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The percolation staircase model and its manifestation in composite materials

I. Balberg[a], D. Azulay, Y. Goldstein, J. Jedrzejewski, G. Ravid, and E. Savir

The Racah Institute of Physics, The Hebrew University, 91904 Jerusalem, Israel

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Abstract. We studied the tunneling percolation conductivity dependence on the site or bond occupation probability in the square lattice. The model predicts that in both, lattice and continuum systems in which there is a hierarchy of the local conductances, the dependence of the global conductivity on the site or volume occupation probability will yield a conductivity staircase. In particular we evaluate the implications of the staircase on the critical behavior of the conductivity. We then show experimental evidence for the predicted percolation-tunneling staircase in a Ag-Al_2O_3 granular metal system and in a carbon black-polymer composite. Following that, we propose that for carbon nanotube (CNT) polymer composites the data in the literature give ample support to a percolation-dispersion staircase behavior. The implication of the present findings on the percolation-hopping problem in composite materials is also discussed.

1 Introduction

In the classical lattice percolation problem the conductivity dependence on the site occupation probability p is predicted to exhibit the phase transition power law dependence of [1,2]:

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

where p_c is the threshold for the onset of global connectivity, t is the critical exponent that has, in three dimensional (3D) lattices, the universal value of $t_{\text{un}} \approx 1.7$–$2$ and G is a parameter that is proportional to the local bond conductance g [1–3]. If, however, the conductance distribution function $f(g)$ is such that the lowest local conductance that is necessary for the onset of percolation, g_c, behaves as $g_c \propto (p - p_c)^u$ (where $u > 0$), a critical behavior with a $t = t_{\text{un}} + u$ will result [3–5]. We wrote equation (1) here for the site percolation problem because of its relevance to systems that consist of conducting particles such as a system of rigid spheres that can occupy sites on a lattice [2,6]. Recalling that in the classical site percolation problem two sites are considered connected if they are nearest neighbors and both are occupied. In this extension, two spheres that occupy nearest neighbor sites are considered geometrically and thus electrically connected by touching. In that system the fractional occupied volume of the conducting spheres is $x = \gamma p$ where γ is the filling factor of the lattice. The important finding of Scher and Zallen (S&Z) in 1970 [6] was however that $x_c (= \gamma p_c)$ is essentially a dimensional constant (~0.16 in 3D) that applies also to systems of a random mixture of touching conducting and insulating spheres. The fractional volume of the conducting spheres x is proportional then to the concentration of the conducting particles in the system, N. Hence, it follows immediately from equation (1) that in the S&Z model the conductivity of the system obeys the relation:

$$\sigma(x) \propto (x - x_c)^t. \tag{2}$$

While the latter result is not unexpected topologically (the critical average number of bonds per occupied site, B_c, at x_c, should be the same for the above lattice and random systems) the extension of equation (2) to other systems in the continuum is not straightforward. In particular, this is not trivial when the particles involved are not spheres and when the particle touching-connectivity criterion of S&Z does not apply. Apart for some specially designed experiments on S&Z like systems, one or both of these two criteria is usually not fulfilled in systems of conducting particles. In particular, in composite materials where the particles do not actually touch and the interparticle conduction is by tunneling, the essence of percolation, i.e., the limited connectivity of the system, seems to lose its meaning [7,8]. This is since no particles are geometrically connected but all particles are electrically connected for any value of x. It is quite surprising then that even for such systems the relation given by equation (2) is generally observed experimentally [3,9].

In order to derive an explanation for the latter, a priori, unexpected behavior, we consider another experimentally observed behavior that has not been analyzed previously

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[a] e-mail: balberg@mail.huji.ac.il

while it was exhibited by very many composites. For this purpose let us define as a "single-smooth" behavior the case where equation (2) applies with the same single t and x_c values, for any subset of a given set of data. The other possible behavior is that equation (2) is fulfilled but with a different x_c for different intervals of x such that between these intervals there are "jumps" in the $\sigma(x)$ dependence. As systems of strong deviation from the "single-smooth" (Eq. (2)-like) behavior we can mention granular metals [10], metal particles embedded in polymers [11,12], and various carbon allotropes embedded in a polymer. The latter include carbon black [13], carbon nano fibers [14], carbon nanotubes [15,16], graphene [17] and graphite [18] composites. In the analyses of the corresponding many data, the above deviations from the "single-smooth" behavior have been either overlooked or ignored completely. In particular, the entire experimental data was fitted to an equation (2)-like dependence with a single x_c and with a non universal t exponent, a fit that actually describes the "single smooth" approximation of $\sigma(x)$ to the measured dependence. This approach appears then to conceal the more detailed information on the active electrical network in the studied systems.

In the work reported here we tried to explain the non "single-smooth" experimental observations by presenting a simple quantitative lattice percolation model and its extension to the case of non lattice systems where spherical conducting particles are distributed in an insulating matrix. Then, we present a verification of our model by comparing its predictions with the results of our measurements on a granular metal and a carbon-black composite. We will also suggest a further extension of the model that can be applied to other systems such as carbon nanotube (CNT) polymer composites. Our models are based on the assumption of the presence of a hierarchy of the local conductances in both, the lattice and the continuum. Since the conspicuous feature of the models is a staircase of equation (2)-like relations we will refer to it then below in general, as *the percolation staircase model.* We will call the specific model in which we assume interparticle tunneling conduction, the *tunneling percolation staircase model.* While following our basic staircase model [8] we previously presented a simulation confirmation of it for lattices [19], we do not know of a simple transparent analytic formulation of the model or an extension of it to composite like systems. In particular, we do not know of any attempt to analyze experimental data that takes into account the presence of a staircase in the observed $\sigma(x)$ dependence. Most importantly, no connection was made between the reported universal ($t = t_{\mathrm{un}}$) or non universal ($t > t_{\mathrm{un}}$) behavior of the conductivity and the observed staircase like behavior. The purpose of the present article is then to deal systematically with the staircase behavior and to utilize the understanding that we derive to explain the unexpected equation (2)-like behavior that is usually observed in composites, in spite of the fact that all the particles in them are electrically connected by tunneling. In turn we expect that our findings will provide a framework that is common to percolation and hopping theories.

The structure of the paper is as follows. In Section 2 we present the simple physical basis of the percolation staircase models and its manifestation in the tunneling percolation problem. We do that by quantifying a very simple lattice model that we have sketched qualitatively previously [8] and by extending it to continuum systems. In particular, we provide specific criteria by which experimental results obtained on composites reveal details of the conducting network within these systems. In Section 3 we present experimental confirmations of the continuum extension of the lattice model that we developed in Section 2. Finally, in Section 4, we evaluate the results of Section 3 and discuss possible extensions of the model to systems of non spherical particles. This enables, in particular, to explain the many data of conductivity stairs in systems of CNT-polymer composites. Finally, we will be able to conclude that the observation of a percolation behavior with a non zero percolation threshold in composites is always associated with the presence of a cutoff or a hierarchy in the values of the conductances of the conducting elements in these systems.

2 Theoretical considerations and predictions

In order to present the basic physical idea of our approach we start by considering a simple-general system that consists of four types of local conductances g_i with values $g_4 \ll g_3 \ll g_2 \ll g_1$ and corresponding concentrations N_1, N_2, N_3 and N_4. Now let us assume that $N_1 < N_c$ where N_c is the concentration of the conductors that is needed for the onset of a globally connected (i.e., percolation) network. Obviously, the network of only the g_1 conductors cannot conduct. Now let us assume that $N_1 < N_c$ but $N_1 + N_2 > N_c$. In this case, the g_1 conductors are practically shorts while the g_3 and g_4 conductors (that may be connected in all sorts of parallel configurations to the combinations of g_1 and g_2 conductors) are practically insulators so that they do not contribute significantly to the global conductance, σ. Hence, in the system described, the global conductance will be proportional to g_2 [8,20,21]. Similarly, if $N_1 + N_2 < N_c$ but $N_1 + N_2 + N_3 > N_c$, the global conductance will be determined by the value of g_3 while the values of the other g_i's will not be reflected by the global conductance of the system. In general, we conclude then that the smaller the sum of the N_is, the smaller that is needed for the global network conductance, the smaller the value of g_i that will dominate this conductance. The above picture predicts a series of "down jumps" in σ with the decrease of $\Sigma N_i (\equiv N > N_c)$ that will take place when, $N_1 = N_c$, when $N_1 + N_2 = N_c$, etc. We call the interval of N values that is dominated by the value of one of the given local g_i's a stair.

To consider the above picture more specifically we examine a lattice where the number of the jth nearest neighbors is z_j, and the "total" number of its neighbors, up to and including the nth neighbor, is Z_n by $Z_n = \Sigma z_j$. Let us evaluate now the connectivity of the system when we assume that only the first n nearest neighbors ($1 \leqslant j \leqslant n$) are involved. The random occupation of the bonds with a probability p^b yields that the average number of occupied bonds, of the jth neighbors per site, will be $p^b z_j$. Correspondingly, the total average number of occupied bonds per site, including all the occupied bonds that are associated with the first n nearest neighbors, is $p^b Z_n$. The dependence of the percolation threshold on Z_n follows simply the topological requirement that, for a given dimension, there is a universal number of bonds per site B_c that is needed for the onset of percolation. The necessary probability for the bonds to be occupied, in order to achieve the onset of percolation, is then $p^b_{nc} = B_c/Z_n$ [2]. We note in passing that in the classical percolation theory one usually considers only the $n = 1$ case [1]. Following the increase of Z_n with $n > 1$ (i.e., the involvement of farther neighbors in the lattice) it is obvious that a corresponding series of percolation thresholds, p^b_{nc}, can be obtained and that $p^b_{nc} < p^b_{(n-1)c}$. The above considerations apply also to the site percolation problem since the involvement of more participating neighbors for the same p will yield a larger number of occupied bonds per site. Hence, we have that the corresponding percolation thresholds in the latter case, the p_{cn}'s, will also obey the rule that $p_{c(n+1)} < p_{cn}$ [2,3,22]. Of course p_{c1} corresponds to the classical first neighbor percolation problem and (for the convenience of the following discussion) we define here also p_{c0} as 1.

Let us consider now a hierarchy of the local-bond conductors by assigning relatively smaller conductances to the farther neighbors. In general, we can associate the local conductance between a site and its nth neighbor, g_n, with their intersite distance, ℓ_n. In particular, in the interesting case of tunneling, $g_n = g_0 \exp(-2\ell_n/\xi)$, where g_0 is a constant and ξ is the tunneling decay distance [8,23]. In that case one can follow the global conductivity of the lattice, $\sigma(p)$, with the decrease of p, by noting that for all n's $\ell_n > \ell_{n-1}$ and assuming initially that for all $n > 1$, $\ell_n/\xi \gg 1$. The global conductance $\sigma(p)$ for p in the interval $p_{cn} < p < p_{c(n-1)}$ could be expected [3], as in other cases with a wide distribution of local conductances [8,20,21], to be dominated by g_n. Thus the overall $\sigma(p)$ dependence should have a conductivity "jump" around each p_{cn}, exhibiting a *staircase dependence* such that between the p_{cn}'s (i.e., in the p range within a "stair") a monotonous increase of σ with p will follow equation (1) with the proper $p - p_{cn}$ and with a t that is equal to t_{un}.

