Limits on the continuum-percolation transport exponents

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(Received 25 November 1997)

The many experimental data that have been accumulated for the critical resistance exponent, t, and the relative resistance noise exponent, κ, in percolation systems, are generally in disagreement with the original predictions of the random void and the inverted random void models of continuum percolation. In this paper we show that by allowing a nonrandom distribution of the voids (or the particles) in these models, one can account for *all* the experimental data. In particular, we show that, except for the two-dimensional inverted random void system, the exponent t may have any value larger than its universal value, while the κ/t ratio will be bound. [S0163-1829(98)00121-0]

About 20 years ago, Kogut and Straley[1] showed that the transport properties of percolation systems may exhibit nonuniversal behaviors. The reason for that nonuniversality is the possibility of a diverging distribution of the high resistance values in the corresponding resistors network. A few years later, Halperin, Feng, and Sen[2] (HFS) showed that such a diverging distribution can be found in systems that can be modeled by a system of insulating spheres embedded in a continuous conducting matrix. The extension of this model to two-dimensional systems is obvious[2,3] and thus we define here ''real systems'' as measurable two- or three-dimensional systems. Such real systems then include sedimentary rocks, composite materials (where the conducting particles are much smaller than the insulating particles), and porous films. The model describing these systems is known as the random void (RV) model. The relative resistance noise of this model for two- ($d=2$) and three- ($d=3$) dimensional systems was predicted by Tremblay, Feng, and Breton[4] (TFB) to have a nonuniversal behavior. For the other model considered, i.e., that of conducting spheres embedded in a continuous insulating matrix, which became known as the inverted random void (IRV) model, a universal behavior for the resistance[3] was predicted, but its relative resistance noise for $d=3$ was shown to yield a nonuniversal behavior.[4] Sometime later, Balberg[5] showed that if the resistance between adjacent particles is determined by a tunneling process, a nonuniversal behavior of the resistance is also possible.[5] Since the presentations of those theories, numerous experimental data were accumulated.[6–22] In particular, the resistance exponent t and the exponents ratio κ/t, where κ is the relative resistance noise exponent, have been determined experimentally for many systems for which the RV or the IRV models should be good descriptions.[6–19] We note in passing that the tunneling model for the resistance[5] and the relative resistance noise[20] yields values that are not too different from those discussed in this paper. Hence, the limits found here also cover the corresponding experimental findings.[5,21,22] However, the expected tunneling mechanism in the latter systems calls for more specific predictions. The corresponding model and the systems considered in Refs. 21 and 22 will be discussed then in detail elsewhere.[20] In this paper we consider the systems that can be described by the RV or the IRV models.

Examining all the data given in Refs. 6–19, one notes two basic facts. First, there is a spread in the values of the exponents t and κ/t (which are far beyond the correction to scaling,[23] and second, there are many t and κ/t values that are much larger than the values predicted by the RV and the IRV models.[2–4] This is *a priori* surprising since the theories are not sensitive to the exact shape of the particles (or voids) in the system, provided they have curved surfaces, as is indeed the case for all the systems that were studied. In particular, two conspicuous deviations from the RV and IRV model predictions should be mentioned. In three dimensions, taking the universal value[24] of t to be $t_{un}=2$, the RV model[2,3] predicts t to have a value of 2.5 while the IRV model predicts that $t=t_{un}$. However, Pike[7] found a value as high as $t=5.01$ for a composite system for which the RV should be a good model, while Wu and McLachlan[19] found values of $t\approx 6$ for systems for which the IRV should be a good model. In two dimensions, taking the universal value of κ to be 1.12 and that of t to be $t_{un}=1.3$, the prediction[4] of the RV model was that $\kappa/t=3.16$ and the prediction of the IRV model was that $\kappa/t=0.86$. However, Garfunkel and Weissman[9] found κ/t values in the range 5.4–8.1 in sand-blasted films for which the RV or the IRV model should provide a good description. The explanation they gave for this conspicuous discrepancy was that their samples were nonuniform. In fact, in the many experimental works in which significant deviations from the theoretical predictions were found, the discrepancies have been attributed[23] to causes that are beyond the simple percolation theory.[24] For example, for the problem of the electrical noise, the causes suggested for the deviations included changes in the microscopic noise mechanism,[25] additional tunneling,[11] noisy hopping conduction,[26] noisy insulators,[27] heating,[28] and nonlinear effects.[22]

