Segregated tunneling-percolation model for transport nonuniversality

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We propose a theory of the origin of transport nonuniversality in disordered insulating-conducting compounds based on the interplay between microstructure and tunneling processes between metallic grains dispersed in the insulating host. We show that if the metallic phase is arranged in quasi-one-dimensional chains of conducting grains, then the distribution function of the chain conductivities g has a power-law divergence for $g \to 0$, leading to nonuniversal values of the transport critical exponent t. We evaluate the critical exponent t by Monte Carlo calculations on a cubic lattice, and show that our model can describe universal as well nonuniversal behaviors of transport depending on the value of few microstructural parameters. Such a segregated tunneling-percolation model can describe the microstructure of a quite vast class of materials known as thick-film resistors, which display universal or nonuniversal values of t depending on the composition.

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I. INTRODUCTION

When the conductivity σ of an insulating-conducting compound is measured as a function of the volume concentration p of the conducting phase, one finds that by reducing p the system eventually undergoes a conductor-to-insulator transition at a particular critical value p_c of the volume concentration. In the critical region $0 < p - p_c \ll 1$ the conductivity follows a power law behavior of the form

$$\sigma = \sigma_0 (p - p_c)^t, \tag{1}$$

where σ_0 is a prefactor which depends on the particular system considered, and t is a positive number typically larger than the unity.

Percolation theory explains the power-law form of Eq. (1) as being due to the lack of any cutoff length scale apart the linear size of the sample, and predicts that the exponent t is universal and depends only upon the dimensionality of the system.[1] This prediction is confirmed by various granular metals compounds and model systems which have been found to follow Eq. (1) with $t \simeq 2.0$,[2–5] that is the value obtained by numerical calculations on three-dimensional random resistor network (RRN) models.[6] In addition to systems showing universality, a large number of disordered compounds displaying values of t larger than $t \simeq 2.0$ have been repeatedly reported,[7–13] so that in the present situation it appears that t can assume any value between $t \simeq 2.0$ up to about $t \sim 6.0$–7.0.

Within percolation theory on a RRN, Kogut and Straley showed that a universality breakdown of the transport may arise from anomalous distributions of elemental conductivities.[14] By assigning to each neighboring couple of sites on a regular lattice a bond with finite conductivity g with probability p and zero conductivity with probability $1 - p$, the resulting bond conductivity distribution function becomes

$$\rho(g) = p h(g) + (1 - p)\delta(g), \tag{2}$$

where $\delta(g)$ is the Dirac delta function and $h(g)$ is the distribution function of the finite bond conductivities. For well behaved $h(g)$, transport is universal and follows Eq. (1) with $t = t_0 \simeq 2.0$ for three dimensional lattices. Instead, if $h(g)$ has a power law divergence for small g of the form

$$\lim_{g \to 0} h(g) \propto g^{-\alpha}, \tag{3}$$

and α is larger than a critical value α_c, Kogut and Straley showed that transport is no longer universal and the conductivity exponent becomes dependent on α.[14] A renormalization group analysis predicts, in fact, that

$$t = \begin{cases} t_0 & \text{if } (D-2)\nu + \dfrac{1}{1-\alpha} < t_0 \\ (D-2)\nu + \dfrac{1}{1-\alpha} & \text{if } (D-2)\nu + \dfrac{1}{1-\alpha} > t_0 \end{cases}, \tag{4}$$

where D is the dimensionality of the lattice and ν is the correlation-length exponent ($\nu = 4/3$ for $D = 2$ and $\nu \simeq 0.88$ for $D = 3$).[15–17] For $D = 3$ and by using $t_0 \simeq 2.0$ and $\nu \simeq 0.88$ the critical value of the exponent is $\alpha_c \simeq 0.107$.

Microscopic models which may justify Eq. (3) are the random void (RV) model proposed by Halperin, Seng, and Fen,[18] and the tunneling-percolating model of Balberg.[11] The RV model describes a system of insulating spheres (or disks in two dimensions) embedded randomly in a continuous conducting material. In this situation, transport is dominated by the conductivity of the narrow necks bounded by three interpenetrating insulating spheres. Such necks have a wide distribution in widths resulting in a wide distribution of conductivities. The original formulation of the RV model predicted $t = t_0 + 0.5$ for the conductivity exponent of the whole sample. A recent generalization of the RV model by Balberg has shown that t can assume even higher values, and that, in principle, it is not bounded above.[19]