In the above we have used the term "dominating" conductance intuitively and thus before a further presentation of our staircase model let us dwell on this concept within the framework of the lattice that we consider, from two points of view. From the topological point of view the conductors that are associated with the nth nearest neighbor bonds are necessary for percolation within the entire p range of the nth stair ($p_{cn} \leqslant p < p_{c(n-1)}$). This means that we can have throughout this range various configurations of conductors but there is no configuration that will yield percolation without having in each conducting path at least a single g_n conductor, or a combination of g_n-only conductors, that will not be shunted by the g_m's for $1 \leqslant m < n$. The second aspect of the dominant conductance, g_n, is that it is assumed to have a value that is much smaller than that of the other ($1 \leqslant m < n$) possible conductors in the system. It is obvious then that the resultant conductivity of the system and its role as the dominant conductance in a stair will be determined not only by the topology but also by the g_m/g_n ratios in the system. In general then, for a given system's connectivity, the overall electrical conductivity of the system will be determined by an additional constraint that the special properties of the electrical conduction impose on the topological network. In the electrical case this yields that the "resultant" conductivity is essentially controlled by a subsystem of the whole network. The effect of this constraint is manifested in particular when there is, as in our model of conductors, a distribution of the values of the local conductances, and this brings to "the physical idea that (in the case of a distribution of the values of the local conductances) the bottlenecks determine the overall conductance of the network" [20,21]. In other words, the constraint imposed by the conductance value distribution on the overall conductivity of the system leads to the selection of a particular "dominating" electrical network from the many topological equivalent networks that can be present in the system. However, it was noted that this conclusion "cannot be quantitatively valid for arbitrary conductance distribution" [20,21]. In our case, the bottlenecks are the g_n's since in the p-range of the nth stair there will be no percolation without the g_n's and since these have the lowest local conductance in the system. On the other hand we do not know a priori if the assumption of the domination of g_n throughout a stair (i.e., that the resultant conductivity there Θ_n, is simply proportional to g_n) is valid for the lattice system that we describe. In particular, in our model the inclusion of the nth nearest neighbors (i.e., that $p^b_{cn} = B_c/Z_n$) means also the inclusion of all the nearer neighbors, $1 \leqslant m < n$ (but not farther neighbors). Hence, using the relation $\Theta_n \propto g_n$, suggests that except for some configuration-dependent factor the resultant macroscopic conductivity, in a given stair range, is determined "only" by its g_n, while in the classical percolation theory the conductivity of the system is determined by the average local the system [3–5]. In the Appendix we discuss this point in detail showing that the use of the $\sigma_n \propto g_n$ relation is well justified for the typical $g_{(n-1)}/g_n$ ratios (10^3) considered in our model and experimental results throughout this work. In passing we also note that the fact that the g_n conductors may be connected with other g_n's or with g_m's in series or in parallel is accounted for by the universal value of the t exponent that we use below. The model that deals with the electrical conductivity in percolating systems and that essentially addresses this particular issue is known as the links nodes blobs (LNB) models [1,3,23]. A brief introduction to this model and its consequences regarding parallel configurations of conductances are given in the Appendix. Here, for the sake of simplicity of the quantitative of the presentation of our model, we assume then that $\Theta_n \propto g_n$ relation is indeed valid.

Turning to a quantitative description of the above model let us estimate the macroscopic conductivity within the nth neighbor stair where the conductivity is assumed to be dominated by the g_n conductances as follows. We approximate then the conductivity in this stair by $\sigma_n[(p-p_{cn})/(1-p_{cn})]^t$, where σ_n is the value of the conductivity that would have been obtained for the corresponding network of conductors if this dependence would have been extrapolated to $p = 1$. While we know that this approximation does not really hold for large $p - p_{cn}$ values [1], this convenient normalization of the conductivity does not affect any of the semiquantitative consequences of the model that will be concluded below. Applying our assumption regarding the dominance of g_n in the global conductance within the nth stair we write then that $\sigma_n = A_n(p)g_n$ where $A_n(p)$ is assumed to be a weakly dependent function (in comparison with $(p-p_{cn})^t$) of p. The above assumptions enable us to express the conductivity in the nth stair by the percolation modified step function:

$$\Theta_n(p) = \begin{cases} \sigma_n[(p-p_{cn})/(1-p_{cn})]^t & \text{if} \quad p > p_{cn} \\ 0 & \text{if} \quad p < p_{cn}. \end{cases} \tag{3}$$

While for each stair the conductivity is assumed then to be dominated by the particular local conductance g_n, the contribution of the lower conductance stairs to the overall conductivity of a given stair has to be considered when one calculates the "actually measured" conductivity of the system. Correspondingly, the global-resultant conductivity in the mth stair, $G_m(p)$, can be approximated by:

$$G_m(p) = \sum \Theta_k(p), \tag{4}$$

where we sum over all the $m \leqslant k \leqslant \infty$ step functions. As apparent from equation (3), with increasing n, the terms in the sum become smaller as do the g_n's, and the σ_n's. A more detailed discussion and more refined expressions for Θ_n are given in the Appendix.

Turning to the global $\sigma(p)$, the consequence of the application of equation (4) can be appreciated as follows. The smaller the $g_m/g_{(m+1)}$ ratio, the smaller will be the corresponding "jump" of the global conductance. Also, assuming a priori a $t = t_{\text{un}}$ in equation (3) yields that for $p > p_{cm}$, but very close to p_{cm}, the $\sigma(p)$ dependence will show some deviation from the universal behavior, while if larger p's in this mth stair are considered this deviation (when best fitting is attempted) will be negligible.

To give a tangible description of our model let us consider a lattice in which, either the occupation of the nearest neighbors is enough for the onset of global conductivity, or, the occupation of both, the nearest and next nearest neighbors is needed for the onset. We do that in Figure 1, by examining a sketch of the above two scenarios in a finite small section of an "infinite" square lattice. The sites of the lattice are presented by the various dots and circles in the figure. Now let us occupy the sites with a probability p such that we have occupied (large circles) and unoccupied (dots) sites. We define first the nearest neighbor connectivity criterion as follows: two sites are considered connected if they are nearest neighbors and occupied. Such connected pairs of sites yield bonds and each bond has a given conductance (or resistance) assigned to it. The corresponding bonds in the system are denoted in Figure 1 by resistor symbols. A similar consideration applies to the second nearest neighbors. The hierarchy of the local conductances is introduced then by assuming a relatively large first nearest neighbor bond conductance (relatively high-value resistors, thin resistor symbols) and a smaller second nearest neighbor bond conductance (with relatively high-value resistors, bold resistor symbols). As seen in the figure, the nearest neighbor resistors constitute a connected network between two opposite edges of this small portion of the "infinite lattice". Now let us reduce the site occupation probability p by assuming that the new network is the same as above, except that some (bright, blue circles) sites are excluded so that the system of the nearest neighbor bonds (or resistors) does not provide a connected network. Yet, the inclusion of the conductances associated with the second nearest neighbors, in addition to the prevailing nearest neighbors, does still provide a connected network. In this latter case the global conductivity will be determined by both types of resistors and if there is a significant difference between their values the high resistor (lower local conductances) will dominate the global conductivity. We see then that in the infinite lattice there will be well defined values of p and p_{c1}, such that for $p > p_{c1}$ there is a global connectivity of the system of nearest neighbor bonds while for $p < p_{c1}$ there is no such connectivity. However, the involvement of the second nearest neighbors provides, for $p < p_{c1}$, a continuous network, that is dominated by the resistors of these neighbors, and, again, this will hold for p values only down to some threshold $p_{c2} < p_{c1}$, for which the combined system of

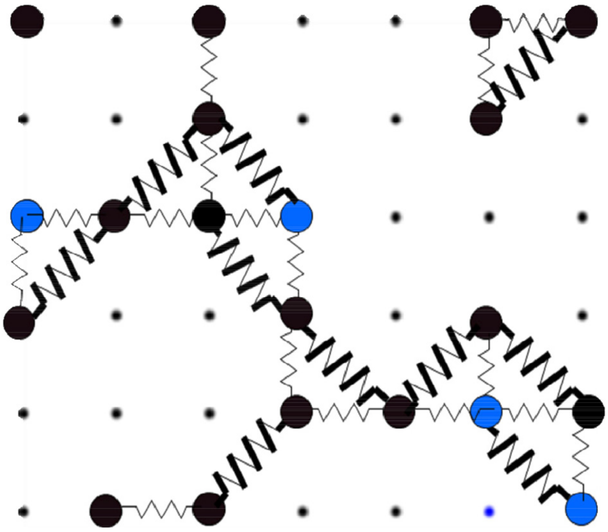

Fig. 1. An illustration of a small portion of a square lattice where some of the sites (large circles) are occupied. Two nearest neighbors that are occupied yield a bond with a relatively small resistance (thin resistor symbols). Two second nearest neighbors that are occupied are connected by resistors with a relatively high resistance (bold resistor symbols). If some of the occupied sites (light-blue circles) are removed the network of nearest neighbors will not yield a globally conducting system while if the resistors between the second nearest neighbors are also included the global network will conduct.

nearest neighbor and second nearest neighbor bonds provides a connected network.

Now let us consider the predictions of our quantitative model that is given by equations (3) and (4) for the simple case of the above infinite square lattice with a significant hierarchy of conductors, as can be the case when the inter-site conduction is by tunneling. To make the presentation of this model as clear as possible we make some simplifying assumptions in the choice of the model parameters. Starting from the series of percolation thresholds, it is well known [2] that the thresholds derived for the site percolation problem are numerically close to the bond percolation thresholds. For example, in the 2D (near neighbor only) square lattice $p_{c1} = 0.59$, which is not too far from the corresponding bond (near neighbor) percolation threshold, $p^b_{c1} = 0.5$. Following that and the fact that the bond percolation thresholds, p^b_{cn}, have the much more topologically transparent numerical values [2,22] than those of the percolation site problem [24], we "adopt" the bond percolation values as presenting the "proper" site percolation thresholds, p_{cn}. The topological transparency follows the above mentioned simple fact that $p^b_{cn} = B_c/Z_n$ where B_c is the critical number of bonds (2 in 2D systems) [2], while for the p_{cn} there are more complex approximations [24]. Hence, for the p_{cn} values in the square lattice illustration (up to $n = 6$ here) we use the corresponding $p^b_{cn} = B_c/Z_n$ series ($n = 1, 2, \ldots, 6$) that is then, 1/2, 1/4, 1/6, 1/10, 1/12, 1/14. The important point to note here is the shrinking of the widths of the expected stairs, $B_c(1/Z_{(n-1)} - 1/Z_n)$, with increasing n (where we note that for the width of the first stair, $n = 1$, one has, formally, to take $Z_0 = 2$). Let us point out that the same consideration applies for 3D lattices but, since the width of the stairs is much larger in the 2D case (the Z_n's are much smaller) we have chosen, for the sake of a clearer illustration, to present our model on the square lattice system.