The purpose of the present paper is to show that *all* the many experimental results reported thus far can be accommodated within the framework of the original HFS and TFB theories if one of the original assumptions is removed, i.e., the assumption that the void sizes ε are distributed uniformly as $\varepsilon\to 0$. The removal of this assumption for *real* systems is very reasonable since during the natural (such as rocks[8]) or artificial (such as composites[11–13,19] or thin films[9,10,15,18]) formations of the systems, the acting forces may yield a more, or a less, ‘‘diverging’’ ε distribution than that of the exactly uniform one. In fact, for carbon black-polymer composites it has been shown[5] that there is a correlation between the dispersion of the conducting particles in the composite and the corresponding critical behavior of the resistance. We must emphasize that such correlations were suggested also for other systems,[2,29] but a single rule that can account for *all* nonuniform distribution and for *all* available experimental data has not been suggested. We also note in passing that our computer simulations[30] have confirmed that if the particles in the systems are randomly dispersed (for $2 \leqslant d \leqslant 6$), the predictions of the theories[2–4] of the RV and the IRV ideal models, for both t and κ, are fulfilled. This implies, as was suggested originally by HFS,[3] that the above uniform distribution as $\varepsilon \rightarrow 0$ is a good description of the *ideal* RV and IRV system.

For the sake of brevity we present here our results using a short version of the well known theories of HFS (Ref. 3) and TFB,[4] keeping the underlying Nodes-Links-Blobs (NLB) model.[24,31] The difference between those theories and the present work is only in the assumed distribution function $h(\varepsilon)$ of the geometrical proximity parameter ε. In the previous theories it was assumed that $h(\varepsilon)$ has a uniform distribution for $\varepsilon \rightarrow 0$, i.e., that there is some ε_0 such that for $\varepsilon < \varepsilon_0$ the distribution can be approximated by $h(\varepsilon) = h_0$, where h_0 is a constant. Here, we suggest that for this regime one can assume more generally that

$$h(\varepsilon) \propto \varepsilon^{-\omega}. \tag{1}$$

For $\omega = 0$, we recover the assumption and the predictions of the HFS and TFB theories,[2–4] while for $\omega > 0$, we have a ‘‘neck’’ distribution that prefers the smaller ε values as $\varepsilon \rightarrow 0$. We know of course that in order for the distribution to converge and be normalizable we must have that $\omega < 1$. Similarly, for $\omega < 0$ we have a ‘‘neck’’ distribution that prefers the larger ε values as $\varepsilon \rightarrow 0$. We further note that there is nothing special about the mathematical form of Eq. (1) and we use it since it retains the convenience of describing distributions that are ‘‘more divergent’’ or ‘‘less divergent’’ than that of the uniform distribution.

Since the resistance r of each resistor in the link depends on the geometry of the resistor,[3,4,30] i.e., $r \propto \varepsilon^{-u}$, we have that the average resistance of a resistor in a link containing L_1 singly connected (bonds) resistors is given by

$$\langle r \rangle_{L1} \propto \int \varepsilon^{-(u+\omega)} d\varepsilon, \tag{2}$$

where the integration is over the interval $\varepsilon_{\min} \leqslant \varepsilon \leqslant \varepsilon_0$ and $\varepsilon_{\min}$ is the typical smallest ε in the link. For the $\omega = 0$ case, $\varepsilon_{\min}$ is easily shown[3] to be proportional to $1/L_1$. Using the same argument but with $\omega \neq 0$, we find that $\varepsilon_{\min} \propto L_1^{-1/(1-\omega)}$. Now, if $u + \omega < 1$, the integral (2) does not diverge as $\varepsilon \rightarrow 0$ and one can take the average between $\varepsilon_{\min} = 0$ and some $\varepsilon = \varepsilon_0$. This yields that $\langle r \rangle_{L1}$ has a finite value that is independent of the proximity to the percolation threshold. Considering the fact that the critical behavior of the macroscopic resistance of the system, R, made of resistors of a constant $\langle r \rangle$ is universal and that R is given by[24] $R \propto (p - p_c)^t$, where $(p - p_c)$ is the ‘‘proximity’’ to the percolation threshold, we get that $t = t_{\text{un}}$. If, however, $u + \omega > 1$, the value $\langle r \rangle$ diverges at $\varepsilon = 0$, and we must take[3] the average in Eq. (2) while considering the finite, L_1 dependent, value of $\varepsilon_{\min}$. Using Eq. (2) and the above value of $\varepsilon_{\min}$ this yields that $\langle r \rangle_{L1} \propto L_1^{(u+\omega-1)/(1-\omega)}$. Noting[24,31] that $L_1 \propto (p - p_c)^{-1}$ we get then that $\langle r \rangle_{L1} \propto (p - p_c)^{-(u+\omega-1)/(1-\omega)}$. The critical behavior will be determined then by the nonuniversal exponent

