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## Critical Behavior of the Electrical Resistance and Its Noise in Inverted Random-Void Systems

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A computer simulation of a representative continuum system, the inverted random-void model in three and six dimensions, is reported. It is the first such simulation where the local geometry of the conducting particles is taken into account. The results show that the critical behaviors of both the electrical resistivity and the resistance noise, near the percolation threshold, are well described by the recently suggested models of links in the nodes-links-blobs picture.

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The prediction of Rammal and co-workers[1] regarding the universal behavior of the resistance noise in percolating networks has inspired many experimental studies on a variety of systems.[2-7] These systems include various thin films and composite materials in which the electrical properties appear to be associated with a network formed by the connection of conducting elements. At some critical density of the conducting-material phase, the concentration of the conducting elements is sufficient for the onset of percolation. However, all the experimental studies have shown a clear disagreement with the universal predictions of Ref. 1: While the electrical resistivity has exhibited the expected universal behavior, the relative resistance noise has exhibited a nonuniversal behavior. In particular, the values of the resistance noise exponent, $\kappa$, have been found to be much larger (2 to 5 times) than the values predicted in Ref. 1. Garfunkel and Weissman,[3] who noted that the experimental systems have a distribution of resistor values, have attributed this discrepancy to continuum corrections. Following them, Rammal[8] was able to predict such corrections, and Tremblay, Feng, and Breton[9] (TFB) presented a comprehensive theory for the continuum corrections that is capable of predicting the noise exponents in any given continuum model. Indeed, the TFB theory seems to qualitatively account for the experimental data as well as to establish the validity of the continuum corrections explanation. There is, however, considerable difficulty in making a *quantitative* comparison of the experimental data with the TFB theory. This difficulty is a result of the fact that the structural features of the tested experimental systems are usually not known in detail, and even when they are known in enough detail, the corresponding parameters (e.g., $u$ and $v$, see below), which are necessary for a comparison with the theory, have not been determined so far for any experimental system. Indeed, no quantitative comparison of experiment and the TFB model has been attempted. Furthermore, not only did each experimental work[2-7] report a different value for $\kappa$, but even within groups of seemingly identical systems[3,5] quite different $\kappa$ values were found. This indicates that the structural details are extremely important and one cannot compare experimental results with predictions of general or idealized models. This situation is in sharp contrast with the universal behavior (e.g., electrical resistivity in two-dimensional experimental systems[7]) where the critical exponents are independent of the structural details.

Following these considerations we applied a direct test of the TFB theory by considering an idealized "experimental system" which can be provided by a computer simulation. We have used for the test an inverted random-void system[9,10] and compared the results quantitatively with the predictions of the theory.[9] Another motivation for this work is the fact that a study by computer simulation makes possible a much more general test of the theory, since systems of dimensionality higher than 3 can be investigated. In particular, predictions[9,10] of transitions from a universal to a nonuniversal behavior of physical transport properties, as a function of the system dimensionality, can be examined. As far as we know, however, no previous simulation study of these properties, in which the local geometry of the conducting particles is taken into account, has been reported. In previous computer studies *predetermined* global conductance distribution functions of the system were used, and resistance values were randomly assigned to the resistor network. The topology of the network was determined separately and independently.[11-14] This was also the case in the computer simulation of the resistance noise, which was reported in Ref. 9 for a two-dimensional system. Hence, a simulation which better approximates real systems (see below) by assigning resistance values which are determined by the local structure of the network (and in particular for higher dimensions) seems to be long called for.