In the tunneling-percolating model of Ref. 11, transport is assumed to be dominated by quantum tunneling between neighboring conducting particles dispersed in an insulating

medium. If the distribution function $P(r)$ of the distance r between two neighboring particles decays with r much slower than the tunneling decay $\exp(-2r/\xi)$, where ξ is the localization length, then the tunneling conductivity distribution function $h(g)$ can be shown to behave as Eq. (3) with $\alpha \simeq 1-\xi/2a$, so that the transport exponent t becomes dependent of the mean tunneling distance a.[11] Interactions between the conducting and insulating phases as well as properties of the microstructure are argued to concur as to the r dependence of $P(r)$. Due to the complexity of the problem, explicit calculations of the interparticle distance distribution function are missing, and one must relay on phenomenological forms of $P(r)$.

In this paper we provide a microscopic derivation of $P(r)$ which has been inspired by the peculiar microstructure observed in a particular class of insulating-conducting compounds: the so-called thick-film resistors (TFRs). These compounds are based on RuO_2 (or $Bi_2Ru_2O_7$, $Pb_2Ru_2O_6$, and IrO_2) grains mixed and fired with glass powders.[20] Typically, TFRs are often in a segregated structure regime in which large regions of glass constraint the much smaller conducting grains to be segregated in between the interstices of neighboring glass grains. Micrographs reveal that the conducting grains are arranged in a network of filaments spanning the entire sample.[7,21,22] By taking into account the quasi-one-dimensional structure of such filaments and by neglecting interactions with the insulating phase, we show that the resulting $P(r)$ can decay much slower than the tunneling decay, leading to a nonuniversal behavior of transport.

This paper is organized as follows. In Sec. II we construct a RRN model which captures the essential structure of the filamentary network of TFRs and calculate the resulting $P(r)$ and the distribution function of the conductivity of filaments. In Sec. III we perform Monte Carlo calculations and calculate the conductivity exponent t for a variety of situations. Section IV is devoted to discussions and conclusions.

II. MODEL

Before describing our model in details, we find it useful to first discuss in general the interplay between the spatial distribution of the conducting phase within the insulating matrix and transport properties. Let us consider a generic insulating-conducting compound where the conducting grains are embedded in an insulating host. In this situation, electron transfer is governed by electron tunneling from grain to grain. The grain charging energy and the Coulomb interaction between charged grains affects the overall transport properties especially regarding their behavior in temperature. Here we focus on systems where the temperature is high enough to possibly neglect charging and Coulomb effects, so that the main electron transfer is dominated solely by tunneling leading to intergrain conductivity of the form

$$\sigma(r) = \sigma_0 e^{-2(r-\Phi)/\xi}, \tag{5}$$

where σ_0 is a constant which can be set equal to the unity without loss of generality, $\xi \propto 1/\sqrt{V}$ is the tunneling factor (or localization length) and V is the intergrain barrier potential. In Eq. (5) we have approximated the conducting grains by spheres of diameter Φ and r is the distance between the centers of two spheres which we treat as impenetrable ($r \geqslant \Phi$).

Due to the exponential decay of Eq. (5), contributions to $\sigma(r)$ from far away spheres can be neglected,[11,23] so that from now on r denotes the distance of two nearest-neighboring spheres. Hence, the ensemble dependence of $\sigma(r)$ upon r is completely defined by the distribution function $P(r)$ of the distance between nearest-neighboring spheres. In fact, once $P(r)$ is known, the conductivity distribution function $h(g)$ can be obtained as follows:

$$h(g) = \int dr P(r) \delta[g - \sigma(r)]. \tag{6}$$

In this preliminary discussion, we are interested in studying how the form of $P(r)$ affects $h(g)$ via Eq. (6) and which are the requisites of $P(r)$ which eventually could generate a power-law distribution function as that of Eq. (3). As already pointed out, $P(r)$ depends on the microstructure of the composite and on eventual interactions between the insulating and conducting phases. In principle, therefore, the form of $P(r)$ depends on the particular composite considered. However, if we imagine that interactions can be neglected, then it is natural to assume that the conducting spheres are Poisson distributed within the insulating phase. Then if D is the dimensionality of the system, by following Refs. 24 and 25 the nearest-neighbor distance distribution function is approximately of the form

$$P(r) \sim \frac{e^{-(r/a_D)^D}}{a_D}, \tag{7}$$

where a_D is a constant depending on the mean distance between neighboring spheres. Equation (7) is an asymptotic approximation of the true $P(r)$, and is valid only in the $r/a_D \gg 1$ limit. This is, however, the limiting region of interest to us since it governs, via Eq. (5), the $g \ll 1$ regime. It is also worth pointing out that Eq. (7) holds true for penetrable as well as impenetrable (hard-core) spheres, the only difference being in the explicit expression for a_D which is however of not importance at the moment.[25]

