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Cluster structure and conductivity of three-dimensional continuum systems

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While the cluster statistics exponents for three-dimensional continuum systems have been shown to be the same as those of lattices, the cluster structure and the conductivity exponents of such continuum systems have not been reported before. Here we present the first determination of both the fractal dimension of clusters, D, and the conductivity exponent, t, for these systems. We further describe the "porcupine"-like morphology of the clusters and the conductivity behavior in continuum anisotropic systems.

In recent studies it has been established that critical behavior of percolating two-dimensional continuum systems,[1] as well as their physical properties,[2] are the same as those of lattice systems. This universal behavior is expected then to hold also in three dimensions. Indeed, a study[3] which has considered the percolating clusters statistics for a three-dimensional continuum system has shown that the corresponding critical exponents, β, γ, τ, and ν are the same as those obtained for three-dimensional lattices.[4] On the other hand, no study of clusters structure (dimensions and morphology) and of a physical property (such as resistivity) has been reported for three-dimensional continuum systems.

In the present Rapid Communication we would like to report such a study in order to establish that the universality applies beyond the cluster statistics and in order to present completely new information that is obtained for three-dimensional anisotropic continuum systems.

For that purpose we have used a Monte Carlo procedure which is in principle (but not in detail) similar to that reported before for two- (Refs. 2 and 5) and three- (Ref. 6) dimensional systems. We have randomly implanted (one by one) N capped cylinders, all having a length L and radius r, in a unit cube. Correspondingly, all the lengths to be mentioned below are given in this unit. The cylinders considered[6] are allowed to penetrate each other. Such interpenetrating or overlapping capped cylinders will be called here intersecting "sticks." The orientation of a "stick" is defined by the angle it makes with the axis of uniaxial symmetry, z. We have used[6] a uniform random distribution of angles between the two limits $-\theta_\mu$ and θ_μ. Hence, the isotropic case is given by $\theta_\mu = \frac{1}{2}\pi$ and the smaller the θ_μ the larger the anisotropy. The macroscopic anisotropy $P_\parallel / P_\perp$ is defined as in the two-dimensional case[5] yielding for the above described random orientation

$$P_\parallel / P_\perp = \cot(\theta_\mu / 2) \quad . \tag{1}$$

For each stick "thrown" into the cube, its intersection with all previous sticks is checked. If it does not intersect any other stick it is given a cluster number, while if it does intersect, it is given the cluster number of the stick it has intersected. If a stick makes two previously separated clusters join, the joint cluster receives one of the previous cluster numbers. For the cluster statistics and cluster structure study the computer stores the following information for every N: the number of sticks in every cluster, s, the number of clusters of size s, N_s, and the center (x_i, y_i, z_i) of each stick belonging to a given cluster. Using this information the program computes the center of gravity of each cluster (x_c, y_c, z_c) and its radius of gyration[7]

$$R_s = \left(\sum_{i=1}^{s} r_i^2 / s \right)^{1/2} , \tag{2}$$

where $r_i^2 = (x_i - x_c)^2 + (y_i - y_c)^2 + (z_i - z_c)^2$. We have also introduced a new density profiling check by counting the sticks between parallel planes [e.g., $x = x_c + (n-1)a$ and $x = x_c + na$] so that this "slicing" provides information concerning the "mass distribution" in the cluster. Finally, the critical stick concentration for the onset of percolation, N_c, is the smallest value N for which there is a cluster that intersects the opposite boundaries. Because of the finite samples used, there is a statistical deviation between the values of N_c obtained in the different directions.[6] In what follows we shall use the notation $N_{c\parallel}$ for the threshold along the z axis, and $N_{c\perp}$ for the threshold along the x axis.

For the determination of the resistance of the cube along the z axis, $R_\parallel$, and perpendicular to this axis, $R_\perp$, we have attached a unit resistor to each intersection of two sticks in the percolating cluster. This cluster contains N_p sticks. Hence, as in our two-dimensional study,[2] the stick is assumed to be an equipotential and the resistor network made of the unit resistors is resolved by the well known matrix representation.

