Unified Model for Pseudononuniversal Behavior of the Electrical Conductivity in Percolation Systems

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Many values of the observed conductivity percolation exponent t cannot be explained by the classical universal theory or by the existing nonuniversal theories. In particular, the $1.3 \leq t \leq 4.0$ clustering of t values, in both composite materials and porous media has not been accounted for. In this work we were concerned with a *pseudo*nonuniversal percolation behavior that, unlike the genuine nonuniversal behavior, explains the statistics of the experimentally observed percolation conductivity exponents in continuum systems.

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It is well established by now that the electrical conductivity σ in a two-phase disordered system is given by [1–9]

$$\sigma \propto (x - x_c)^t, \qquad (1)$$

where x is the fractional volume of the conducting phase in the system, x_c is the observed threshold for the onset of the conductivity and t is an exponent. By the universality of percolation as a phase transition, t is expected [1,4], and in many cases observed [3–11], to have the "universal" value μ that is well established in the theory of lattice percolation [12]. Hereafter, we use μ to denote either of its two dimensional $\mu(2\mathrm{D}) \approx 1.3$ or three dimensional $\mu(3\mathrm{D}) \approx 2.0$ values. In 1979 Kogut and Straley (K&S) [13] have shown (see SM1 in the Supplemental Material [14]) that t will be larger than μ if the values of the local conductances in the system g have a distribution that diverges as $g \to 0$. Following this "K&S effect" it was shown that the tunneling between conducting particles in composites [15] and the bottlenecks between the nonconducting elements in porous media [16] can bring about such a nonuniversal $t > \mu$ behavior. In particular, for the composites, in principle, the $t > \mu$ values have no bound [15], while for porous media [16], in 2 dimensions $t = \mu(2\mathrm{D})(=1.3)$ and in 3 dimensions t cannot exceed $\mu(3\mathrm{D}) + 1/2(=2 + 1/2)$.

Indeed, $t = \mu$ values and $t > \mu$ values were found in many continuum systems. However, examining in more detail the available t-value statistics in experimental works, on composites [6,7,9] and porous media [17,18], and in simulation studies [19–23], reveals that the many $t > \mu$ values have specific bounds that cannot be accounted for by the K&S [13], or deviation from isotropy [24], effects.

The principal characteristics of the t-value statistics are as follows (for more details see SM2 in Ref. [14]). For composite materials [6,7] it was concluded that "*t is predominantly in the range 1.3–4 peaking around 2*" [7]. While for those systems one can attribute $t > \mu$ values to the K&S effect [15], this effect cannot explain the sharp drop in the number of observed t values at the particular value of $t = 4.0$. For porous media [17,18] the t exponent "has been found to vary anywhere between 1.3 and 4 depending upon a variety of factors" [17]. The $t = 4.0$ bound in those media is even harder to explain because of the above $t \leq 2.5$, K&S value, limitation [16]. The most striking fact is that the above $1.3 \leq t \leq 4.0$ clustering is the same for such *a priori*, very different kinds of systems. Moreover, one finds (see SM3 in Ref. [14]) that there is a subclustering of t values in the $1.3 \leq t \leq 2.6$ and $2.0 \leq t \leq 4.0$ ranges in both types of systems [6,25–28], which again, has no explanation within the framework of the above effects. All these imply that there is another source for these t-clustering phenomena and this source is more abundant than the K&S [13] and other possible $t > \mu$ effects [23,24] (see more in SM2 and SM3 in the Supplemental Material [14]).

In this Letter we explain the above t clustering by adding to the previous universal [12,29,30] and nonuniversal [4,13–16] theories a "unified" theory of percolation conductivity that applies to both lattices and continuum. As our theory considers t values for an x range somewhat removed from x_c, we call the t values predicted by it the pseudononuniversal exponents. Our explanation is based on showing that these pseudoexponents can reach values as high as 2μ, provided that the x range studied is below the x value for the onset of the effective medium conditions. We start then by considering the transition from the percolation-scaling regime to the effective medium regime in both the bond percolation and the site percolation problems. Next, we derive the behavior expected when the bonds in the lattice constitute the local conductances but the available information is given in terms of the site occupation probability. This is followed by showing how corresponding considerations in continuum systems lead to the understanding of the above described t-values clustering.

Since the electrical conductivity in lattices is a bond related problem [12,29,30] its percolation dependence is given by [1–5]

$$\sigma \propto (1/R_b)(p_b - p_{\mathrm{bc}})^\mu, \qquad (2)$$

where R_b is the resistance associated with any occupied bond, p_b is the occupation probability of that bond, and p_{bc} is its critical (percolation threshold) value. One notes here that the meaningful measure for the deviation of p_b from p_{bc} is given by the normalized $p_{db} \equiv (p_b - p_{bc})/p_{bc}$ parameter [19,31].