By inserting Eqs. (5) and (7) into Eq. (6), the resulting conductivity distribution function becomes

$$\begin{aligned} h(g) &\sim \int \frac{dr}{a_D} e^{-(r/a_D)^D} \delta[g - \sigma(r)] \\ &= \frac{\xi}{2a_D} \frac{1}{g} \exp\left(-\frac{\xi}{2a_D} \ln g^{-1} \right)^D, \end{aligned} \tag{8}$$

which, after some manipulations, reduces to

$$h(g) \sim \frac{\xi}{2a_D} g^{(\xi/2a_D)^D (\ln g^{-1})^{D-1} - 1}. \tag{9}$$

For $D=2$ and 3, the $g \to 0$ limit of the above expression goes to zero irrespective of the value of $\xi/2a_D$. In this case, therefore, no power-law divergence of $h(g)$ is encountered and,

as discussed in Sec. I, transport is governed by the universal critical exponent $t=t_0\simeq 2.0$. Instead, when $D=1$, Eq. (9) becomes

$$h(g)\sim \frac{\xi}{2a_1} g^{(\xi/2a_1)-1}, \tag{10}$$

which is exactly of the form of Eq. (3) if we identify α with $1-\xi/2a_1$. We have arrived, therefore, at the result that if the spheres are Poisson distributed along a one-dimensional line, the resulting conductivity distribution function has a power-law behavior for small g and, consequently, transport is non-universal for sufficiently large values of $1-\xi/2a_1$.

The difference between the $D=2$ and 3 and $D=1$ cases stems from the decay of Eq. (7) which for $D=2$ and 3 is much too fast with respect to the simple exponential decay of Eq. (5). In fact from Eq. (6) it is simple to show that as long as $\lim_{r\to\infty} P(r)/\sigma(r)=0$ then $\lim_{g\to 0} h(g)=0$ irrespective of the detailed structure of $P(r)$. Hence to construct a RRN model having $h(g)$ of the form of Eq. (3), we must consider forms of $P(r)$ whose decay for $r\to\infty$ is sufficiently slow. The result of Eq. (10) suggests that for this scope one dimensionality is an important ingredient, at least as long as interactions between conducting and insulating phases can be neglected.

Among the various insulating-conducting compounds, thick film resistors are systems whose microstructure can be appropriately described in terms of quasi-one-dimensional units. Let us consider the highly non-homogeneous microstructure typical of TFRs. These systems are constituted by a mixture of large glassy particles (with size L of order $1-3\mu$m) and small conducting grains of size Φ typically varying between ~ 10 and ~ 200 nm. Due to the high values of L/Φ, the small metallic grains tend to occupy the narrow regions between the much larger insulating zones, leading to a filamentary distribution of the conducting phase.[7,21,22] A classical model to describe such a segregation effect was proposed already in the 1970s by Pike.[7] This model replaces the glassy particles by insulating cubes of size $L\gg\Phi$ whose edges can be occupied by chains formed by adjacent metallic spheres of diameter Φ. Let us assume that an edge has probability p of being occupied by a chain of $n+1$ spheres and probability $1-p$ of being empty. As depicted in Fig. 1, the set of occupied and empty edges form a cubic lattice spanning the entire sample.

To define the RRN relevant for this model we proceed as follows. The conductivity g of a single occupied channel is governed by the conductivities of the metallic spheres and those between pairs of two-neighboring spheres. The conductivity of the metallic spheres adds only a negligible contribution to g which is then given by n conductivities σ_i of pairs of nearest-neighboring spheres in series:

$$g^{-1}=\sum_{i=1}^{n}\frac{1}{\sigma_i}. \tag{11}$$

We assume that the inter-sphere conductivities σ_i are due to the tunneling processes between two adjacent spheres, so that their sphere-to-sphere distance r dependence is that of