The inverted random-void model[9,10] chosen for our study is composed of conducting permeable spheres (hyperspheres) which are randomly distributed in space (hyperspace). In this model each intersection between two permeable spheres has its own resistance. The value of the resistance is determined by the degree of overlap of the intersecting spheres (see below). In the simulation we have used our previously reported procedure[14] for the "implantation" of permeable spheres of radius $a$ in a unit cube (hypercube). If two spheres have some overlap they are considered connected, and the onset of percolation is at a concentration of spheres, $N_c$, which is associated with the formation of a continuous path of connected spheres between opposite faces of the cube. The various "measured" properties of the system are then computed as a function of $N/N_c-1$, where $N$ is the density of spheres in the cube. In the present work we are interested in the resistance of the cube, $R$, and the corresponding relative resistance noise which is given by[9]

$$S_R=\left[\sum\langle\delta r^2\rangle i^4\right]\left[\sum ri^2\right]^{-2}. \tag{1}$$

Here $r$ is the resistance of the bond between two intersecting spheres, $\langle\delta r^2\rangle$ is the corresponding resistance-fluctuation-correlation function, $i$ is the current through the resistor (i.e., the bond), and the sum is over all the resistors in the network. One notes, of course, that for a unit current between the opposite plane electrodes of the cube (as will be assumed in the present work) the denominator in Eq. (1) is simply $R^2$.

If in a continuum model, such as the one used here, equipotentials are assumed throughout the intersecting region of two spheres (the so-called "neck"[9,10]), one can easily correlate both $r$ and $\langle\delta r^2\rangle$ with the neck geometry. This has been done for $r$ in Ref. 10, while for $\langle\delta r^2\rangle$ one can make the correlation by following[15] the argument given by Wright, Bergman, and Kantor[16] for a network of discrete resistors. The most concise argument, however, follows from the fact that the resistance fluctuations of the individual resistors add up randomly. Hence, as in random process, the squared variance, $\langle\delta r^2\rangle$, divided by the average squared, $\langle r\rangle^2$, is inversely proportional to the number of elements in the system. If the elements are the individual volume parts of a continuous slab of material of volume $V$, one obtains that

$$\langle\delta r^2\rangle/\langle r\rangle^2\propto 1/V. \tag{2}$$

One notes that $\langle r\rangle$ is an average over one resistor and thus it is the same as the $r$ given in Eq. (1). If the bond resistance depends only on one variable geometrical parameter $\epsilon$ such that

$$\langle r\rangle\propto\epsilon^{-u}, \tag{3}$$

and if the volume of the slab also depends only on $\epsilon$, such that

$$V\propto\epsilon^{v}, \tag{4}$$

one finds that

$$\langle\delta r^2\rangle\propto\epsilon^{-(2u+v)}. \tag{5}$$

In order to apply the general expressions given by Eqs. (3) and (5), one has to derive[9,10] the values of $u$ and $v$ for the system under consideration. In the present case this is done by our taking the slab discussed above as the "neck" region of the random-void system. Such a region is illustrated in Fig. 1. As in other cases of spreading resistance, the dominant resistance contribution is due to the narrowest portion of the neck. This part is well approximated[10] by a cylinder with a length of the order $2(a\epsilon)^{1/2}$ and a radius of the order of $(a\epsilon)^{1/2}$. Since only the parameter $\epsilon$ varies from one intersection (neck) to another, both $r$ and $V$ will depend only on this parameter. It is easily realized that in a space of $d$ dimensions, $r\propto\epsilon^{1/2-(d-1)/2}$ and $V\propto\epsilon^{d/2}$. Hence for the inverted random-void model the $u$ and $v$ values which should be substituted in Eqs. (4) and (5) are

$$u=d/2-1 \tag{6}$$

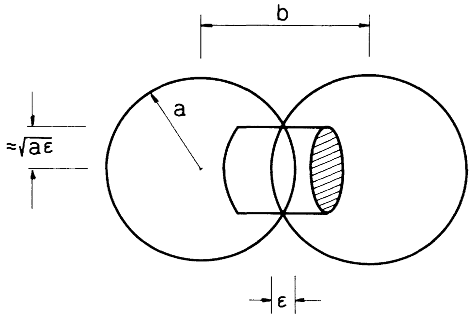


FIG. 1. The geometry of intersecting permeable spheres. The cylinder represents the region which yields the dominant contribution to the resistance of the corresponding "neck."