While Eq. (2) [12] and the K&S effect [13,14,32] are associated with the asymptotic $p_b \to p_{bc}$ scaling-critical regime [12,30], the deviation of p_b from p_{bc} gives way to the regime of the well-known effective medium approximation (EMA) [8,17,33–35], where

$$\sigma \propto (1/R_b)(p_b - p_{bec})^v. \tag{3}$$

Here, $v \equiv 1$, is independent of the type of lattice and dimension and p_{bec} is the predicted EMA threshold (where $p_{bec} \geq p_{bc}$). For the present work the important point is that the transition between the two regimes is continuous and smooth [33–35] and that the details of the transition are system dependent (see SM4 in Ref. [14]).

In Eqs. (2) and (3) the resistors R_b were attached to lattice bonds. As illustrated in Fig. 1 one can, however, attach the resistors to sites with an occupation probability p_s [17,33,36]. An occupied site can be described by four resistors of an $R_s/2$ value each. If two adjacent sites are occupied there is a bond with a resistance R_b between them. Following the expected [4,12,29,30,37] scaling-critical behavior of the conductivity and its experimental [1,38] and computational [24,33,34] confirmations, one concludes that as the conductivity of the bonds-only system

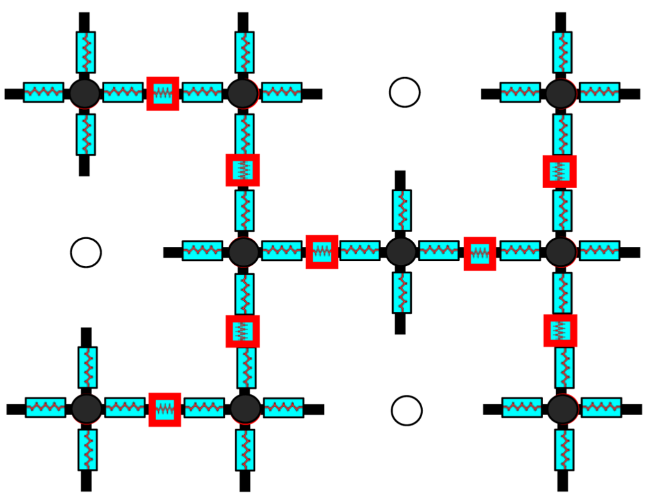

FIG. 1. An illustration of a segment of a square lattice where a site is either occupied (full circles) or unoccupied (empty circles). Each occupied site can be considered as made of four resistors each of which has a value of $R_s/2$ so that to cross it the resistance encountered is R_s. If two nearest neighbor sites are occupied there is a bond between them with a resistance R_b that is illustrated by a resistor in a red box. In principle, a "pseudononuniversal" behavior can arise when $R_b \gg R_s$ but the conductivity is monitored as a function of p_s (rather than as s function of p_b).

($R_s = 0$) is given by Eq. (2), the conductivity of the sites-only ($R_b = 0$) system will be given by [33,37]

$$\sigma \propto (1/R_s)(p_s - p_{sc})^{\mu}, \tag{4}$$

where p_{sc} is the sites percolation threshold.

Let us examine now the conductivity when the resistors are attached to the bonds, or $R_b \gg R_s$, but we want to (or we can only) express the conductivity in terms of p_s, as done first by Adler *et al.* [38]. Being concerned below with continuum systems we illustrate in SM5 [14] a possible corresponding porous medium. For lattices, however, we know that the relation $p_b = p_s^2$ applies to any lattice and any dimension [17,33,39]. This well-known relation follows intuitively [40] from the fact that for a lattice-spacing bond to be occupied, its two end sites must be occupied. A rigorous proof and a simulation confirmation of this relation will be provided elsewhere. This $p_b = p_s^2$ relation yields that, if we only know p_s and R_b [38], we will have to replace Eq. (2) by

$$\sigma \propto (1/R_b)(p_s^2 - p_{sc}^2)^{\mu} \propto (p_s - p_{sc})^{\mu}(p_s + p_{sc})^{\mu}. \tag{5}$$

As expected [37] [and as in Eq. (4)], for $p_s \to p_{sc}$ the behavior is controlled by $(p - p_c)^{\mu}$). Then, as p_s departs from p_{sc} but assuming that the effective medium conditions did not set in, a $\sigma \propto p_s^{2\mu}$ dependence will be approached. Following Eqs. (3) and (5), and as has been proven rigorously by Sahimi [17], one would also expect that, with a further increase of p_b,

$$\sigma \propto (p_s^2 - p_{sec}^2)^v = (p_s - p_{sec})(p_s + p_{sec}), \tag{6}$$

where, p_{sec} is the corresponding threshold of the EMA site problem.