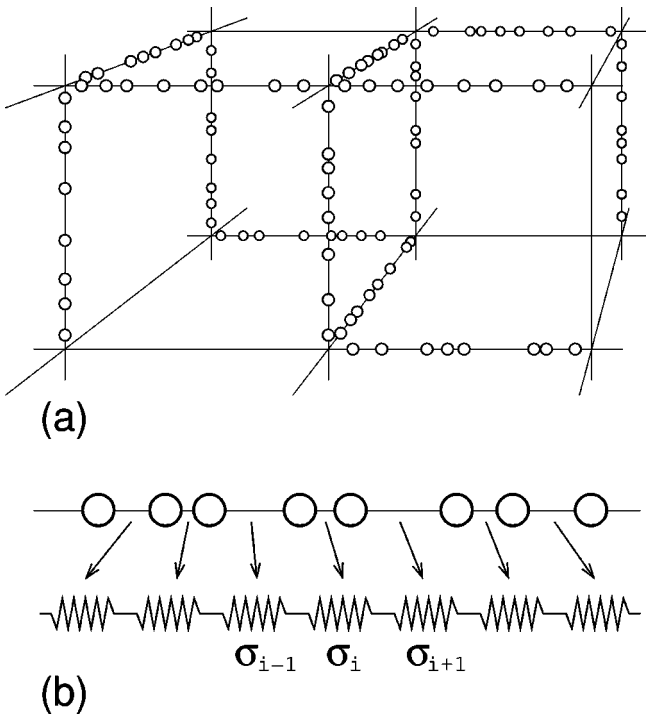

FIG. 1. (a) Pictorial representation of the segregated tunneling-percolation model. The cubes represent insulating grains while the spheres are conducting particles. The spheres are arranged to occupy the edges of the insulating cubes with probability p. The total ensemble of occupied and unoccupied edges forms a cubic lattice spanning the entire sample. (b) Equivalence between an edge occupied by $n+1$ spheres and a conducting element. The set of inter-spheres tunneling conductivities is equivalent to a conductor with n resistive elements with conductivities σ_i in series. The fluctuation in distance between two neighboring spheres leads to fluctuating tunneling conductivities. With this equivalence, the model depicted in (a) can be considered as a bond-percolation model where a fraction p of bonds has variable conductivities and a fraction $1-p$ is insulating.

Eq. (5). For TFRs, the tunneling hypothesis is well sustained by their high values of piezoresistance (i.e., the strain sensitivity of transport),[20] and the low temperature dependence of transport indicating some kind of assisted hopping. As done in the introductory part of this section, we neglect interactions between the insulating and conducting phases, and assume that the sphere centers are Poisson distributed along the cube edge. In doing so, we implicitly assume that finite size effects of the channels can be neglected and that periodic boundary conditions are applied. In this way the last sphere on one end of the channel is identified with the first one on the opposite end, so that we have n individual spheres and n intersphere tunneling junctions. In this situation, the distances r change according to the distribution function $P_n(r)$ of the nearest-neighbor distances r of n impenetrable spheres arranged randomly in a quasi-one-dimensional channel. By following Ref. 25, $P_n(r)$ can be calculated exactly, and it is given by

$$P_n(r)=\frac{1}{a_n-\Phi}e^{-(r-\Phi)/(a_n-\Phi)}\Theta(r-\Phi), \tag{12}$$

where Θ is the step function and

$$a_n = \frac{\Phi}{2}\left(1+\frac{L}{n\Phi}\right), \tag{13}$$

is the mean intersphere (center-to-center) distance. In the above expression $n\Phi/L$ cannot be larger than the unity since no more than L/Φ spheres can be accommodated inside a channel. Note that the asymptotic expression Eq. (7) for $D=1$ coincides with Eq. (12) if a_1 is identified with $a_n-\Phi$. Hence the distribution function $f(\sigma)$ of the intersphere conductivities should be of the same form of Eq. (10). In fact,

$$f(\sigma)=\int_0^\infty dr P_n(r)\,\delta[\sigma-\sigma(r)]=(1-\alpha_n)\sigma^{-\alpha_n}, \tag{14}$$

where

$$\alpha_n = 1-\frac{\xi/2}{a_n-\Phi}. \tag{15}$$

Having obtained an explicit expression for the distribution function $f(\sigma)$ of the intersphere conductivities, we can now calculate the total distribution function $h_n(g)$ of the whole channel. From Eq. (11), $h_n(g)$ can be defined as

$$h_n(g)=\int d\sigma_1 \ldots d\sigma_n f(\sigma_1)\ldots f(\sigma_n)\,\delta\left[g-\left(\sum_{i=1}^n \frac{1}{\sigma_i}\right)^{-1}\right], \tag{16}$$