Having defined the p-intervals, i.e., the stairs, by the percolation thresholds, let us consider now the local tunneling conductance associated with a given nth nearest neighbor (or the nth shell of neighbors) $g_n = g_0 \exp(-2\ell_n/\xi)$ that dominates the network conductivity in the nth stair. The corresponding contribution of the nth stair conductance will be determined then, according to equation (3) with $\sigma_n = A_n g_n$. Now, assuming for simplicity that the A_n values do not vary significantly with n we can approximate the "down jumps" between the stairs i.e., between the σ_n values by $g_n/g_{n+1} = \exp[2(\ell_{n+1}/\ell_n)/\xi]$. In the square lattice example that we consider (starting with the second stair, i.e., from $n = 1$), the corresponding $\ell_{n+1} - \ell_n$ increments, in units of the lattice constant a, are 0.4, 0,6, 0.24, 0.6 and 0.17. To evaluate these σ_n's with the increase of n we assume in our calculations that $2a/\xi = 10$ and normalize σ_6 to be 1. The interesting observation here is that since the width of the stairs $\Delta p_{cn}(= p_{c(n-1)} - p_{cn})$ is reduced considerably with increasing n, the variation of the global $\sigma(p)$ dependencies will appear as dominated by the decreasing g_n's rather than by the conducting network (as in the LNB model, see the Appendix) variations of the

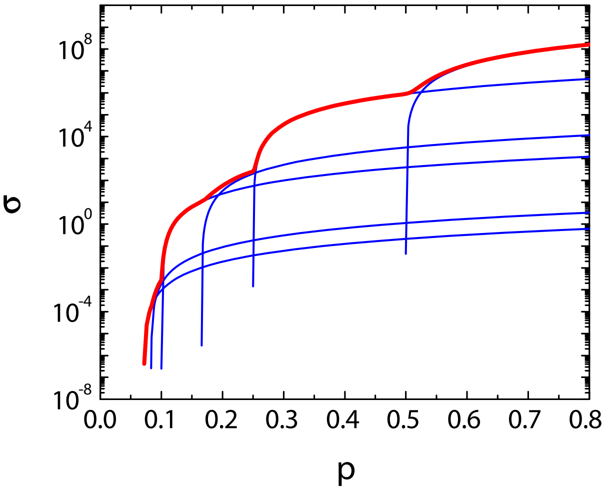

Fig. 2. An illustration of the expected staircase behavior of a normalized percolation tunneling conductivity in a 2D square lattice. Both, the separate stair functions (Eq. (3), for $n = 1, \ldots, 6$) and their sum (Eq. (4), bold red curve) are shown in the figure for the first six stairs. This illustration was derived for $2a/\xi = 10$.

classical percolation theory [1,3]. Indeed, as shown below (in Fig. 3), this effect will be manifested by an apparent non universal behavior i.e., by a t exponent larger than t_{un}.

Having selected the parameters let us present now the corresponding predictions of equations (3) and (4) in Figure 2. In this figure we tried to accommodate as many stairs on the one hand, but to have as clear as possible separation between them, on the other hand. To make the behavior more reminiscent of composites that are 3D systems we have preferred (in spite of the 2D lattice that we have chosen) to use the 3D value of $t_{\mathrm{un}} = 2$ for the plot of the separate stairs according to equation (3). In the figure we show then the upper six conductivity stairs, i.e., the functions Θ_n for $n = 1$–6. Also shown is the envelope function $\sigma(p)$ (heavy red curve), which is determined in the range of stair k by $G_k(p)$ as given by equation (4).

In Figure 2 the behavior of $\sigma(p)$ for $p > p_{c1}$ is due to the contribution of two parallel conductivity networks, one of g_1-only conductors (Θ_1) and one of a network that is made of a mixture of g_1 and g_2 conductances (Θ_2). The $n = 2$ curve represents then the extrapolation of the virtual second stair behavior to the $n = 1$ stair regime. This extrapolation is the contribution of the $n = 2$ percolation network if the percolation cluster of the g_1-only conductors is eliminated. Obviously since the two contributions are in parallel they add up yielding the resultant "conductivity envelope" in the first stair regime. Note that the envelope is the overall system's conductivity that will be actually measured or simulated (concealing the relative contributions of the two networks of which it is made). We see then that (as also expected from the construction of our model and as discussed in the Appendix) the ratio of the $\sigma_n(p)$'s, for a given p farther above p_{c1}, will be of the order of g_1/g_2.

It can be seen that, due to the contribution of the lower conductivity stairs, the resulting $\sigma(p)$ dependence

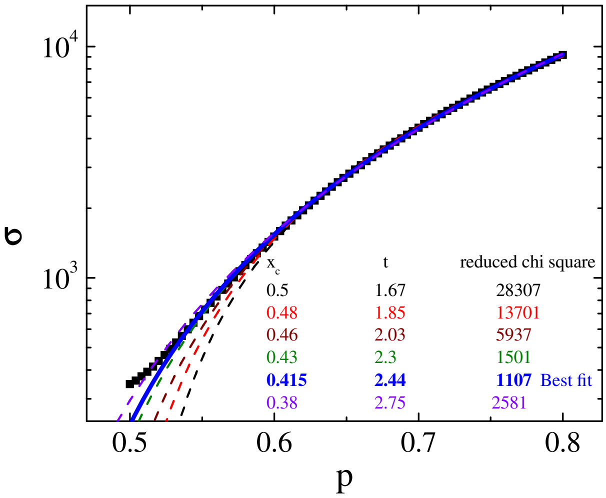

Fig. 3. The analysis of the behavior of the normalized conductivity in the region of the first stair as derived by the application of equation (4) to data such as the conductivity envelope in Figure 2 but for $2a/\xi = 5$.

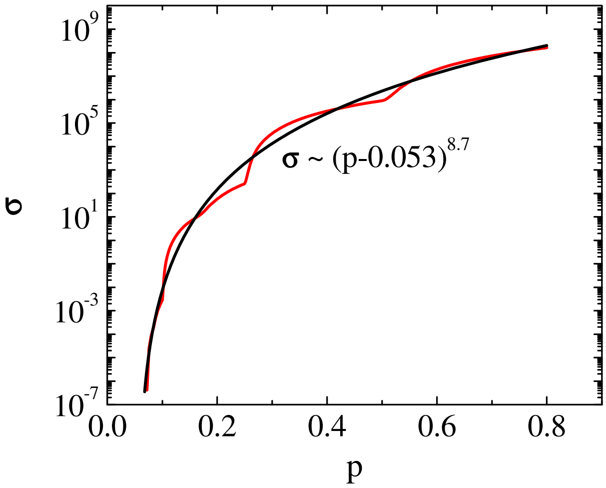

Fig. 4. The fitting of the conductivity envelope shown in Figure 2 to an equation (1) dependence.

deviates from the single Θ_n in each stair, mainly close to the corresponding percolation threshold p_{cn}. We also see that the larger the conductivity jumps, the smaller the effect of the $n+1$ (lower conductivity) stair on the conductivity of the nth (higher conductivity) stair. The importance of the latter result is that the conductivity contribution of the lower stair affects the t values that will be derived for the following higher conductivity stair. This is since the lower conductivity stair causes an upper shift of $\sigma(p)$ around p_{cn}, and this leads to an apparent lower p_{cn} value and a corresponding apparent larger t value. This behavior is demonstrated in Figure 3 where different p_{cn} values are tried in order to fit the $\sigma(p)$ (or the envelope $G_1(p)$ at the first stair, with data that were obtained as in Figure 2, but (for clarity) with $2a/\xi = 5$. We see that the best fit (see the reduced chi square values) that is presented by the solid (blue) curve in the figure, yields now a value of 0.415 for p_{cn} and 2.44 for t, rather than the "original" imposed $p_{cn} = 0.50$ and $t = 2.0$ values. Hence, the effect of the participation of the smaller g_n's as p decreases is to shift the percolation threshold for a given set of data to lower values than the value that would have been observed without the participation of the lower g_n's. In order to account for an equation (1) like behavior for such a set of data the t value must increase. This is since the corresponding $p - p_c$ interval has increased then (note that all the p's are smaller than 1). The end result is that the increase of the t value beyond its universal value is associated with the presence of smaller g_n values below the p range of a given set of data. This decrease of the g_n values as p decreases is however the basis of the a non universal behavior in the tunneling percolation problem [3,5]. In particular, if the observed decrease of the dominating g values (with the decrease of p) in the system can be presented by $g \propto (p - p_c)^u$ for some p_c value, we get that $t = t_{\text{un}} + u$ in accord with the general theory of the non universal behavior [3,4]. We conclude then that when t values somewhat larger than t_{un} are derived from experimental data, by fitting them to an equation (1)-like behavior, they simply reflect the non negligible contribution of the smaller conductors in the system. In passing we note that the presence of the smaller (non dominant) g_n's in a given $(n+1)$ stair is usually ignored in many previous analyses of experimental data. This is apparently, since the lower conductivity stair, where these smaller g_n's are dominant, was not detectable. On the other hand, as shown here, the contribution of these g_n's to the higher conductivity stair that is revealed by the data may be important.

In general, the results of computer simulations and experimental measurements are given essentially by the data points of an "envelope" such as the (bold, red) curve shown in Figure 2. However, the analysis commonly used in the literature is to fit the data to a single stair dependence by the application of equation (1). This kind of analysis, that ignores the deviations from the smooth equation (1)-like curve, yields a single p_c and a single t. For example the best fit of the data points of the "envelope" in Figure 2, to a single equation (1) dependence yields, as shown in Figure 4, a percolation threshold of $p_c = 0.053$ and a large $t = 8.7$ value. This behavior confirms the suggestion made above that while the individual stairs may exhibit a universal behavior (or a behavior close to it) the general trend of decreasing g_n's leads to the observed non universal behavior. In particular, since $t > t_{\text{un}}$, we can account for this decrease of the g_ns as yielding the non universal behavior when the "effective" g_n behaves as $g_n \propto (p - p_{cn})^u$, our case $u \equiv t - t_{un} = 8.7 - 2 \approx 6.7$. Moreover, this u value can be mapped onto the original $f(g) = (1-\alpha)g^{-\alpha}$ distribution function of Kogut and Straley [4] with $\alpha = u/(1+u) = 0.87$. We also note that while the best fit of the limited data of the envelope extrapolates to $p_c = 0.053$, the "true" threshold of the tunneling system is the one that would have been obtained by equation (4) for $k \to \infty$, i.e., $p_{ck} = 0$. Fitting the envelope of the data in Figure 2 with this $p_c = 0$ value yields (as discussed following Fig. 3) to a somewhat larger non universal value of t ($\approx$11).