$$t = t_{\text{un}} + (u + \omega - 1)/(1 - \omega). \tag{3}$$

Turning to the relative resistance noise, instead of repeating the procedure used in Ref. 4, we apply, for brevity, a short argument[30] that yields the same result. The argument is that in a random process, the squared variance (the averaged squared resistance fluctuation of a given resistor, $\langle \delta r^2 \rangle$ in our case) divided by the average squared ($\langle r \rangle^2$ in our case) is inversely proportional to the number of elements in the system. When the elements are individual volume parts of a continuous slab of material of volume V, one obtains then that $\langle \delta r^2 \rangle / \langle r \rangle^2 \propto 1/V$. Here, $\langle r \rangle$ is the average over a single resistor and thus it is given by $\langle r \rangle \propto \varepsilon^{-u}$. The corresponding volume element is related then to the ε parameter by[4,30] $V \propto \varepsilon^{v}$, yielding that $\langle \delta r^2 \rangle \propto \varepsilon^{-(2u+v)}$. Now, in order to find out the squared fluctuation in the link, $\langle \delta R^2 \rangle_{L1}$, one can apply Cohn's theorem[32] by which the power dissipated by the fluctuations of the ‘‘macroscopic’’ system (i.e., the fluctuations of the entire link $\langle \delta R^2 \rangle_{L1}$) equals the sum of the powers dissipated by the fluctuations in the individual resistors. In our case this means that $\langle \delta R^2 \rangle_{L1} \propto L_1 \langle \delta r^2 \rangle_{L1}$. The latter average is given, in our model, by

$$\langle \delta r^2 \rangle_{L1} \propto \int \varepsilon^{-(2u+v+\omega)} d\varepsilon, \tag{4}$$

where the integration is over the interval $\delta \leqslant \varepsilon \leqslant \varepsilon_0$, and δ, as in Ref. 4, is the typical ε value of the single resistor, R_ξ, which has the average resistance of an entire link. In the NLB model (where $R_\xi = L_1 \langle r \rangle_{L1}$), δ is given then by $R_\xi \propto \delta^{-u}$.

Let us consider first the case where R_ξ has a universal behavior (i.e., $\langle r \rangle_{L1}$ is independent of L_1). In this case one gets[4] that $\delta \propto L_1^{-1/u}$. Under these conditions one finds from Eq. (4) that

$$\langle \delta r^2 \rangle_{L1} \propto L_1^{(2u+v+\omega-1)/u}. \tag{5}$$

Since the resistance is universal, the relative resistance noise $S_R = \langle \delta R^2 \rangle_{L1} / \langle R \rangle_{L1}^2$ will deviate from the universal expression only by the term given in Eq. (5). Following the $L_1 \propto (p - p_c)^{-1}$ relation we get, provided $u + \omega < 1$ but $2u + v + \omega > 1$, that the relative resistance noise exponent will be given by

$$\kappa = \kappa_{\text{un}} + (2u + v + \omega - 1)/u, \tag{6}$$

where κ_{un} is the universal value of the relative resistance noise exponent. If, however, $2u + v + \omega < 1$, the integral in Eq. (4) converges and $\kappa = \kappa_{\text{un}}$.