which, by using Eq. (14), reduces to

$$\begin{aligned} h_n(g) &= (1-\alpha_n)^n \int d\sigma_1 \ldots d\sigma_n \left(\prod_{i=1}^n \sigma_i\right)^{-\alpha_n} \\ &\quad \times \delta\left[g-\left(\sum_{i=1}^n \frac{1}{\sigma_i}\right)^{-1}\right] \\ &= (1-\alpha_n)^n g^{-\alpha_n} \int d\sigma_1 \ldots d\sigma_n \left(\sum_{i=1}^n \prod_{j\neq i}^n \sigma_j\right)^{-\alpha_n} \\ &\quad \times \delta\left[g-\left(\sum_{i=1}^n \frac{1}{\sigma_i}\right)^{-1}\right]. \end{aligned} \tag{17}$$

It is clear that $h_n(g)$ behaves as $g^{-\alpha_n}$ for $g \ll 1$ since the integral appearing in the last equality of the above expression is well behaved in the $g\to 0$ limit. In fact the $g\to 0$ limit of the Dirac δ function appearing in Eq. (17) reduces to

$$\lim_{g\to 0}\delta\left[g-\left(\sum_{i=1}^n \frac{1}{\sigma_i}\right)^{-1}\right]=\sum_{l=1}^n \delta(\sigma_l)\left(\frac{\sum_{i=1}^n \prod_{j\neq i}\sigma_j}{\prod_{i\neq l}\sigma_i}\right)^2, \tag{18}$$

so that, finally,

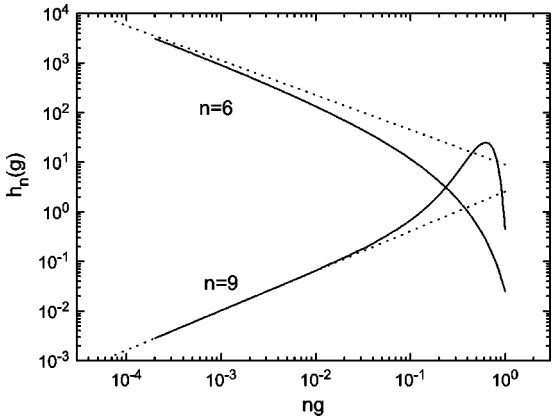

FIG. 2. Distribution function $h_n(g)$ of the conductivity of the occupied channels for $L/\Phi=10$, $\xi/\Phi=0.2$, and different values of n. Solid lines are the result of a numerical calculation of Eq. (16), while the dotted lines are the asymptotic results of Eq. (19).

$$\begin{aligned} h_n(g) &\simeq (1-\alpha_n)^n g^{-\alpha_n}\int d\sigma_1 \ldots d\sigma_n \sum_{l=1}^n \delta(\sigma_l)\left(\prod_{i\neq l}\sigma_i\right)^{-\alpha_n} \\ &= n(1-\alpha_n)g^{-\alpha_n} \quad \text{for} \quad g\ll 1. \end{aligned} \tag{19}$$

The above equation is the main result of this paper, i.e., the distribution function of the occupied channel conductivities $h_n(g)$ is of the same form of Eq. (3). In this situation, for sufficiently large values of α_n the RRN conductivity can behave in a nonuniversal way with exponent $t>2.0$. The condition for universality breakdown is given by Eq. (4) which for a three-dimensional network implies $\alpha_n>\alpha_c \simeq 0.107$. From Eqs. (13) and (15) this condition corresponds to

$$n<n_c=\frac{1-\alpha_c}{1-\alpha_c+\xi/\Phi}L/\Phi, \tag{20}$$

so that, for fixed values of ξ/Φ and L/Φ, the value of the transport exponent t is governed solely by the number of spheres that can be arranged within the occupied one-dimensional channels. The overall behavior of $h_n(g)$ is reported in Fig. 2 where we report a numerical calculation of Eq. (16) (solid lines) together with the asymptotic behavior obtained in Eq. (19) (dotted lines). In this example we have set $\xi=2$ nm, $\Phi=10$ nm, and $L=0.1$ μm, corresponding to $L/\Phi=10$, $\xi/\Phi=0.2$, and $n_c\simeq 8.17$. For $n=9>n_c$ the distribution function goes to zero as Eq. (19) with $\alpha_n\simeq -0.244$ while for $n=6<n_c h_n(g)$ diverges for $g\to 0$ with exponent $\alpha_n=0.7$. Since $\alpha_c\simeq 0.107$, we expect that for $n=9$ transport is universal while for $n=6$ the exponent t becomes larger than $t_0\simeq 2.0$ as in Eq. (4).