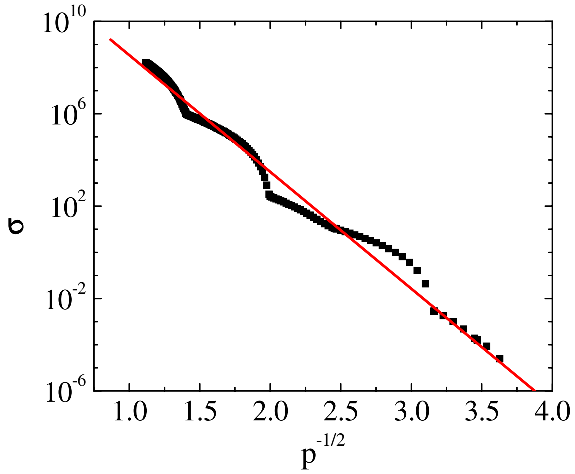

Fig. 5. The data points of the conductivity envelope in Figures 2 and 4, as a function of the distance between occupied sites, that is presented here by the $p^{-1/2}$ scale of a 2D lattice.

Following the non universal behavior that we obtained by taking the $p \to 0$ limit let us turn to examine the tunneling hopping prediction in that limit of dilute site occupation. It is known that the distribution function of the interparticle distance when applied to 2D in hopping theory [23] yields that $\sigma \sim \exp(-\omega p^{-1/2})$ (where ω is a constant [8,23]) and thus we also examined whether that behavior will be manifested by the envelope shown in Figures 2 and 4. For the examination of the asymptotic behavior of this envelope as $p \to 0$ we plot in Figure 5 the data points of the envelope as a function of the inverse density of the occupied sites in the 2D lattice, $p^{-1/2}$. The straight line obtained as $p \to 0$ does indeed indicate that $p = 0$ is the ultimate threshold of the system and that the hopping theory of randomly implanted sites applies in this dilute limit of the lattice. We conclude then that the basic physics that determines the non-universal-like and the hopping-like behaviors in this dilute limit ($p \to 0$), is that the $\sigma(p)$ behavior is dominated by the decrease of the conductances rather than by the decrease in the connectivity of the system [8].

The four predictions of our above tunneling staircase model that were illustrated in Figures 2–5 are then: (a) each stair is expected to be practically described by equation (1) with its proper p_{cn}, and, for large local conductance jumps, with $t = t_{\rm un}$. The smaller the jumps, the larger will be the effect of the lower conductivity stair on the critical behavior of the subsequent higher conductivity stair. This leads to a lower fitted p_{cn} and a larger fitted value of t with respect to $t_{\rm un}$. (b) The conductivity ratio between the stairs will be of the order of $g_n/g_{n+1} = \exp[2(\ell_{n+1} - \ell_n)/\xi]$. (c) The participation of lower and lower conductances as $p \to 0$ can be presented by a $g_n \propto (p - p_{cn})^u$ dependence where p_{cn} is the corresponding threshold. Such a dependence yields the non universal behavior with $t = t_{\rm un} + u$. (d) The "true" percolation threshold of the system is at $p = 0$. As in the hopping transport the closer the p to 0, the more hopping-like the system gets. This behavior, that is well demonstrated by a $p^{-1/2}$ dependence in 2D or the corresponding $p^{-1/3}$ in 3D, is due to the fact that when the lattice site occupation becomes very dilute it is essentially that of randomly implanted sites. One should note here that this result follows the fact that in the p range where this behavior is found, the variation in the g_n's (that is manifested by $u > t_{\rm un}$) is much larger than the variation of the conductivity due to the change of the connectivity (that is manifested by $t = t_{\rm un}$) with the decrease of p.

The extension of our lattice tunneling-percolation staircase model to systems of conducting spheres that are embedded in an insulating matrix appears to be relatively simple. In that case the hierarchy of the local conductances, g_n, will be determined by the radial distribution function (RDF) of the particles distances around a reference particle [25]. Of course this function depends on the assumptions made in the simulation models [26–28] or on the preparation conditions of the systems. In particular, in real composites the RDF depends on the value of x, on the components of the composites and on the fabrication process. For simplicity, we distinguish between two cases assuming that in both of them the values of the local conductances, g_n, decrease monotonically with the interparticle distance. In the first case, the RDF has well defined narrow peaks, and in the other, the RDF is a smooth monotonic function. Let us consider then the conductivity in these two cases when the interparticle conduction is by tunneling. In the first case the peaks in the RDF of the interparticle (surface to surface) distances are the equivalents of the ℓ_n's defined for the lattice. Indeed, distinguishable peaks of the RDF represent clear shells of near neighbors [25] and as the density of the spheres increases, the spheres arrangement approaches that of a lattice [27,28]. Hence, a corresponding hierarchy of local tunneling conductances, associated with the interparticle distances ℓ_n, is likely to occur. Following our discussion on lattices we expect then conductivity stairs in these systems. In particular, the magnitude of the conductivity "down jump" between the n and the $n+1$ stair in $\sigma(x)$ (i.e., $\sigma_m/\sigma_{(m+1)}$) is expected, as for lattices, to be given by $\exp[2(\ell_{n+1}-\ell_n)/\xi]$. Turning to the second case of no peaks, or relatively wide peaks, the distribution of the ℓ_n values (that determines the distribution of the g_n's) will lead to a smooth (single stair like) $\sigma(x)$ dependence which is actually the envelope of the hardly distinguishable or quasi continuous series of stairs. Following our predictions for the lattice model this may also lead to a deviation of the value of t from its universal value in the $\sigma(x)$ dependence. Thus, one will obtain, computationally or experimentally, a single universal ($t = t_{\rm un}$) stair, or a single-like stair with a $t \geq t_{\rm un}$ value. Having these guidelines and the above (a)–(d) predictions will enable us to understand the computational and experimental results that are obtained for various composite systems.

3 Experimental considerations and results

As mentioned in the introduction there are many systems [10–18] in which conductivity stairs have been found

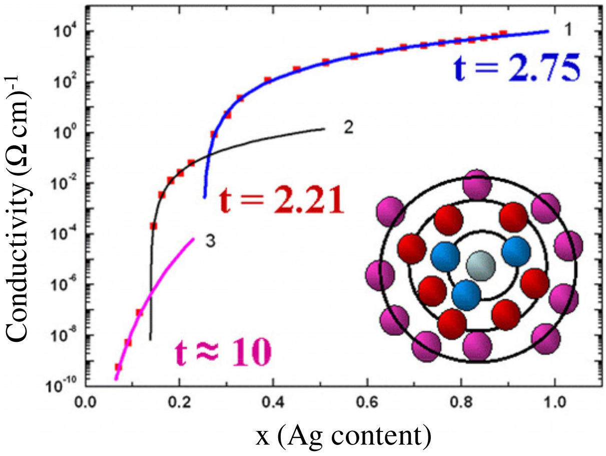

Fig. 6. Our experimental results of the electrical conductivity dependence on the silver fractional volume content in granular Ag-Al_2O_3 composites. The fitting of the data to equation (2) for the three stairs (1, 2, 3) and the corresponding fitted t-exponents are also shown. In the figure we also illustrate the well-known shell structure of near neighbors in a three dimensional ensemble of hard spheres. The circles represent the average position of the first, second and third nearest neighbor shells.

experimentally but have not been analyzed, accordingly. These systems are of different complexity and the relation of each of them to the simple model suggested in Section 2 is yet to be determined. It is apparent that systems of spherical conducting particles that are embedded in an insulating matrix are the most reminiscent of our simple model and thus would be the most convincing ones in providing evidence for the validity of the model.

In trying to obtain convincing experimental evidence for the existence of a percolation-tunneling staircase we looked for a system that consists of spherical or nearly-spherical conducting particles so that the predictions ((a)–(d)) of the lattice model presented in Section 2 can be tested. However, before choosing such a system we noted that the theoretical calculations of the RDF of non touching collections of hard spheres, as illustrated schematically in the inset of Figure 6, seem to put a severe experimental constraint that in practice does not enable to measure more than a couple of stairs. This constraint is that the distance between the RDF peaks [23-26] should not be too large compared to the tunneling decay length, ξ, which is of the order of 1 nm [29–31]. If this condition is not met, the measurement of the lower conductivity stairs will be quite difficult since it is associated with very high tunneling resistances to the farther neighbors. Thus we concluded that the predictions of our staircase model can be tested conveniently only in systems with particles size that is of the order of a few nm's.

The system that seems to fulfill the latter requirement is that of granular metals [10,32] for which samples of small particles, that are roughly of the same size [33], cover almost the entire x range and for which the existence of tunneling between the particles is well established [10,32]. However, as far as we know, no more than two stairs have been identified in those systems and the first one is usually attributed to particles coalescence [10,32–34]. Hence, a granular metal, for which at least three stairs are observed, seems promising for the confirmation of the predictions of Section 2.

Granular metal systems of nearly spherical metal particles embedded in an insulating matrix are relatively simple to prepare by the co-sputtering method [10,32–37]. This method has the advantage that it enables to prepare a quasi continuous set of samples that differ by relatively small increments of x where x is clearly proportional to the concentration of the grains. The challenge for the present work was then to prepare an appropriate granular metal i.e., a one for which we could expect at least three peaks in the RDF that will lead to the observation of three conductivity stairs. Following these considerations we noted that both Ag-SiO_2 [35,36] and Ag-Al_2O_3 [37] composites appear to consist of well separated grains that form near-neighbor like shells. The Ag-Al_2O_3 system seems, however, to have the advantage of consistency with the RDF models that usually assume equal size spheres [26–28]. This is since the Al_2O_3 matrix (for non annealed films [33]) tends to keep the average size of the metallic grains, 5.2 ± 1.8 nm [37], essentially independent of x in the interesting range of $0.1 < x < 0.4$ [35–37] which is below the grains coalescence transition. Hence, the first, second and third nearest neighbors shells in the Ag-SiO_2 [35,36] and Ag-Al_2O_3 [37] composites are reminiscent of those of the ideal models of spheres where the RDF has peaks [25]. Moreover, since in the granular systems of noble metals the first percolation thresholds are at relatively low x (<0.3) values, it may be that the first stair is already associated with tunneling, in contrast with the typical higher x values for the coalescence associated transitions in other granular metals [10,32]. In fact, the available structural data in references [35,37] show that the interparticle (surface to surface) distance to the first neighbor shell is of the order of 1–2 nm while the distance to the second neighbor shell is of the order of 5–6 nm and to the third neighbor shell it is 10–12 nm. This suggests that the $\sigma(x)$ dependence is not too sensitive to the assumption whether the first shell results from coalescence ($\ell_1 = 0$ and $g_1 = g_0$) or from a tunneling conductance between the nearest neighbors ($\ell_1 =$ 1–2 nm and $g_0/e^2 < g_1 < g_0/e$, where e is the base of the natural logarithms). Hence, the behavior of the first stair in these systems may provide additional support to the model (in addition to the behavior expected from the second and third stairs). In fact, the assumption that a first stair in those composites can be approximated by the tunneling model is further supported by the two conductivity stairs that have been observed [11] in systems of metal grains embedded in a polymer, where there is definitely no coalescence of particles.