and

$$v = d/2. \tag{7}$$

In our simulation, $N$ permeable spheres (or hyperspheres) of radius $a$ are randomly implanted as described before. For each pair of intersecting spheres the distance between their centers $b$ is determined, and thus the value of $\epsilon$ ($=2a-b$) is recorded. Correspondingly, the values of $\langle r\rangle$ and $\langle \delta r^2\rangle$ are found for each intersection by use of Eqs. (3) and (5) (with a proportionality factor of 1). After knowing the $r$ values we find the currents in the system. This is done by our solving the Kirchhoff equation[17] for the network of the resistors, the values of which were determined as described above. In the present work, the equation was solved by use of a preconditioned conjugate-gradient algorithm[15] rather than our previous inverse-matrix method,[17] since the former enables the solution of much larger systems. The procedures for finding the percolation threshold and the exponent which fits the data were previously reported,[18] while the details of the above-mentioned algorithm will be presented elsewhere.[15] Finally, substituting the values compared for $r$, $\langle \delta r^2\rangle$, and $i$ in Eq. (1) we found $R$ and $S_R$ of the sample.

One notes that in our determination of $r$ and $\langle \delta r^2\rangle$ we use an idealization of real systems, since we estimate these quantities by the geometry of the cylindrical necks and consider multiple overlaps of spheres as separate overlaps. Close to the percolation threshold, however, these simplifications are expected to play a minor role. The neck approximation becomes better the smaller the overlap (i.e., the larger the resistance), and we know that close to $N_c$ the larger resistors determine the critical behavior.[9,10] The multiple overlaps also represent parallel connections of resistors which form "miniblobs." Since both the effect of the blobs and the average critical number of bonds per sphere[19] (4.5 in 2D, 2.8 in 3D, and 1.3 in 6D) decrease with dimensionality, the importance of these effects on the resulting values of $R$ and $S_R$ decreases with dimensionality and proximity to the threshold.

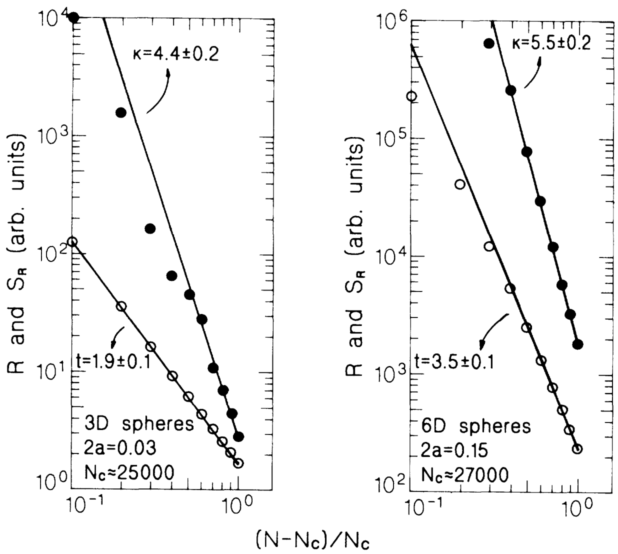


FIG. 2. The dependence of the sample resistance (open circles) and the relative resistance noise (filled circles) on the proximity of the percolation threshold in three- and six-dimensional inverted random-void systems. Each circle represents an average of ten different samples.

In the present Letter we show the results obtained for three- and six-dimensional inverted random-void systems. This is done in order to check some of the predictions of the continuum theories[9,10] in distinctly different cases. For example, one expects[10] a transition from a universal to a nonuniversal behavior of the electrical resistivity with increasing dimensionality. On the other hand, the electrical noise should[9] exhibit a nonuniversal behavior in systems of these dimensions. To make the best comparison possible we show here results which represent averages of ten large samples (of $N_c \approx 25\,000$ for 3D and $N_c \approx 27\,000$ for 6D). These are the largest samples which we could generate at present. In Fig. 2 we show the results which were obtained for the studied inverted random-void systems. The resistivity exponent derived from the data for three dimensions ($t=1.9 \pm 0.1$) is indeed the one expected for a universal behavior[16] ($t=2.0\pm 0.1$). The resistance noise exponent for three dimensions is found to have the nonuniversal value of $\kappa=4.4\pm 0.2$. This value is in excellent agreement with $\kappa=4.56$ $[=1.56+(v+2u-1)/u]$, the value predicted by the TFB theory.[9] We should point out that we have previously[15,19] confirmed the universal value of $\kappa=1.56$, which is expected from scaling considerations.[1,16] For the six-dimensional hypercontinuum the electrical resistivity exponent is found to be $t=3.5\pm 0.1$, compared to the value of 4.0 $[=3.0+(u-1)]$ to be expected[10] for such an inverted random-void system. The resistance noise exponent is found to be $\kappa=5.5\pm 0.2$, again agreeing well with the TFB result[9] for this case, $\kappa=6.0$ $[=2.0+v+1]$.