Before discussing our numerical results on the RRN conductivity, it is worth pointing out that our model can be easily generalized to consider, also, situations in which the number of spheres accommodated in the one-dimensional channels is not fixed. More specifically, if $\mathcal{P}_{n'}$ is the distribution function of the number n' of spheres, then the distribution function of the occupied channels is generalized to

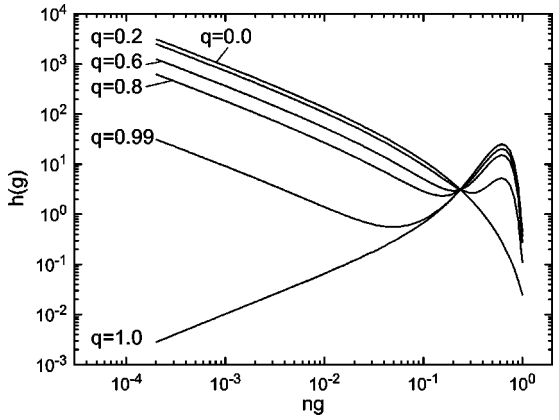

FIG. 3. Channel conductivity distribution function $h(g)$ [Eq. (21)] for the bimodal distribution of Eq. (22) and for different values of q. $L/\Phi=10$, $\xi/\Phi=0.2$, $n1=9$, and $n2=6$.

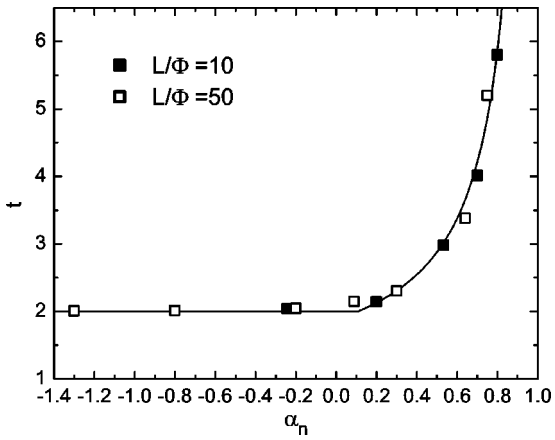

FIG. 4. Critical exponent t as a function of the tunneling exponent α_n for $\xi/\Phi=0.2$ and different values of L/Φ and of the number n of inter-sphere tunneling junctions accommodated within the occupied channels of a cubic random-resistor network. From left to right: $n=9,8,\ldots,5$ for $L/\Phi=10$ (filled squares) and $n=46,45,43,41,39,33$, and 27 for $L/\Phi=50$ (open squares). The solid curve is the theoretical result $t=t_0\simeq 2.0$ for $\alpha_n<\alpha_c\simeq 0.107$ and $t=\nu+1/(1-\alpha_n)$ for $\alpha_n>\alpha_c$ [see Eq. (4)].

$$h(g)=\sum_{n'} \mathcal{P}_{n'} h_{n'}(g). \tag{21}$$

As an instructive case let us consider a bimodal distribution of the form

$$\mathcal{P}_{n'}=q\,\delta_{n',n1}+(1-q)\,\delta_{n',n2}, \tag{22}$$

where $0\leqslant q\leqslant 1$. For $q=0$ or $q=1$ we recover the previous case in which the occupied channels have the same number of spheres and whether transport is universal or not depends on the specific values $n1$ (for $q=1$) or $n2$ (for $q=1$). An interesting case is given by $0<q<1$ and $n2<n_c<n1$, according to which there is a concentration q of channels conductivities with distribution function with exponent $\alpha_{n1}<\alpha_c$ and $1-q$ channels with $\alpha_{n2}>\alpha_c$. This case is depicted in Fig. 3, where $n1=9$ and $n2=6$ and, as in Fig. 2, $L/\Phi=10$ and $\xi/\Phi=0.2$. For all values $q<1$ the $g\to 0$ limit is governed by the diverging part of the total distribution function. In this case we expect that at the critical point the transport exponent in not universal for any value $q<1$. Note, however, that for q sufficiently close to unity, the asymptotic regime is reached for relatively small values of the conductivity. As we shall see in Sec. III, this has the effect of shrinking the region where criticality sets in with $t=\nu+1/(1-\alpha_{n2})$.