In contrast with all the above considerations that seem to be in favor of examining the above granular systems [35,37], we noted that the observed shells of the RDF peaks in these systems appear for $x < 0.4$, while in the many computations of the RDF of ensembles of spheres [26–28] the conspicuous peaks appear for denser ensembles. Since a priori there may seem to be some disagreement here we suggest that there is a good reason for the slight discrepancy between the model computations and the actual structure of the granular composites. In the computations, the models used were of particles that are dispersed under equilibrium or non equilibrium (e.g., random sequential addition) conditions, while in the granular metals the possible arrangement of the particles follows the very different process of particles nucleation and growth. Trying to evaluate the effect of the latter process on the spatial distribution of the grains, we found that there is no derivation or a discussion of the RDF for co-sputtered metal-insulator composites. This is in spite of the fact that there are quite a few studies and interpretations [38–40] of the corresponding processes in metal-only deposited particles and even in those deposited by sputtering. Since this material issue is far beyond the scope of the present work and since we are concerned here only with the outcome of the deposition process i.e., the experimentally observed RDF in the granular metals of interest [35,37], we do not elaborate further on this issue. The important observation from the point of view of the present work is that, in both, Ag-SiO_2 [35,36] and Ag-Al_2O_3 [37] systems, the above peaks in the neighbor-shells distance distribution and the observed percolation transitions in them, take place in the $x < 0.3$ range. This observation strongly suggests that the two are related in accordance with our expectations.

Following the above considerations we have co-sputtered Ag-Al_2O_3 samples under similar conditions to those used by other authors [10,36,37] and by us [34,41] for depositing various granular metals in general [32,33], and Ag-Al_2O_3 [37] in particular. As in our previous works with granular metals we deposited 1 μm thick films and the electrical conductivity measurements were carried out in the (two or four probe) coplanar configuration [41]. Indeed, as exhibited in Figure 6, we were able to find three clear conductivity stairs fulfilling the predictions of the lattice staircase model as listed in Section 2. First, the values of t (as marked in Fig. 6) of the upper two stairs are close to (but somewhat larger than) the universal value t_{un}, and the corresponding thresholds of the percolation transitions take place at x values that are very close to those of other granular metals where the metal was Ag or Au [10,32]. Second, the conductivity "jumps" between subsequent stairs are of the expected orders of magnitude, if we assume that $\xi \approx 1$ nm [29–31], as follows. Taking $\ell_1 = 1$–2 nm and $\ell_2 = 5$–6 nm (see the above structural data), the expected ratio between the values of the local conductivities of the first (1) and second (2) stairs will be $(g_1/g_2 = \exp[2(\ell_2 - \ell_1)/\xi]) \sim 10^3$. Considering then the proportionality between the global conductivity (σ_n) and the dominant local (g_n) conductance of the nth stair, we expect from Section 2 that $\sigma_1/\sigma_2 \sim g_1/g_2$ where the σ_n's are taken at a common x (but far from the percolation thresholds). Indeed, this is consistent with the data in Figure 6, where the corresponding conductivity ratio is of the order of 10^3. Similarly, the same expectation of 10^3 is also fulfilled by the ratio of the conductivities of the second (2) and third (3) stairs, since the average interparticle distance between the second and the third neighbors is also about 5–6 nm [37]. Third, if one tries to fit the three data points (that we managed to achieve in our samples for the third stair) to equation (2) one gets a value of $t \approx 10$, which is (as we concluded above for lattices) simply indicative of the dominant role of the decrease in the g_n values as x (or p), approach the dilute limit. Hence, the corresponding $\sigma(x)$ dependence appears as a single "non universal" ($t > t_{\mathrm{un}}$) behavior [4,5,42]. Fourth, high t values translate, as we have shown in Figure 5, to a good agreement with the hopping model. Of course, we note the fact that we have here only three data points for the third stair does not allow us to demonstrate convincingly the expected high t and hopping like behavior. However, a more convincing evidence for this prediction will be provided below.

As pointed out in the introduction there are many systems where conductivity stairs are observed experimentally but their structure is quite different from that of granular metals and, correspondingly, from the very simple model that we proposed in Section 2. While one can think of a more special extensions of our model (see Sect. 4) we would like to widen here the base of our simple model to other systems of spherical particles but for which a hierarchy of the tunneling conductances is dominated by the microstructure of the insulating matrix rather than by the global RDF of the conducting spheres. As will be shown below this scenario is of particular relevance to carbon black polymer composites which are a prominent example of the most studied electrically conducting composites [7,13,34,41]. In these composites (unlike the example of granular metals) the size of the conducting particles is very large compared to the tunneling distance [34]. Hence, only the first near neighbors (say, along a given direction from the reference particle) are within the tunneling range and only they can contribute significantly to the global conductance. With the increase of x in such a system, the density of the particles increases and the average nearest neighbor tunneling conductance will also increase leading to the increase of the global conductivity. If the distribution function of the distance between a particle and its neighbors in the first shell, $h(r)$, is "smooth" (monotonic and having a monotonic derivative), we expect a "simple-single" $\sigma(x)$ dependence with no stairs and an exponent t, that depends on $h(r)$ [3,5,42,43]. Hence, in the ideal case of a smooth $h(r)$, as usually found in calculations or simulations [44,45], no stairs are expected. However, for some real composites, in particular for those that are prepared by melt molding of polymers, a local modification of the $h(r)$ function may occur due to some friction [5,15,16] and other interactions [46,47] between the particles and the polymer. In this case two particles may be separated by a few discrete polymer layers such that each layer is of the order of a nm. The layers that encapsulate the individual conducting particle will have then a nm scale width distribution [46–49] that is superimposed on the $h(r)$ distribution function. Thus, the discreteness

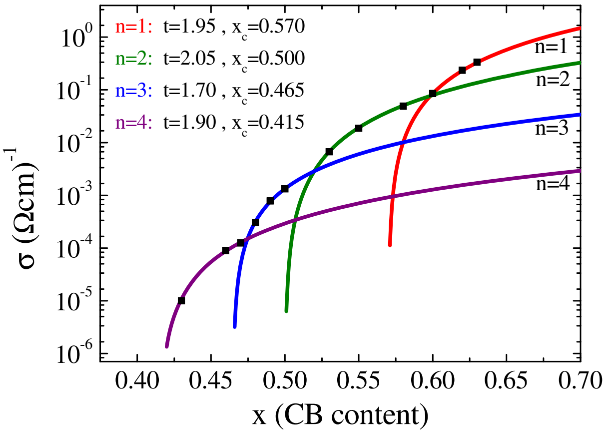

Fig. 7. The CB volume fraction dependence of the electrical conductivity in CB-polymer composites in which the CB particles are essentially spherical. The curves show best fits to the data, for each of the four distinct stairs, in the staircase. The corresponding stairs parameters are given by the table in the inset.

of these features may be reflected in the observed behavior of $\sigma(x)$. Following this possible scenario we turned to carefully examine the $\sigma(x)$ dependence of a carbon black polymer composite in which the carbon particles are nearly spherical with a diameter of 250 nm [43,50].

The composite samples used in our measurements were prepared and measured as previously described [43,51,52]. The results that we obtained on three sets of samples were very much the same. In Figure 7 we present the $\sigma(x)$ dependence that we obtained on one of these sets. As shown in the figure we can extract from this dependence four stairs of relatively small magnitudes. We note that while the total number of data points that we have is relatively large and dense in comparison with other relevant works, for each of the distinguishable stairs (see below) we have only three data points and thus we cannot extract accurate t values for them. However, we fitted (using the least squares criterion) the data for each stair by an equation (2) like behavior and, as given in the table within the figure, we got t values that are very close to the 3D universal 1.7–2.0 value. The fits that we show indicate that it is possible to associate the presented data with separate stairs.

Of course, at first sight, there is a difficulty involved in the analysis of the data and their interpretation as suggested by the curves in Figure 7. This is the possibility that the stairs in the figure simply reflect some fluctuations in the measurements. To eliminate this doubt and to convincingly show that the data reflect a genuine property of the tested composites we have carried out detailed measurements of the $\sigma(x)$ dependence of the same set of samples but by the completely different method of microwave transmission through the samples [53]. We have found that indeed the results of these latter measurements can be fitted, within the resolution shown in Figure 7, by stairs that have the same thresholds x_c that we observed in our dc conductivity measurements (as listed in the table within this figure).

Establishing this experimental amenability of our data analysis in terms of stairs let us examine the results shown in Figure 7 in light of the about 1 nm thick polymer layers that are known to be formed around particles in such composites [11,47,48,50,54]. In particular, the wrapping [55,56] or coating [57] of particles by the polymer elements, that can be small as 0.4 nm [58], suggest the formation of thin polymer shells around the conducting particles. It was even suggested that the thickness of the polymer layer between the particles is independent of x [47]. Also, there are various indications for the formation of subsequent polymer layers with different densities and possible different properties that may provide various kinds of tunneling barriers [47,59]. On the other hand, there are many indirect experimental results that suggest the dominance of a nm-distance related tunneling conduction mechanism in such composites. For example, the temperature dependence of the conductivity [54] and the well known electrothermal switching effect [60] are well explained by such tunneling conduction. Hence, it is not impossible that several “quantized”, nm thick, polymer layers (“onion like”) form around a CB particle and thus a series of different tunneling barriers separate between the near carbon particles. All this may correlate well with the almost equal and small stairs that we observed in Figure 7. Correspondingly, the variation or the jumps of the dominant conductances is at least an order of magnitude smaller than expected from the peaked RDF case that we encountered for granular metals.

Finally, let us consider the best fit to the entire data that were presented in Figure 7. This fit represents essentially the $\sigma(x)$ dependence that is expected from the smooth two particles $h(r)$ function dependence that follows the well known (structure less) distribution of the near neighbor interparticle distances [44,45]. As we have discussed previously in detail [5,43] the latter function yields a non universal behavior of the conductivity. Indeed, fitting the entire data in Figure 7 to equation (2) we find that in accordance with our expectation ((c) in Sect. 2), $t \approx 11$ (or $u \approx 9$) and $x_c = 0.3$. Not less important is the experimental verification here of the prediction ((d) in Sect. 2) made following Figure 5, that the non universal behavior in the tunneling-percolation theory [3,5] reflects the behavior expected from hopping theory [23], in particular when the local conductances decrease in a similar manner with decreasing x in the two theories [5,23,42,43]. This verification is demonstrated here, in Figure 8, by plotting the data of Figure 7 in accordance with the hopping like, $\sigma \sim \exp(-\omega x^{-1/3})$, dependence [8,23]. These results fulfill then our two expectations for a high t value and a hopping behavior, for which the data in Figure 6 was not as convincing.