The assumption made needs some elaboration since the distribution in the values of the resistors which can take place in "real" continuum systems is unaccounted for. One reason for such a distribution is that the resistance between two conducting particles may depend on the degree of their overlap. While no analog experiment or theoretical model deals with this problem we note that at least for some composites[8,9] (for which a one-point tunneling contact has been suggested) our assumption approximates the real situation. Another reason for having a distribution in the values of the resistors is the distribution of distances between intersections of one conducting particle by other particles. That such a distribution does not affect the critical behavior of the resistance has been demonstrated, for two-dimensional systems, by an analog experiment.[2] The emphasis in this work, however, is on the "correlation" aspect of the continuum systems described. We know that a percolating system in the continuum can be presented by a correlated lattice.[10] In both the lattice and the continuum, we have random resistors networks. The question which is addressed here is how will the correlations in the resistors network affect the values of the network's resistance and its critical behavior? In lattices, there are small discrete numbers of resistors which have a common junction, while in our continuum model there is a distribution of such numbers. In particular, it is interesting that the center of this distribution will depend on the aspect ratio of the particles involved. This is borne out by a recent observation[11] that, for a system of spheres close to the percolation threshold, the average number of spheres intersecting a given sphere is 2.8, while the same quantity for long ($L >> r$) sticks is 1.4. Correspondingly, the "average building block" of the spheres system is a three resistor junction, while the junction of the long stick has, on the average, half the number of resistors. Following the above considerations our results are expected to shed light on the "correlation" aspect of the continuum while the effect of the resistors values distributions deserves a separate study. Based on the much greater amount of data for two-dimensional systems, one expects, however, that both aspects of the continuum will not yield a different critical behavior from that found for lattices.

To examine consistency with the previous reports,[3] we have checked first whether the reported critical exponents are obtained in our isotropic system of three-dimensional sticks. Using double logarithmic plots (but determining the exponents by least-squares fit[2]) we found from the N_s dependence on s (at N just above N_c) that $\tau = 2.2 \pm 0.2$, from the dependence of N_p/N on $(N/N_c - 1)$ that $\beta = 0.4 \pm 0.1$, from the dependence of $(\Sigma N_s s^2/N)$ on $(N/N_c - 1)$ that $\gamma = 1.8 \pm 0.2$, and from the correlation length, i.e., the average[7] of R_s, that $\nu = 0.83 \pm 0.09$. These values are, within the experimental confidence limits, the same as those reported before.[3] The new observation, however, is that these values are obtained for spheres ($L << r$) as well as for elongated sticks, i.e., that they are independent of the aspect ratio of the corresponding sticks. This suggests that the critical behavior is independent of the type of objects of which the continuum system is composed.

The second search was for the dependence of the radius of gyration R_s [see Eq. (2) above] on s for N just above N_c. This search was done for both spheres and sticks in the isotropic three-dimensional system. Typical results for large (we have chosen $s \geq 10$) clusters[7] are shown in Fig. 1. The value obtained from the slope, i.e., $D = 2.5 \pm 0.5$ is in excellent agreement with the predictions[7] for the fractal dimension of large clusters, $D = d - \beta/\nu$, where d is the dimensionality of the system and β/ν, for the three-dimensional system, is about 0.5.

Having a sticks system enables the introduction of anisotropy into the system (see above). How will the introduced anisotropy affect the structure of the clusters? First, we found, as expected, that in isotropic systems the clusters have a spherical shape but the radius of gyration increases with increasing aspect ratio $L/2r$. The shape of the clusters in the anisotropic systems has been revealed by the density profiling described above. Typical results of such a sectioning study are shown in Fig. 2. The x's indicate the number of sticks between two successive planes parallel to the yz plane while the z's indicate the number of sticks between successive planes parallel to the xy plane.