III. MONTE CARLO RESULTS ON THE CUBIC LATTICE

In this section we discuss our Monte Carlo calculations for the conductivity σ of the RRN model defined in Sec. II. In constructing the RRN we must first implement numerically the conductivity of the channels occupied by a given number n of spheres. If $x_i(i=1,\ldots,n)$ is a set of random numbers equally distributed in the interval (0,1) then it is easily found that the channel conductivity g having Eq. (17) as its corresponding distribution function is

$$g=\left[\sum_{i=1}^{n} x_i^{1/(\alpha_n-1)}\right]^{-1}. \tag{23}$$

The RRN is then defined to have a fraction p of channels (in a cubic lattice) with g as given by Eq. (23) and a fraction $1-p$ with $g=0$. The generalization to a bimodal distribution [Eqs. (21) and (22)] is straightforward.

To calculate the transport exponent t numerically, we use the transfer-matrix method of Derrida and Vannimenus applied to a simple cubic lattice of $N-1$ sites in the z direction, N sites along y and $\mathcal{L}$ along the x direction.[26] Periodic boundary conditions are used in the y direction while a unitary voltage is applied to the top plane and the bottom plane is grounded to zero.[27,28] For sufficiently large $\mathcal{L}$ ($\mathcal{L}\gg N$) this method permits one to calculate the conductivity per unit length of a cubic lattice. We calculate the conductivity σ_N for different linear sizes N at the percolation thresold $p_c\simeq 0.2488126$ for bond percolation on a cubic lattice,[29] and then by least-square fits we extract the critical exponent t from the finite size scaling relation[28,31]

$$\sigma_N=aN^{-t/\nu}(1+bN^{-\omega}), \tag{24}$$

where $\nu\simeq 0.88$ is the correlation length exponent, a and b are constants, and ω is the first correction to the scaling exponent t/ν. In performing the calculations we have considered the following geometries: $N=6$ ($\mathcal{L}=5\times 10^7$), $N=8$ ($\mathcal{L}=2\times 10^7$), $N=10$ ($\mathcal{L}=1\times 10^7$), $N=12$ ($\mathcal{L}=8\times 10^6$), $N=14$ ($\mathcal{L}=2\times 10^6$), and $N=16$ ($\mathcal{L}=2\times 10^6$).

In Fig. 4 we report the obtained values of the critical exponent t for $\xi/\Phi=0.2$ and for two different values of the ratio L/Φ between the length channel and sphere diameter. Each square corresponds to a particular number n of inter-sphere tunneling junctions arranged in the channel (see the caption) which, from Eqs. (13) and (15), also gives the corresponding value of the tunneling exponent α_n reported in the abscissa. As a function of α_n, the critical exponent t nicely follows Eq. (4) (solid curve) confirming that a univer-

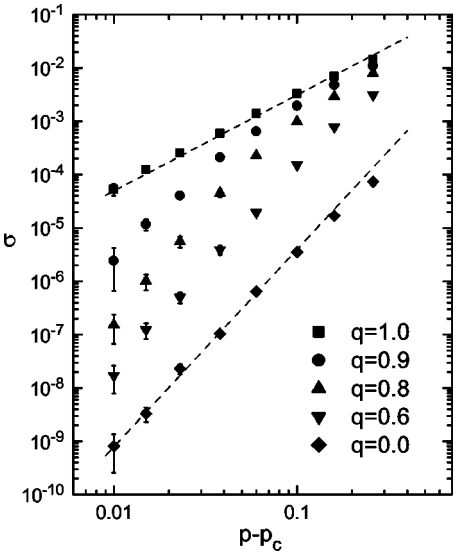

FIG. 5. Conductivity σ of a $40\times40\times40$ cubic lattice with the bimodal distribution of Eq. (22) with $n1=9$, $n2=6$, $L/\Phi=10$, and $\xi/\Phi=0.2$. Symbols are mean values of ten different runs with standard deviations given by the error bars. Dashed lines are fits to Eq. (1) with $t=1.8\pm0.1$ for $q=1$ and $t=3.7\pm0.2$ for $q=0$.

sal ($t\simeq2.0$) or nonuniversal ($t>2.0$) behavior is obtained just by changing the number of spheres accommodated in the channels. In our least square fittings to Eq. (24) we have found that the minimum χ^2 is obtained by setting $b\neq0$ and $\omega\sim1.0$ for $\alpha_n<\alpha_c$ and $b=0$ for $\alpha_n>\alpha_c$. It is worth noticing that our Monte Carlo results on the cubic lattice agrees with Eq. (4) much better than the corresponding problem [Eqs. (2) and (3)] on the two-dimensional square lattice.[32,33]