4 Discussion

The observation of a percolation behavior when there is a finite tunneling conductivity between any two particles in a system (rather than a contact with a constant resistance and only between their nearest neighbors, as in the

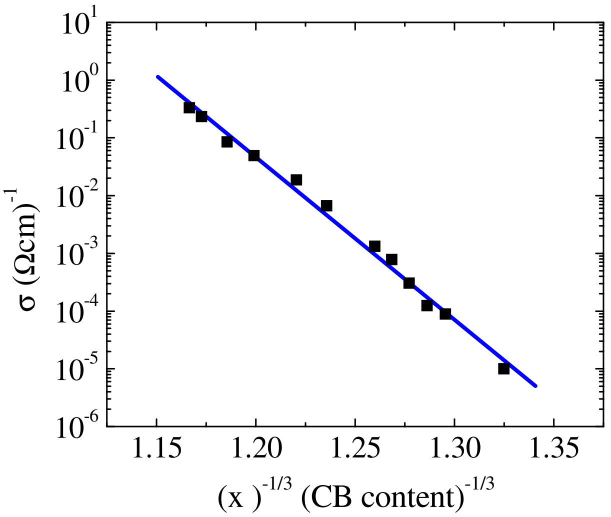

Fig. 8. The experimental data points of the conductivity in Figure 7, as presented by their dependence on the inter particle 3D distance scale of $x^{-1/3}$.

S&Z model [6]), is not trivial [7]. Our work enables then, beyond a better understanding of the electrical conductance in composites, to find an answer to the fundamental question that was raised in the introduction, i.e., how is it that the presence of tunneling does not eliminate the percolation critical-like behavior? For answering this question let us summarize the relevant conclusions that follow our staircase model. We saw that a single-smooth equation (2) like universal percolation behavior can be observed only when there is a single dominating local conductance value in the system throughout the studied x regime. In simulations, this is a result of the cutoff in the maximum interparticle distance that is taken into account, and in the experiments, this is a result of the limited global conductivity that can be measured. If there is an hierarchy of the local conductances and the corresponding global conductance is measurable in an x regime, within which the dominating local conductance varies, a conductivity staircase will be observed.

When the hierarchy is determined by interparticle distances, farther and farther neighbors are needed for the onset of percolation with the decrease of x. Since for tunneling the charge transfer probability decreases monotonically with interparticle distance, the dominating local conductance that must be involved for the onset of global conductivity in corresponding systems will become smaller and smaller. Hence, conductivity stairs such that each is characterized by its percolation threshold and its dominant local conductance will be obtained as a function of x. The more dilute the system, the denser will be the series of stairs and the fitted conductivity to their envelope will have an apparent non universal behavior. On the other hand, a hopping like behavior, well known for randomly distributed dots [23], is expected in the corresponding dilute system limit. In fact, we have illustrated that even if one starts from the discrete lattice percolation approach (as in Fig. 5), when the system becomes dilute or the dominant local conductance decreases continuously with decreasing x (as in Fig. 8), the behavior does become of the hopping type. We see then that hopping is always the dilute limit of tunneling percolation and in systems where the interparticle distance distribution is monotonic and continuous they are essentially the same. Correspondingly, our finding that in the tunneling percolation problem the "true" threshold is always at p (or x) $= 0$, shows that the finite percolation thresholds observed in computations of, or measurements on, composites, are a result of the limited conductivity data (i.e., only above a given finite x value) that are usually available. The corresponding $\sigma(x)$ dependence that is usually observed provides then only a part of the full connectivity-conductivity picture of the system. On the other hand, analysis of the given data in terms of the conductivity staircase, i.e., in terms of the built in hierarchy of the local conductances in the system, gives a more detailed picture of the percolation network. The answer to the question of why a percolation like critical behavior is observed in tunneling percolation systems, is that one usually monitors the conductivity in the relatively high density (and larger conductivity) first stair regime. Note that this regime is usually wide enough so that the observed critical behavior is not masked by the contribution of the lower conductivity stairs. In particular, this analysis in terms of the first stair, explains the rather general observation (as emerges from the statistics over numerous data on all types of composites [9,15,16]) that the conductivity values of the critical exponents, t, reported in the literature are quite narrowly distributed around the universal value of t_{un}. On the other hand, while relatively few (in comparison with that corresponding majority), there are still many works in which the values derived for t deviate significantly from t_{un} [9,15,16]. These $t > t_{\text{un}}$ values are well understood now to describe the envelope of the first or the first few conductivity stairs when the effect of lower stairs is not negligible (as in Fig. 3) for the measurable x range.

In Section 1 we mentioned the very many systems in which conductivity stairs were observed [10–18] but have not been analyzed accordingly. Many of these systems consist of non spherical conducting particles so that our models based on spherical particles do not simply account for them. However, the fact that for such systems stairs-like behaviors have been observed can be explained by the principle stressed throughout this article, i.e., that in such systems a hierarchy of the local conductances must exist. Of these systems there is quite a great interest in composites in which the conducting particles are carbon nanotubes (CNTs) [15,16] or metallic nanowires [61]. For such systems two scenarios have been suggested. In one, the local conductances are determined by tunneling [58] or by the resistance [62] between two "touching" particles, while in the other, the dominating local conductances are associated with the segments between subsequent contacts of two particles along a third one [63]. As one would expect a priori, and as we have demonstrated by corresponding simulations [64,65], in both cases an association of the global conductance with (practically [65]) constant values of the local conductances provides a good approximation for the system and thus no conductivity stairs are anticipated. However, surprisingly, in numerous composites of elongated particles such stairs are observed [14] and thus the implied hierarchy must be a scenario that is not associated with the interparticle transport mechanism. Noting that the effect of the particles dispersion due to processing is well known to determine the electrical properties of such composites [15,16,66] we suggest that in those composites the hierarchy of local conductances follows possible aggregation configurations such as the ones suggested by Hu et al. [66].

To illustrate a simple aggregation configuration that can result in conductivity stairs let us assume a percolation dispersion model where two types of local conductances are randomly implanted in an insulating matrix. For this version of a dispersion model we adopt the structure of the system that was experimentally found by Maaroufi et al. [67] for a CNT-polymer composite. They found that their composite consists of a distribution of isotropic CNT aggregates as well as individual non aggregated CNTs. Let us assume accordingly that the aggregates of the CNTs act as high conductivity spheres (that have a diameter which is of the order of the length of the single CNTs) such that the local conductance between two of them, g_a, is mainly due their touching. On the other hand, the conductances of a single CNT, or between two "touching" single CNTs or between a single CNT and an aggregate that are "touching", are of the order of g_s, such that $g_s \ll g_a$. Therefore, the conductance between two non touching aggregates that are "bridged" by a single CNT, is also of the order of g_s. Let us denote the concentration of the aggregates by N_a and the concentration of the individual, none aggregated CNTs, by N_s. We assume (as in the very simple model described in Sect. 2 and the general lattice model in the Appendix) that for the onset of percolation we need at least N_c particles (aggregates and individual CNTs) in the system and that for high enough x values we have that $N_a > N_c$ (or x_c), so that the contribution of the g_s's to the global conductance is negligible. It is clear then that for x values lower than x_c a global conductance can be achieved only with the participation of some of the g_s's. Correspondingly, the conductivity of the first (high conductivity) stair will be determined by the conductance of the aggregates (i.e., g_a) while the conductivity of the second (lower conductivity) stair will be dominated by the conductance of the individual CNTs, g_s. The onset of percolation of the latter stair will be then at a proper combination of both the aggregates and the individual CNTs that is enough to provide the connectivity of the system. This model of two stairs can be extended to more than a single size (or a single type of) aggregates and thus, in general, several conductivity stairs may result.

This model is strongly supported by recent computer simulations of a somewhat more complicated model by Hu et al. [66]. These authors simulated samples of a few compact CNT aggregates and found that the increase of x yielded steps in $\sigma(x)$. Hence, conductivity stairs as obtained experimentally [15,16] can form when the aggregates configurations or the dominant conductance in them vary with the increase of x. We also note that, unlike in the simple tunneling staircase model of Section 2, the "jumps" of the global conductivity between stairs, in the presently discussed dispersion models, do not have to be large since the magnitudes of the "jumps" are determined primarily by the aggregates configurations that form in the CNT-polymer composites during the melt molding. In fact, the stairs can be "smeared out" if the dominating aggregate or the dominating configuration vary monotonically and continuously with increasing x. However, as in the tunneling staircase model we suggest that the apparent non universal behavior observed in many of these systems is due to the analysis of the corresponding "envelope" of the measured $\sigma(x)$ rather than with a continuous diverging distribution of the conductors, as we have seen here for the tunneling-percolation scenario in the carbon black-polymer composites.

Finally, while we discussed in detail the tunneling percolation staircase and mentioned briefly its possible generalization to the dispersion percolation staircase, other types of percolation staircases or a combination of them seem to be possible. In fact a system that does not fit the models of tunneling or aggregation is that of ion implanted silicon where the conductivity was measured as a function of the carrier concentration [68]. In that system two stairs were observed and the percolation behavior of $t = 2$ was deduced for the second, lower conductivity stair. This further suggests the generality of our model as well as its ability to explain many behaviors in terms of the dominant conductivity network in percolation systems.

In conclusion, in this work we have considered a lattice model in which the intersite conduction is by tunneling. We have used this model to analyze the electrical behavior of a granular metal and a carbon black polymer composite and we have suggested a percolation dispersion model for composites in which the conducting particles (e.g. CNT, metal nano-wire or graphene) are anisotropic. In our study we have shown that a staircase in the global conductivity dependence on the conducting phase content, $\sigma(x)$, in composite materials, is a general phenomenon. Two cases were described in some detail. The case where there is a peaked distribution of interparticle tunneling distances and a case where the configurations of particles changes with x. We made then a connection between the percolation threshold and the critical exponent of the conductivity which has not been considered previously in the tunneling percolation problem. This enabled us to draw three important classification criteria for the understanding of the $\sigma(x)$ behaviors in composites. First, if a single universal behavior of the conductivity is observed the composite either consists of a single dominant local conductance or the contribution of the possibly present smaller conductances is beyond the computing or experimental resolution. Second, if stairs are observed in the global conductivity they are due to the presence of a hierarchy in the values of the conducting elements that constitute the percolation network. Third, if only a single stair with a non universal behavior is observed there is a continuous monotonic distribution of decreasing conductances in the system. Finally, and most importantly, as $x \to 0$, the dilute nature of the composite can be well described by both, a non universal percolation behavior, with an $x = 0$ threshold, and a hopping like model.