A simple reconstruction which utilizes the sectioned profiles shown can yield the cluster cross section. While it is clear from the data presented in Fig. 2 that the cluster has a cylindrical shape the distribution of the sticks at the cylinder's ends, where the sticks are sparse, requires much larger clusters (and thus larger sticks ensembles) than those available in our study. Following this difficulty we have stu-

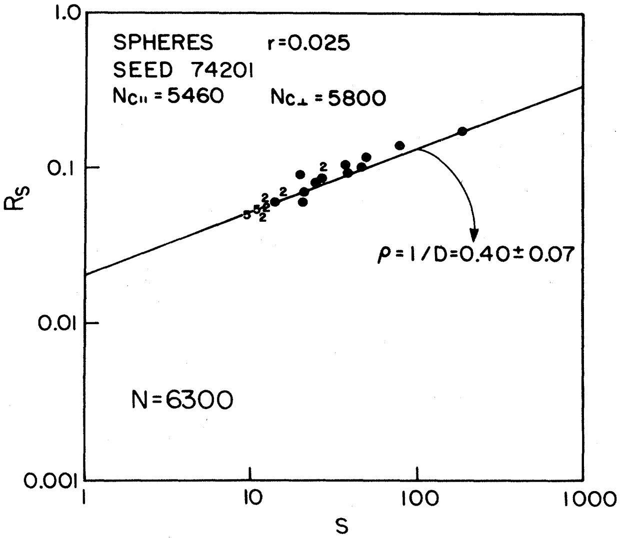

FIG. 1. Dependence of the radius of gyration of a cluster on its size. A point indicates the existence of one cluster of the corresponding s while a number indicates the number of clusters of the corresponding s and their averaged R_s.

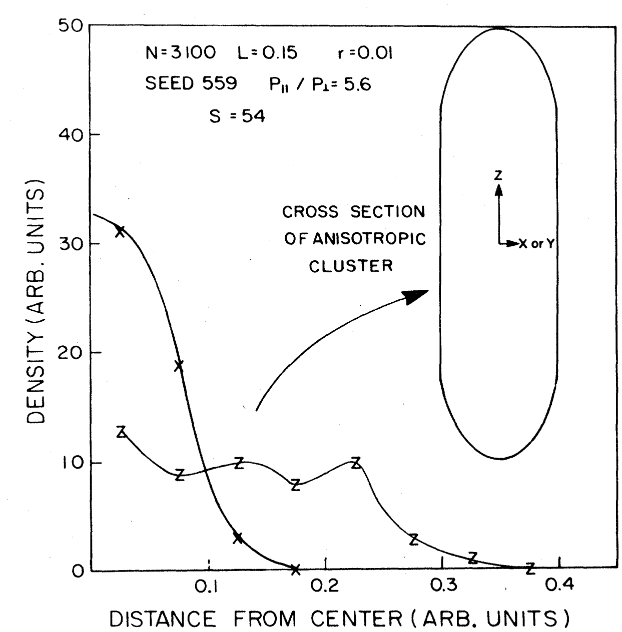

FIG. 2. Number of sticks centers between $x_c + (n-1)a$ and $x_c + na$ (the x's) and between $z_c + (n-1)a$ and $z_c + na$ (the z's) as a function of the distance $(n - \frac{1}{2})a$ from the cluster center (x_c, y_c, z_c). Here, $a = 0.05$ and $n \geq 1$.

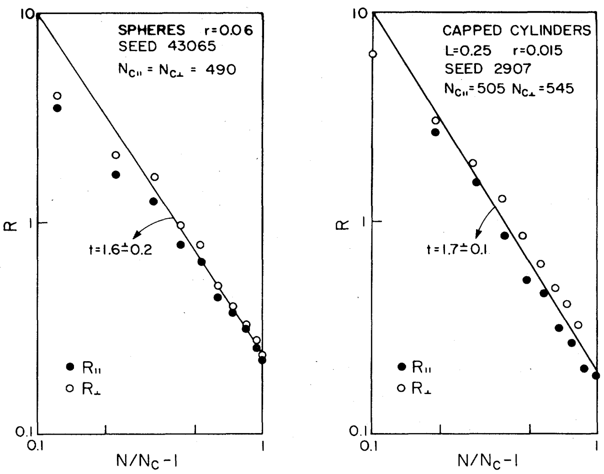

FIG. 3. Longitudinal and transverse resistance of two isotropic three-dimensional continuum systems as a function of the conducting particles concentration. The values of the exponents are derived by a least-squares-fit procedure.