The new results above are that while close to the percolation threshold, the percolation dependences [Eqs. (2) and (4)] apply, "far enough" from p_{sc} i.e., for $p_d \equiv (p_s - p_{sc})/p_{sc}) \gg 1$ (see the above definition for p_{db}) these behaviors may sequentially approach the $\sigma \propto p_s^{2\mu}$ and the $\sigma \propto p_s^{2v} \equiv p_s^2$ dependences [Eqs. (5) and (6), respectively]. We note here, however, that the largest possible p_s/p_{sc} ratio, in 2D and 3D lattices, can hardly fulfill the $p_s \gg p_{sc}$ requirement for the $\sigma \propto p_s^{2\mu}$ behavior [41]. On the other hand, the range of $p_{sc} \leq p_s \leq 1$ is usually enough for the onset of the effective medium conditions [17,19,33–36]. For the sake of argument, let us visualize then an imaginary lattice with a site percolation threshold $p_{scil} \ll 1$, so that there is a p_s regime where $p_{scil} \ll p_s$, but p_s is still small enough ($\ll$1) that the effective medium behavior has not dominated yet the conductivity. In such a lattice, in addition to the well-known expected [8,17,33] decrease of t with the increase of p_s from μ to $v \equiv 1$, some new $t(p_s)$ scenarios, that depend on the p_s regime, may be found. In principle then, considering the possible competition between the $\mu \to 2\mu$ [Eq. (5)] and the $\mu \rightarrow v(\equiv 1)$ [Eq. (6)] transitions, we conclude that the following possible sequences of t-exponent transitions, or a part of one of them, may be observed with the departure of p_s from p_{scil}. These are (a) $\mu \rightarrow v(\equiv 1)$, (b) $\mu \rightarrow 2\mu \rightarrow 2v \rightarrow v$, (c) $\mu \rightarrow 2\mu \rightarrow v \rightarrow 2v$, (d) $\mu \rightarrow 2v \rightarrow v$, and (e) $\mu \rightarrow v \rightarrow 2v$.

The main result to be emphasized here is the (in principle) possibility of the increase of the values of t beyond μ, in particular the $\mu \rightarrow 2\mu$ transition [as in scenarios (b) and (c)], in addition to the well-known [8,17,33] decrease of t from μ to $v = 1$ [scenario (a)]. In Fig. 2 we illustrate then, schematically, the corresponding expected $t(p_s)$ behaviors of scenarios (a) and (b). The important novelty here is that unlike the $t > \mu$ values in the K&S effect [13] the present results give t bounds that cannot exceed 2μ (i.e., $t = 2.6$ in two dimensions and $t = 4.0$ in three dimensions). This can provide then a clue, for the $t = 2.6$ and $t = 4$ boundaries which we were set to explain at the outset of this work (see also SM2, SM3, and SM6 in Ref. [14]).

Following the above predictions, we turn from the above imaginary behavior in lattices to the many continuum systems of composites and porous media where $x_c \ll 1$ [42,43] and in which the scaling behavior was found to extend to $x \gg x_c$ [44–48]. To consider those systems we use the well-known formal bridge of Scher and Zallen (S&Z) [1,49,50] between lattice and continuum percolation (SM5 in the Supplemental Material [14]). Indeed, in accordance with this and the expectation from the universality [4,37], one finds the fulfillment of Eq. (1) with $t = \mu$ and/or a $\sigma \propto (n - n_c)^{\mu}$ dependence in numerous continuum systems [3–7,10,11]. Here, n is the concentration of the conducting particles or pores, in composites or porous media, respectively, and n_c is the relevant threshold value. Correspondingly, in continuum systems the proximity to the threshold can be given by $x_d \equiv (x - x_c)/x_c$ [5] or $n_d \equiv (n - n_c)/n_c$ [19–22] (see SM5 in Ref. [14]) rather than by $p_d \equiv (p_s - p_{\mathrm{sc}})/p_{\mathrm{sc}}$ that is well defined in lattices [19,31]. For our purpose then, systems such as carbon nanotube (CNT) and graphene composites are of interest since for these, very small x_c ($\ll 1$), or very large n/n_c ($\gg 1$), values were found [22,44–48,51–55] and explained [42]. Hence, these small x_c values enable the study of systems at x values that are small enough ($x \ll 1$), so that the onset of the effective medium conditions is not appreciably manifested but, at the same time, they are large enough ($x/x_c \gg 1$) so that scenarios (b) and (c) are possible. Indeed, the universal $t = \mu(\mathrm{3D})$ value has been obtained in many systems with $x/x_c > 10$ values [44–46,48,51,56], indicating the relatively wide extent of the scaling-critical regime there. In particular, the increase of t, with the decrease of x_c, exactly within our predicted boundaries of $\mu(\mathrm{3D}) \leq t \leq 4.0$, as observed in Ref. [9], appears to provide a strong support to our scenario (b). Following that, we associate most of the many $\mu(\mathrm{3D}) < t \leq 2\mu(\mathrm{3D}) = 4.0$ values, obtained in the case of $x_d(\equiv x - x_c)/x_c) \gg 1$, [51,52,55,57] with this extension of the critical regime (see SM2 in Ref. [14]). A similar discussion for porous media will be given elsewhere.