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Appendix: The network of minimal resistance

The physical picture that we utilize for describing the evolution of the overall electrical conductivity in a lattice, with the increase of the bond percolation probability p^b, as well as for the justification of the application of equations (3) and (4), is that of the well-known links nodes blobs (LNB) [1,3,23] model. Briefly, the percolation backbone is the electrical conducting part of the infinite-size percolation cluster [1,2]. The bonds (or resistors) that make the backbone are the bonds through which there is an electric current if a bias is applied to the system. This backbone can be envisioned as consisting of a network (say, a square or a cubic lattice) of parallel "links" that are made of "singly connected bonds" (SCBs) and blobs, such that the "links" intersect at "nodes" that define the conducting net [1,23]. The SCBs are bond-resistors that are not by-passed by other resistors that belong to the same link, and the blobs are bunches of resistors such that some resistors or groups of them are connected in parallel with other resistors or groups of them. The length of the links are easily envisioned to be of the order of $\lambda \propto (p^b - p^b_c)^{-\upsilon}$, the average size of the finite clusters of connected bonds, since the "holes" in the infinite backbone net can encapsulate only finite clusters, and the diameter of these finite clusters is distributed around λ [1]. Here, p^b is the bond occupation probability, p^b_c is its value at the percolation threshold and υ is the correlation length exponent.

Turning to the expected resistance of the cubic LNB network, let us assume that the resistance of a link is R_λ. If a cubic sample of length L is considered we have, on the average, L/λ links that connect one face of the sample with the opposite one. Correspondingly, in a D dimensional system there will be $(L/\lambda)^{D-1}$ parallel links along the edge of the cube. Since the link's resistance is R_λ the resistance of the whole network of the R_λ's will be given by

$$R_L = R_\lambda(\lambda/L)^{D-2}. \tag{A.1}$$

To find R_L we need then "only" to estimate the value of R_λ. Assuming that all the occupied bonds, i.e., all the resistors in the system, have the same value r_o, we can estimate the value of R_λ in the LNB model as follows. We saw that the link consists of SCBs and blobs. The number of singly connected bonds L_1 in a link of length λ can be easily shown [3,23] to be given by $L_1 \propto (p^b - p^b_c)^{-1}$. Now, if we neglect the resistance of the blobs (that being made of parallel resistors have lower resistance than the same length chain that is made of singly connected resistors) the resistance of the link can be estimated by its lower bound which is $R_\lambda = r_o L_1 \propto (p^b - p^b_c)^{-1}$. However, the blobs further contribute to the resistance and thus one expects that $R_\lambda > r_o L_1$. One assumes then that $R_\lambda \propto (p^b - p^b_c)^{-\zeta}$ where ζ is an exponent that depends only on the dimensionality of the system [3,69]. While calculating the values of ζ is not straight forward, the fact that its value is larger, but not much larger, than unity is to be expected since, when $p_b \to p^b_c$ there is not only a "dilution" of the network of links, but also a "dilution" in the local structure of the blobs, and the links will approach the limit of a chain of SCBs. In other words as $p^b \to p^b_c$ the resistance will be affected by both, the "dilution" of the network due to the increase of ξ (as in Eq. (A.1)) and the "dilution" of the blobs. The latter effect means a "stronger" divergence of the sample resistance as $p^b \to p^b_c$ and it will thus be manifested by $\zeta \geqslant 1$. Another manifestation of the same picture is the less likelihood for "closed loops" in the structure with the increase of D, suggesting that the value of ζ decreases to unity for $D = 6$, the "critical dimension" of percolation [1,2]. Indeed $\zeta \approx 1.3$ for $D = 2$ and $\zeta \approx 1.1$ for $D = 3$ [3,69]. These simple principles were confirmed by simple hierarchy models [1] and simulations [3,69,70]. All the above results can be summarized then by presenting the critical behavior of the global resistance (or conductance) in a percolation system by:

$$R_L(= 1/G_L) \propto (p - p_c)^{-t}, \tag{A.2}$$

where t is the known critical exponent of the conductivity that is given, in view of the above, by:

$$t = (D - 2)\nu + \zeta. \tag{A.3}$$

These t values, that were derived by analytical approximations or Monte Carlo simulations for lattices and are known as the universal values of the conductivity exponent are, $t_{\mathrm{un}} = \zeta \approx 1.3$ for $D = 2$ and $t_{\mathrm{un}} = \upsilon + \zeta \approx 2$ for $D = 3$.

Turning to the problem of interest here, i.e., to the more general scenario where there are resistors of various values in the system, let us consider the case of $p^b > p^b_c$. For such a p^b there are many possible occupied bond configurations through the network but there is one that has the lowest resistance, while the other conducting configurations can be considered to be connected in parallel to it. Since the occupation of the bonds is random and there is no correlation between the value of the resistor and its location in the network any group of p^b_c bonds out of the available p^b bonds will provide a percolating network. In particular, the group of the p^b_c resistors of lowest values in the system of p^b occupied bonds provides a configuration of a minimal resistance network (MRN) that gives an upper limit to the resistance of the system [4]. Following the above random occupation of bonds we note that in every subgroup of bond-resistors, such as the percolation cluster, the links or the SCBs have the same distribution of the resistors values as the MRN and thus the same average resistance. This enables us to consider an "average" resistance value and replace the above r_o value, of the case where all the resistors have the same value, with the corresponding average of the participating resistors [3–5].

With the LNB and MRN pictures in mind we turn to discuss the analysis that was presented in Section 2 of this paper. For the basic physical justification for the application of equation (3) let us consider the very simple system of the first two types of neighbors (the two highest conductance stairs, where $z_1 = z_2 = 4$) in the two dimensional square lattice, by using the above bond percolation parameters. This use follows the fact that the bond percolation picture, as already mentioned in Section 2, is easier to follow in the present context, but all the semiquantitative results to be developed below apply equally well to the site percolation picture. The extension of the two nearest neighbor case (of $z_1 = z_2 = 4$) to the more general case will be discussed further below. We know that the number of bonds needed for onset of percolation in the square lattice is $B_c = p^b_{2c}(4 + 4)$, where p^b_{2c} is the critical bond occupation probability for the onset of percolation in this lattice when the participation of both the first and second nearest neighbors is allowed. Here B_c is a dimensional constant ($B_c = 2$ in 2D [2]) and we note that from the connectivity point of view the two types of bonds are indistinguishable. Hence, each set of randomly chosen fraction of p^b_{2c} bonds will yield a single backbone, but all the backbones that can form in such a way are topologically equivalent. Since randomly chosen, the number of type 1 (first nearest neighbor) and type 2 (second nearest neighbor) bonds must be the same in the above square lattice system. Let us examine now the evolvement of the structure of the occupied bonds networks in this system and then discuss the evolvement of its MRN with the increase of p^b.

Starting at $p^b = p^b_{2c}$, where we need all the available occupied bonds in order to get percolation, we must have a fraction of $p^b_{2c}/2$ occupied bonds of type 1 bonds and a fraction $p^b_{2c}/2$ of type 2 occupied bonds. As p^b increases such that $p^b > p^b_{2c}$ we have that any randomly chosen subgroup of p^b_{2c} bonds, out of the p^b bonds that are available, will exhibit percolation. This enables various outcomes at the onset of percolation with different proportions of the two types of bonds. For example, we can still have a subsystem of an equal fraction of the two types of bonds i.e., $p^b_{2c}/2$ of each type (as in the $p^b = p^b_{2c}$ case) but we can also have a subgroup of bonds that consists of as many bonds as possible (within the constraint that the overall occupation of bonds will not be less than p^b_{2c}) of say type 1 bonds and as few as possible type 2 bonds. In the latter case the maximum fraction of type 1 bonds can be as high as $p^b/2$ as this includes all the available type 1 bonds. In order to yield percolation we must have then at least $p^b_{2c} - p^b/2$ bonds of type 2. This consideration applies of course as long as $p^b < 2p^b_{2c}$. Larger p^b values will be considered below. Note that the same argument applies for any fraction of the two types of bonds as long as the sum of their fractions sum is p^b_{2c}. In other words, there are many networks that are topologically equivalent but the constraint of the conductance value distribution determines which one of them will determine (or dominate) the resultant (highest possible) conductivity of the system. The above results also confirm the expectation that the higher the p^b, the more significant can be the participation of the type 1 bonds (i.e., the larger the ratio, $p^b/2(p^b_{2c} - p^b/2)$ of type 1 to type 2 bonds in the so chosen network of maximal type 1 bond participation). We further note here that all other configurations (which have lower overall conductivities) are connected in parallel to the above highest conductivity one and as such the latter determines (the lower limit of) the overall conductivity of the system. An interesting case is when $p^b = p^b_{1c}$. Recalling that in the square lattice $p^b_{1c} = 1/2$ [2] and that (as we saw in Sect. 2) $p^b_{1c} = 2p^b_{2c} = 2(1/4)$ we get in that case that there is also a configuration of type 1-only occupied bonds ($p^b/2 = p^b_{2c}$) with no participation of type 2 ($p^b_{2c} - p^b/2 = 0$) bonds, which is exactly the meaning of p^b_{1c} in our staircase model.

Let us turn now to the implication of the above considerations on the resultant resistance of the system by associating a resistance R_1 (or a conductance $g_1 = 1/R_1$) to the type 1 bonds and R_2 (or a conductance $g_2 = 1/R_2$) to the type 2 bonds, and examine in particular the case of $R_2 \gg R_1$. For clarity, since resistors add in series, we will consider here local resistors rather than local conductances. It is obvious that the more the relative participation of the smaller R_1 resistors (instead of the larger R_2 resistors) in the MRN, the lower will be the resultant resistance of the corresponding network. Hence, from the electrical conductivity point of view, the lowest possible resistance of a network of p^b_{2c} (out of p^b) conductors (within the constraint of having p^b_{2c} bond resistors in the subsystem) will be obtained for the occupation ratio of $(p^b/2)/(p^b_{2c} - p^b/2)$ for the type 1 to the type 2 resistors. In other words, the MRN will consist of $p^b/2$, R_1 resistors and $p^b_{2c} - p^b/2$, R_2 resistors. It is seen then that at $p^b = p^b_{2c}$ the "average" resistance will be simply $(R_1 + R_2)/2$, while in the more general case of $p^b_{2c} \leqslant p^b \leqslant 2p^b_{2c}$) the "average" value of the resistors in the MRN, $\langle R \rangle_2$, will be:

$$\langle R \rangle_2 = \left(p^b R_1/2 + \left(p^b_{2c} - p^b/2\right) R_2\right)/p^b_{2c}. \tag{A.4}$$

This equation gives then the evolvement of $\langle R \rangle_2$ with increasing p^b. In the case of interest here, other networks that are possible for $p^b > p^b_{2c}$ will be connected in parallel to that of the MRN. However, all these networks will have smaller ratios than $(p^b/2)/(p^b_{2c} - p^b/2)$ and thus a higher average resistance than that of equation (A.4). Hence as long as the $\langle R \rangle_2 \propto R_2$ approximation (i.e., assuming that $R_2 \gg [(p^b/2)/(p^b_{2c} - p^b/2)]R_1$) is obeyed for the case of equation (A.4) it will be obeyed even more so for all the other possible parallel contributions of those networks. Correspondingly, the resistance of the whole system will be proportional to R_2.