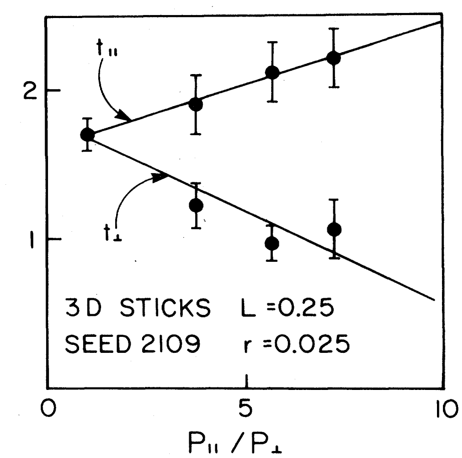

FIG. 4. Dependence of the apparent longitudinal ($t_\parallel$) and transverse ($t_\perp$) conductivity exponents on the macroscopic anisotropy of the system $P_\parallel / P_\perp$.

died the much larger clusters available for the two-dimensional systems of widthless sticks. We found that these clusters have a capped rectangle shape where the caps are parabolic. Hence, we have drawn the curve connecting the data points in Fig. 2 to be consistent with this shape. This result suggests that the shape of the three-dimensional cluster is the body of rotation of a capped rectangle. Correspondingly, we also show the cluster cross section which is in agreement with the data (reduced for convenience by a factor of 2) and is consistent with a capped cylinder shape. Of course, one has to recall that the clusters have a "swiss cheese" structure[7] and thus this overall shape is just the envelope of the cluster. The present clusters have also the special property that parts of the sticks may be piercing this envelope from within, yielding a "porcupine"-like morphology to the cluster. We further see that the bulk of the cluster has quite a uniform density and that its "skin" (where the density drops from the bulk value to zero) is almost one-quarter of its diameter. The important general property of the clusters in anisotropic systems, which is found here, is that they are elongated, but their aspect ratio is always smaller than the macroscopic anisotropy of the system. Hence, a sublinear dependence exists between these two quantities. More quantitative dependences of the cluster shapes on the various possible parameters $[L/r, \theta_\mu, s$, and $(N/N_c - 1)]$ will be reported elsewhere.

Turning to the resistance of the cubes, we have found (for many seeds) its dependence on the sticks concentration, as is shown in the examples of Fig. 3. The clear power-law behavior suggests that the critical exponent t can be obtained from the data. The t values shown in Fig. 3 and their confidence limits were determined using the least-squares-fit procedure of Ref. 2. The points to note are that for these isotropic continuum samples the t value is the same as that found for three-dimensional lattices,[7,12] that this value is independent of the sticks' aspect ratio, and that for the same $N/N_c - 1$ the resistance itself is almost independent of the aspect ratio or the "average building block" mentioned above. While the observed value of t confirms the expected universality, the latter point deserves some attention since one might expect that the longer sticks would conduct more "efficiently." This expectation is fulfilled if one considers the fact that much more "material" is needed for the N spheres than for the N sticks (see the r and L values in Fig. 3) to yield the same conductivity.

The effect of anisotropy on the resistance of a continuum system can be readily studied in our system of elongated particles (sticks). Plotting the $R_\parallel$ and $R_\perp$ data as in Fig. 3 for the various anisotropies has yielded, over the same $N/N_{c\parallel} - 1$ (or $N/N_{c\perp} - 1$) range, apparent power-law dependences. Hence, we have obtained an apparent critical exponent $t_\parallel$ which is associated with $R_\parallel$ and an apparent critical exponent $t_\perp$ which is associated with $R_\perp$. We have chosen this presentation of the data since it was used for the studies of anisotropic two-dimensional[12-14] and three-dimensional[12] lattices. In these studies it was found that $t_\parallel$ increases with the deviation from the percolation threshold and with increasing anisotropy while $t_\perp$ decreases under the same variations. As we show in Fig. 4 the same behavior is also obtained in the continuum system studied here. This finding, which has not been reported before for the two- and three-dimensional continuum, appears to be universal. Our results seem then to be also beneficial for the planning of real practical systems such as composites made of elongated particles.[2,9]

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