{B11}$$

A less trivial distribution, which yields width-dependent averages for both (B2) and (B3) is that of the log-normal distribution of width 2σ and a mean $\ln L_M$. This distribution, which is defined by

$$P(\ln L) = (2\pi\sigma^2)^{-1/2} \exp[-(\ln L - \ln L_M)^2/2\sigma^2] \ , \tag{B12}$$

yields the averages

$$\langle L \rangle = L_M \exp(\sigma^2/2) \ , \tag{B13}$$

and

$$\langle L^2 \rangle = L_M^2 \exp(2\sigma^2) \ . \tag{B14}$$

If the excluded area is an invariant under different distributions, and if we use a sample of a given stick concentration N, we must obtain that $\langle L \rangle^2 = L_{ci}^2$ [according to (B2)] or that $\langle L^2 \rangle = L_{ci}^2$ [according to (B3)], where $L_{ci} = 4.2r_s$ the critical stick length found for equal-length sticks (see Sec. III). For $\sigma = (\ln 10)/2$ the two averages yield $L_M = 2.1r_s$ and $L_M = 1.1r_s$, respectively. The value obtained in the Monte Carlo study[14] was $L_M = (1.1 \pm 0.1)r_s$, in excellent agreement with the $\langle L^2 \rangle$ average. This is very convincing evidence for the invariance associated with Eq. (B3) since the distribution considered is very wide and the predictions based on the two averages are significantly different and are much more distinct than those obtained by using uniform distributions.[4,15]

The next question which arises is whether the above conclusions are special to the stick system or are they more general. Examining the Monte Carlo data[4] for circles and spheres indicates that these conclusions are general indeed. For circles the averages (B2) and (B3) for the uniform distribution [Eqs. (B7) and (B9)] take the form

$$\langle A \rangle = \pi \langle (r_i + r_j)^2 \rangle = 4\pi r_M^2 + 2\pi f^2/3 \tag{B15}$$

or

$$\langle A \rangle = \pi \langle r_i^2 \rangle = 4\pi r_M^2 + 4\pi f^2/3 \ , \tag{B16}$$

respectively. Here r_i and r_j are the radii of the "interacting" circles and r_M is the mean of the distribution of these radii. Again, if invariance is considered, then $\langle A \rangle = 4\pi(1.06r_s)^2$ as obtained for equal radius circles (see Sec. III). For the distribution taken in the literature,[4] $f = r_M$, it is expected that the critical value will be $r_M = 0.98r_s$ according to (B15) and $r_M = 0.92r_s$ according to (B16). The value obtained by the Monte Carlo computation[4] was $r_M = 0.93r_s$, again in excellent agreement with the average of type (B3). The B_c value was found to depend on the distribution width and fulfill relation (B11). Its Monte Carlo B_c ($=4.01$) is indeed between the value 4.5 [obtained for equal-radius circles and expected from (B3)] and the value 3.94 [obtained by using the Monte Carlo result for r_M in Eq. (B16)].

Following the above discussion it is worthwhile checking whether the above conclusions apply to higher dimensions. Here the only Monte Carlo data available is for spheres having a uniform distribution with $f = r_M$. The expected excluded volumes according to (B2) and (B3) are

$$\langle V \rangle = (4\pi/3) \langle (r_i + r_j)^3 \rangle = (8\pi/3)\langle r_i^3 \rangle + 8\pi \langle r_i^2 \rangle \langle r_j \rangle \tag{B17}$$

and

$$\langle V \rangle = (32\pi/3) \langle r_i^3 \rangle \ , \tag{B18}$$

respectively. These distributions for the case $f = r_M$ yield, correspondingly,

$$\langle V \rangle = 16\pi r_M^3 \tag{B19}$$

and

$$\langle V \rangle = (64\pi/3) r_M^3 \ . \tag{B20}$$

Taking $\langle V \rangle = (32\pi/3)r_c^3$ with the value $r_c = 1.41r_s$ obtained[4] for equal-radius spheres, we get that $r_M = 1.22r_s$ according to (B19), while $r_M = 1.13r_s$ according to (B20). The latter value is again in excellent agreement with the Monte Carlo value,[4] $1.131r_s$. Also in agreement with the conclusions reached in two dimensions for B_c we see that its Monte Carlo value[4] $B_c = 2.17$ lies between $B_c = 2.8$ [the expectation according to (B20); see Sec. III] and $B_c = 2.11$ [according to (B19)].

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