We see that our present results do indeed confirm the TFB theory and thus verify the validity of the links (of the nodes-links-blobs picture) analysis in determining the critical behavior of the electrical resistance and its noise. We note that our three-dimensional results for $\kappa$, while being in excellent agreement with the TFB theory, is in disagreement with the prediction of Rammal,[8] $\kappa = 3.7$ $[=2.67+(2u+v-2)]$. Since Refs. 8 and 9 differ only in the two-dimensional random void and the (identical) three-dimensional inverted random-void prediction for $\kappa$, we can state that our result supports the TFB analysis. (In other dimensions and models, the corresponding differences[16] between the predictions of Refs. 8 and 9 are just of the order of $\zeta_R - 1$.)

We note that our results for $t$ and $\kappa$ are somewhat smaller than expected for an infinite sample. This is not surprising, since finite-size effects weaken the divergence of critical behaviors near the threshold, in general,[20] and for electrical properties in particular.[14,18,21] Indeed, application of the simulations described here for smaller samples (average of ten samples of $N_c \approx 5000$) yielded $t = 1.8 \pm 0.1$ in three dimensions and $t = 3.2 \pm 0.1$ for six dimensions. Comparison of these values with the above values (Fig. 2) shows that for this quantity, the finite-size corrections are almost unimportant in three dimensions, and the values converge slowly to the expected value in six dimensions. The same procedure for $\kappa$ (for $N_c \approx 15000$) yielded the values $\kappa = 3.8 \pm 0.2$ in three dimensions and $\kappa = 5.0 \pm 0.2$ in six dimensions. Again, comparison of these values and those reported here (Fig. 2) indicates that the direction of convergence is towards the higher, expected values, as $N_c$ is increased. A more detailed discussion of the finite-size effects and their origin will be given elsewhere.[15]

The proximity of our results to the predictions of the TFB theory[9] shows that the computer simulations do indeed bypass the experimental difficulties which exist at present. In particular, the "sensitivity" of the noise to the current distribution[3] makes the experimental results more critically dependent (compared to the resistivity) on factors such as the true dimensionality of the system,[3,4] heating of the narrowest necks,[3] and parallel conduction channels.[5] The large difference between the critical exponents of seemingly similar experimental systems (compare the various results derived in Ref. 3) is a further demonstration of the sensitivity of the critical behavior to the exact neck-size distribution function. Our results indicate, however, that a true inverted random-void system should exhibit the theoretically predicted behavior. The experimental systems which closely resemble the inverted random-void system are those in which the onset of percolation is associated with the coalescence of conducting particles.[5,22,23] Indeed, a universal behavior of the electrical resistivity has been found in these systems,[22,23] and the suggested value of $\kappa$ derived for one of those systems,[5] the Mo-$Al_2O_3$ granular composite $\{\kappa = [(Q-2)t] = 4.6\}$, is very close to the predicted[9] and presently found value of $\kappa = 4.5$.

In summary, we have carried out a simulation of continuum systems in three and six dimensions. We found that the critical behavior of the electrical resistivity changes from universal to nonuniversal with increasing dimensionality. We have also confirmed (for dimensionality higher than two) that the inverted random-void systems will show the expected nonuniversal behavior of the resistance noise.

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