As TFB,[4] we repeat the same argument for the second case (where the resistance has a nonuniversal behavior), i.e., in our model, when $R_\xi \propto L_1 L_1^{(u+\omega-1)/(1-\omega)}$. Thus, from $R_\xi \propto \delta^{-u}$ we find that the corresponding δ value will be given now by $\delta \propto L_1^{-1/(1-\omega)}$. In this case (since $u+\omega>1$ and $u,v>0$), we have that $2u+v+\omega>1$ and the integration of Eq. (4) yields that

$$\langle \delta r^2 \rangle_{L1} \propto L_1^{(2u+v+\omega-1)/(1-\omega)}. \tag{7}$$

Since in this case [see Eq. (3)] the resistance has the nonuniversal part of $\langle R \rangle_{L1} \propto L_1^{(u+\omega-1)/(1-\omega)}$, one readily obtains from Eq. (7) that $S_R \propto L_1^{(v-\omega+1)/(1-\omega)}$. This yields that the relative resistance noise exponent is

$$\kappa = \kappa_{\mathrm{un}} + (v+1-\omega)/(1-\omega). \tag{8}$$

We readily see that for $\omega \to 0$, Eqs. (6) and (8) give the TFB results.[4]

Now that we have the expressions (3), (6), and (8), we can check the specific numerical predictions for the ''real'' (two- or three-dimensional) RV and IRV models. As is well established,[2–4,30] for the RV model $u=d-3/2$ and $v=d-1/2$, while for the IRV model $u=d/2-1$ and $v=d/2$. Considering the fact that the requirement for obtaining a nonuniversal behavior of the resistance is that $u+\omega>1$ and that the possible values of ω lie in the interval $-\infty<\omega<1$, we see from Eq. (3) that one can have values from $t=t_{\mathrm{un}}$ to $t\to\infty$ for all the cases except for the $d=2$ IRV model. In the latter case, we cannot have that $u+\omega>1$ and thus $t=t_{\mathrm{un}}$, i.e., we will *never* have a nonuniversal t value. These results account well for the extreme $t>2.5$ values obtained in composites for which the $d=3$ RV model appears to be a good description,[7,13] as well as for the extreme $6>t>t_{\mathrm{un}}$ values in composites[19] for which the $d=3$ IRV model appears to be a good description.

Turning to the relative resistance noise, let us consider first the case presented by Eq. (6), i.e., for which the resistance has a universal behavior. As explained above, these are the cases where $u+\omega<1$. Hence, we consider all ω values that can account for this condition for the four models under consideration. For the $d=2$ RV model, a universal behavior can be found for $\omega<1/2$. In this case Eq. (6) yields that $\kappa=\kappa_{\mathrm{un}}+(3/2+\omega)/(1/2)$, so that $\kappa/t=\kappa/t_{\mathrm{un}}\leqslant 4.7$. For the $d=2$ IRV model, we have, for $\omega=0$, that $\langle \delta R^2\rangle_{L1}$ converges, so that $\kappa/t=\kappa_{\mathrm{un}}/t_{\mathrm{un}}$. However, for $1>\omega>0$, Eq. (6) yields that $\kappa=\kappa_{\mathrm{un}}+\omega/u$. Since $u\to 0$ in this case, the value of κ diverges. Hence, any value between $\kappa_{\mathrm{un}}/t_{\mathrm{un}}=0.86$ and $\kappa/t\to\infty$ is possible for this case. This unique situation accounts well for the extreme experimental result of $\kappa/t>5$ observed[9] in two-dimensional sand-blasted films. It further suggests then that an IRV-like resistors network was formed during the preparation of those films. Similar considerations for the $d=3$ systems yield that in the $d=3$ RV model, since $t_{\mathrm{un}}=2$ and $\kappa_{\mathrm{un}}=1.56$, we get that $0.78=\kappa_{\mathrm{un}}/t_{\mathrm{un}}\leqslant\kappa/t\leqslant 2.1$, while for the $d=3$ IRV model, we get that $0.78=\kappa_{\mathrm{un}}/t_{\mathrm{un}}\leqslant\kappa/t\leqslant 2.78$.