We have also applied the transfer-matrix method to the bimodal distribution of Eq. (22) with $L/\Phi=10$, $\xi/\Phi=0.2$, $n1=9$, and $n2=6$, and found that, as expected, at p_c the critical exponent is already nonuniversal for $q=0.9$. However, what is interesting in the bimodal case is the behavior of the conductivity σ away from the critical thresold. The high-structured shape of $h(g)$ for $0<q<1$ reported in Fig. 3 in fact suggests that the p dependence of σ could be affected by the competition between the two exponents $\alpha_{n1}<\alpha_c$ and $\alpha_{n2}>\alpha_c$. To study this problem, the application of the transfer-matrix method for values of the occupied channel concentration p away from the critical thresold p_c is not efficient since the computational time of the algorithm increases as p is moved from p_c.[26,27] Hence we have approached the problem by solving the RRN by the conjugate gradient method, which is more efficient away from the critical point.[30] The resulting σ is reported in Fig. 5 for a cubic lattice of $40\times40\times40$ sites and periodic boundary conditions applied to the sides not connected with the external potential drop. We have considered the bimodal case defined by $L/\Phi=10$, $n1=9$, and $n2=6$, and different values of q. For $q=1$ (filled squares in Fig. 5) all the occupied channels have $n1=9$ tunneling junctions and the conductivity is well approximated by Eq. (1) with the critical exponent $t=1.8\pm0.1$. This value is slightly less than the universal result $t=t_0\simeq2.0$, and this difference signals the limitation of extracting critical exponents from the p dependence of σ in finite size samples. However σ follows the power-law Eq. (1) in the interval $p-p_c<0.1-0.2$. A nice power law is also found for $q=0$ (filled diamonds) for which the occupied channels have $n2=6$ number of junctions. In this case, however, the exponent is $t=3.7\pm0.2$, i.e., slightly less than the nonuniversal value $t\simeq4.0$ obtained by the transfer-matrix method (see Fig. 4).

In Fig. 5 we report also σ calculated for intermediate values of q. For $q=0.9$ and 0.8 the $p-p_c$ dependence of the conductivity can be reasonably fitted by a simple power law only for $p-p_c<\sim0.05$ for which we have found $t=3.2\pm0.3$ and 4.1 ± 0.4, respectively. We interpret this shrinking of the critical region as being due to the large contribution of the fraction of channels with $n1=9$ to the occupied channels distribution function $h(g)$. However, for $q=0.6$ the critical region is already fully restored, and σ follows a power law with $t=3.8\pm0.5$ for $p-p_c\leqslant0.1$.

IV. DISCUSSION AND CONCLUSIONS

As shown in the previous sections, the interesting characteristic of our segregated tunneling-percolation model is the possibility of having a universal or nonuniversal behavior of transport within the same theoretical framework. As we have discussed, if the microscopic physical and geometric parameters (insulating cube size L, sphere diameter Φ, number of spheres, and localization length ξ) are such that the tunneling factor α_n is larger than the critical value $\alpha_c\simeq0.107$ then the critical exponent t is nonuniversal and follows $t=\nu+1/(1-\alpha_n)$; otherwise transport is universal and the critical exponent is $t=t_0\simeq2.0$. This universal/nonuniversal crossover is experimentally observed in thick-film resistors, which are reported to vary between $t\simeq2.0$ and $t\simeq5.0$ for mixtures of chemically identical constituents. It can be argued that different fabrication procedures (for example, the firing temperature) affect the microstructure leading to different effective values of α_n. Of course our model is oversimplified in the sense that interactions between the conducting and insulating phases are completely neglected. However, it is remarkable that only two assumptions, the quasi-one-dimensionality of the conducting channels and the Poisson distribution of the position of the spheres inside the channels, are sufficient to give rise to such a rich phenomenology.

The model discussed in this paper captures the essential physics, but eventually it can be further generalized to include more realistic features. For example, it is possible to account for different sizes of the conducting spheres in a straightforward manner, since for this case the one-dimensional nearest-neighbor distance distribution function is also provided by an analytical and exact expression.[34] Also the tunneling expression [Eq. (5)] can be refined by including, for example, charging energies or distribution functions for the tunneling factor ξ.

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