Within the framework of our paper it is important to justify equation (3) i.e., to find out how appropriate is it to overlook the g_m's ($1 \leqslant m < n$) and thus to use the $\sigma_n \propto g_n$ relation in equation (3). To appreciate the role of the two types of resistors in the system let us examine the typical $R_2/R_1 = 10^3$ ratio that we have considered in Figures 2–4 and in the experimental results, and compare this R_2/R_1 ratio with the $(p^b/2)/(p^b_{2c} - p^b/2)$ ratio of the number of R_1 and R_2 resistors that we derived above. Recalling that in the square lattice $p^b_{2c} = 0.25$ we find that, for example, for p^b at $1.2p^b_{2c}$, $1.9p^b_{2c}$ and $1.99p^b_{2c}$, we get that the corresponding $(p^b/2)/(p^b_{2c} - p^b/2)$ ratios are 3/2, 19 and 199, respectively. In other words, even at p^b as high as $p^b = 0.497$ (noting that p^b_{1c} is just at 0.500) we have that the contribution of the R_2 resistors dominates $[R_2/R_1 = 10^3 > (p^b/2)/(p^b_{2c}-p^b/2) = 199]$ and thus, up to this p^b value which is (by our definition of the second stair range $p^b_{2c} \leqslant p^b < p^b_{1c}$) very close to the end of this stair, our assumption of the R_2 domination is well justified. For the above typical R_2/R_1 ratios we have then that for p^b, up to very close to p^b_{1c}, the $A_2(p)$ term embedded in equation (3) is well approximated by $p^b_{2c}/[(p^b_{2c} - p^b/2)]$. This approximation, within the second stair range has a much weaker dependence than $(p^b - p^b_{2c})^t$. This justifies then our use of a single σ_n value, throughout the second stair, in the application of equation (3) that was made in Figure 2. On the other hand, we saw that at p^b_{1c} we have already a percolating network that consists of R_1-only resistors. We note then, that, the width of the transition from a R_2 to a R_1 dominated global resistance will depend on the R_2/R_1 ratio and that for the ratios considered in this paper this transition is quite narrow. Since we do not use the results of this narrow "jump", (just below p^b_{1c}) in our analysis we have (as explained below) just extrapolated asymptotically the behavior of the second stair through this narrow range. Obviously, for $p^b > p^b_{1c}$ the global resistance of the lattice will be dominated by the network of R_1-only resistors as given by the behavior described by equation (3) for the conductivity throughout the first ($n = 1$) stair as plotted in Figure 2. Of course, in parallel there will be peripheral current carrying configurations that consist of less R_1 and more R_2 resistors, but these are even more dominated by the R_2 resistors than the MRN that we described above. The important point here is that the ratio between the global conductivity just below the transition to that just above the transition is of the order of $g_2/g_1 (= R_1/R_2)$. We have used this ratio also in our analysis of the experimental data. In Figure 2 this fact is presented by the extension of the curve of the second stair ($n = 2$) conductivity so that the ratios of the conductivities of the $n = 1$ curve and the $n = 2$ curve is g_1/g_2 for a common $p^b > p^b_{1c}$ value. Following that we discuss now the meaning of the $n = 2$ curve in the first stair regime. The same interpretation applies to any n in Figure 2.

The extension of the $n = 2$ curve into the first stair regime can be interpreted as a continuation of the peripheral MRN parallel configuration that consists of both R_1 and R_2 resistors that could have been obtained if the presence of the percolation cluster of the R_1-only resistors would have been eliminated. For dimensions higher than 2 (see below) the average resistance of the MRN in the $p^b > p^b_{1c}$ regime will be given by

$$\langle R\rangle_2 = \left\{ \left(p^b/2 - P_1\right) R_1 + \left[p^b_{2c} - \left(p^b/2 - P_1\right)\right] R_2 \right\} / p^b_{2c}, \tag{A.5}$$

where p^b_{2c} here is the percolation threshold of any such system that includes the first and second nearest neighbors and P_1 is the well-known [1,2] occupation probability of the percolation cluster of the R_1-only resistors. The conductivity contribution of the R_1 and R_2 MRN in the first stair will be given as in equation (3) by $(1/\langle R\rangle_2)[(p^b - P_1 - p^b_{2c})/(1 - P_1 - p^b_{2c})]^t$. In passing we note that P_1 cannot be larger than $p^b/2$ and that in our ($z_1 = z_2$) example (that is generalized below) p^b is larger than p^b_{2c} so that within the first stair regime $p^b - P_1 - p^b_{2c} = p^b/2 + (p^b/2 - P_1) - p^b_{2c} > 0$, even though the rate of the increase of P_1 with p^b is larger than that of p^b [2].

Considering equation (A.5) we note that for 2D systems the contribution of the MRN that contains the R_1 and R_2 resistors cannot be separated from the contribution of the R_1-only cluster since the MRN will be always shortened by this cluster. On the other hand, for higher dimensions, various types of independent percolation clusters can coexist and their independent contributions to the global conductivity can be estimated. Since the experimental data that we analyze in our study are 3D systems, the application of equation (A.5) (or its generalized form to be given below) i.e., the extension of the conductivity in the $n = 2$ stair to the regime of the $n = 1$ stair, is justified. We see then that, as illustrated in Figures 2 and 4, the relation $\Theta_2 \propto 1/R_2$ used in equation (3) is also justified there. The importance of this extrapolation of the second stair behavior to the first stair regime in our analysis is that, for high enough p^b, the ratio of the resultant conductivities, as given by the curves $n = 1$ and $n = 2$, is of the order of $R_2/R_1 = g_1/g_2$, which is a central prediction of our model. With a further increase of p^b there are fewer remaining R_1 resistors and more R_2 resistors in the peripheral network that consists of the R_1 and R_2 resistors in the first stair regime, since the backbone of the percolation cluster "consumes" the finite R_1 clusters [2]. Hence, the peripheral system of R_1 and R_2 resistors approaches, with the increase of p^b, the R_2-only cluster for which the overall conductivity is definitely proportional to $1/R_2$ and thus, for p^b appreciably above p^b_{1c}, $A_2(p) \approx 1$.

To summarize, our very important conclusion is that for the typical R_2/R_1 ratios used here the overall conductivity in the second stair is dominated by the R_2 resistance (except for a narrow range just below p^b_{1c}) and that above p^b_{1c} the extension of the corresponding conductance contribution of the second stair configuration is also dominated by R_2. Of course, the overall conductivity in the first stair is dominated by the higher local conductance of the system, R_1. Hence, equations (3) and (4), well describe (up to a weakly, p-dependent, factor) the dominating overall conductance. The use of these equations for carrying out our analysis of the overall conductivity (as given by Eq. (4)) is justified then. We can further conclude that the expected overall conductance that one would actually measure or compute, due to the two kinds of resistors, is the one given by the envelope of the conductivity as shown in Figures 2 and 4. The $n = 1$ curve represents then the contribution of the R_1-only network while the $n = 2$ curve represents the highest parallel conductance of the network that is made of R_1 and R_2 resistors.

The extension of the above case of two types of bonds to the more general case, where all the smaller than the largest local resistor in a stair, say, R_n, may participate in the conduction, is simple. As we saw in the above case of two types of resistors, all the bonds associated with the R_m's for which $m < n$ may contribute to the conduction in the system. For a given p^b the relative concentration of bonds associated with the mth neighbor (where there are n types of neighbors in the system i.e., the nth stair regime) is (see Sect. 2) z_m/Z_n. Following our assumption that $R_{(m-1)} \ll R_m \ll R_n$ for all $m < n$ we can follow the above two-resistor case and compare the cumulative partial participation of bonds associated with all the R_m's ($m < n$) with the participation of the bond-resistors of highest value, R_n. Since $\Sigma z_m/Z_n = Z_{(n-1)}/Z_n$, the fractional participation of all these bonds (as in Eq. (A.4)) is $p^b(Z_{(n-1)}/Z_n)$, while the fractional participation of the R_n resistors (that is necessary for the onset of percolation in the nth stair) in the MRN is: $p^b_{nc} - p^b(Z_{(n-1)}/Z_n)$. Correspondingly, the average resistance of the MRN will be given by:

$$\langle R\rangle_n \approx \left\{p^b\left(Z_{(n-1)}/Z_n\right)R_{(n-1)} + \left[p^b_{nc} - p^b\left(Z_{(n-1)}/Z_n\right)\right]R_n\right\}/p^b_{nc}. \qquad \text{(A.6)}$$

And the resultant conductivity will be given by $(1/\langle R\rangle_n)[(p^b - p^b_{nc})/(1 - p^b_{nc})]^t$.

As in the two resistors case that we considered above, the two interesting cases are that of $p^b = p^b_{nc}$ and of $p^b = p^b_{(n-1)c}$. Indeed, in the first case we get that the partial participation of all the low value resistors is $p^b_{nc}(Z_{(n-1)}/Z_n)$ and that of the high value (R_n) resistors is $p^b_{nc}(1 - Z_{(n-1)}/Z_n)$, while in the case of $p^b = p^b_{(n-1)c} = p^b_{nc}(Z_n/Z_{(n-1)})$ we get that the cumulative fractional occupation of all lower value resistors is p^b_{nc} and that none of the higher value resistors (R_n) is involved. We note again then that the relative contribution of the lower value resistors will increase with the increase of p^b, but the p^b value at which this contribution will become significant, depends essentially on the $R_n/R_{(n-1)}$ ratio. We further note that for the $R_n/R_{(n-1)}$ ratios considered in this work the contributions of the lower value resistors ($m < n$) in the nth stair will be of significance only very close to $p^b_{(n-1)c}$.

The extension of the nth stair to the range of the $n-1$ stair (i.e., to $p^b_{(n-1)c} < p^b < p^b_{(n-2)c}$) is similar to the one used above for the two stairs case. In other words, the contribution of the parallel networks that are dominated by the R_n's for $p^b > p^b_{(n-1)c}$ will be well approximated by $(1/\langle R\rangle_n)(p^b - \sum P_m - p^b_{nc})^t$ where $\langle R\rangle_n$ here is the average resistance value of the local conductors at all the stairs above the nth. In this expression, the P_m's ($1 \leqslant m \leqslant n-1$) are the probabilities to be on the percolation cluster of bonds (or resistors) of the mth type, the corresponding $\langle R\rangle_n$ is given then by:

$$\langle R\rangle_n = \left\{\Sigma\left(p^b z_m/Z_n - P_m\right)R_m + \left[p^b_{nc} - \Sigma\left(p^b z_m/Z_n - P_m\right)\right]R_n\right\}/p^b_{nc}. \qquad \text{(A.7)}$$

We note that here $P_m > 0$ only if p^b is above the pertinent percolation threshold, i.e., for $p^b > p^b_{mc}$. Obviously, if p^b belongs to the kth stair, $P_m = 0$ for all the m stairs for which $1 < m < k < n$. Hence, equation (A.7) is the generalized average resistance of the MRN for any $k < n$ stair. We have then that, as in the simple two stairs scenario, our results justify the general application of equation (3) and our corresponding interpretation of its extension beyond the percolation threshold of the next (higher conductivity) stair as is manifested by equation (4).

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