Except for the above $d=2$ IRV model we can have for all models and for $\omega>1/2$ a nonuniversal behavior of the resistance, and then we should consider Eqs. (3) and (8). This yields that

$$\kappa/t=[\kappa_{\mathrm{un}}+(v+1-\omega)/(1-\omega)]/[t_{\mathrm{un}}+(u+\omega-1)/(1-\omega)]. \tag{9}$$

For $\omega=0$, the only case that applies is that of the $d=3$ RV model yielding that $\kappa/t=2.02$. The largest κ and t exponents are obtained, however, for $\omega\to 1$. In this limit, $\kappa/t=(v/u)$. The latter ratios for the three applicable cases are 3 ($d=2$ RV), 5/3 ($d=3$, RV), and 3 ($d=3$, IRV). For all other possible ω values, the κ/t values for the nonuniversal t are smaller than 3. Indeed, considering all the available data for three-dimensional systems, there is no report of an experimental result with $\kappa/t>3$. Considering the lower limits of the κ/t ratio, we note that since $t\geqslant t_{\mathrm{un}}$ and $\kappa\geqslant\kappa_{\mathrm{un}}$ and since ω can obtain negative values, we will always have that the smallest κ/t ratio can be $\kappa_{\mathrm{un}}/t_{\mathrm{un}}$.

We can summarize now the predictions of the present theory for the relative resistance noise in comparison with that of the previous TFB theory[4] as follows: For the $d=2$ RV model, $0.86\leqslant\kappa/t\leqslant 4.7$ instead of $\kappa/t=3.16$; for the $d=2$ IRV model, $0.86\leqslant\kappa/t\leqslant\infty$ instead of $\kappa/t=0.86$; for the $d=3$ RV model, $0.78\leqslant\kappa/t\leqslant 2.1$ instead of 2.02; and for the $d=3$ IRV model, $0.78\leqslant\kappa/t\leqslant 3$ instead of 2.29. This is to be compared with the $0.9\leqslant\kappa/t\leqslant 8.1$ values observed experimentally in two-dimensional systems,[9,10,15,18] and $1\leqslant\kappa/t\leqslant 3$ values observed experimentally in three-dimensional systems.[11,12,19]

In the previous attempt[23] to account for measured κ/t values, as well as in the discussions given in the experimental reports,[9–12,15–19] the self-consistency between the experimental t and the κ/t values, in view of the prediction of the RV or IRV models, has not been considered. In what follows we will show how this self-consistency is improved considerably if we consider the present models in comparison with the original HFS and TFB models. We consider then the experimental works for which the RV or IRV models may apply, and in which t and κ/t have been determined simultaneously.[11,12,15,18,19] We have already shown above that all the measured κ/t values are within the intervals allowable by the present theory, while many of them deviate considerably from the original TFB predictions (i.e., the $\omega=0$ case). In our self-consistency test we consider first systems for which t was measured to be universal.[11,12,15,18] Indeed, the measured κ/t values are accounted for within the limits predicted here for the corresponding cases. In particular, we see that the largest κ/t value found experimentally,[12] i.e., $\kappa/t=3$, is within the limit of our theory and is significantly higher than the TFB prediction of $\kappa/t=2.3$.

A more stringent test of our theory is the case where both t and κ/t are nonuniversal. This is since this requires the finding of a *single* ω parameter, which will account for both exponents. Such a finding will not only yield quantitative proof for the present theory but will provide a tool for identifying the underlying network (RV or IRV) in real percolation systems. The only such experimental data we know of comes from the recent work of Wu and McLachlan.[19] They found, in a composite that is made of a mixture of conducting particles and insulating particles (where the former are somewhat larger than the latter), that $t\approx 2.66$ and that $\kappa/t\approx 1.55$. If we try to account for t by our $d=3$ IRV model, which is the more likely description of this system, we find from the value of t [using Eq. (3)] that $\omega\approx 0.7$. Using then Eq. (9), this yields that $\kappa/t\approx 1.3$. The difference between this and the above experimental value is less than 20% and can be accounted for even by theoretical considerations (e.g., corrections to scaling[23]). We note that this is not the case for the original HFS and TFB predictions that $t=2$ and that $\kappa/t \approx 2.3$ for this case.

In conclusion, the agreement and self-consistency of the present predictions with all the available experimental results indicates that the present assumption of nonrandom particle (or void) distributions provides an improved description of real percolation systems. Further simultaneous measurements of t and κ/t are called for in order to see whether comparison with the present theory can yield information on the distribution function of particles (or voids) in corresponding percolation systems.

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