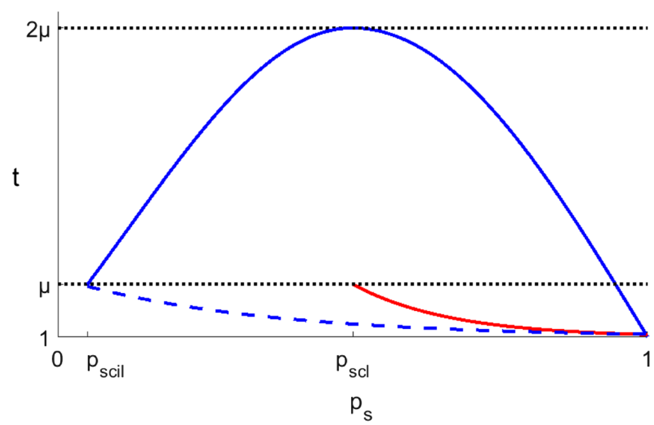

FIG. 2. The $t(p_s)$ dependence as concluded for real (red, $p_s \geq p_{\mathrm{scl}}$, curve) and imaginary (blue, $p_s \geq p_{\mathrm{scil}}$, curves) lattices. The p_{scl} is the threshold of existing lattices and p_{scil} is the threshold of an imaginary lattice. The lower (red and blue-dashed) $\mu \rightarrow 1$ curves illustrate the common $p_s \geq p_{\mathrm{scl}}$ [scenario (a)] behavior for both the bonds and the sites problems. The upper (blue, $p_s \geq p_{\mathrm{scil}}$) curve represents the [scenario (b)] behavior for an imaginary lattice of a very low percolation threshold when the resistors are attached to the lattice bonds but the conductivity is monitored as a function of p_s. It is important to note that the position of the peak and its specific shape depend on the details of the onset of the effective medium conditions. The μ and the 2μ (dotted) lines are separated, for the illustration, according to the $\mu(\mathrm{2D}) = 1.26$ value.

For a simple transparent example of the possible manifestation of the (b)–(e) scenarios in composites in which the $x_d \gg 1$, or the $n_d \gg 1$, condition is fulfilled, let us consider a 2D system of line segment "sticks" [10,11,20,21] such as those frequently used to represent CNT composites [19–22,42]. In the sticks systems, as illustrated in Fig. 3, we have a concentration of n_{st} sticks and there are, say, on the average, m intersections per stick. Doubling n_{st} also doubles m and thus the total concentration of the intersections (hereafter the junctions) n_j, will quadruple with n_{st}, i.e., $n_j \propto n_{\mathrm{st}}^2$. This simple intuitive conclusion has been confirmed in the simulations of Zezelj and Stankovic [21]. Considering the $p_b = p_s^2$ relation that we applied for lattices we can establish now the $p_s \leftrightarrow n_{\mathrm{st}}$ and $p_b \leftrightarrow n_j$ analogs of the two systems. Letting n_{stc} be the threshold for the onset of conductivity, yields that $n_{\mathrm{std}} = (n_{\mathrm{st}} - n_{\mathrm{stc}})/n_{\mathrm{stc}}$ is the analog of p_d and x_d. However, unlike lattices and latticelike systems (see Figs. 1, and SM1 and SM2 in the Supplemental Material [14]), for slender (permeablelike [42]) sticks there is, in practice, no limit to n_{st} [10,11,19,21]. Following that and the above analogy let us start, as in Fig. 1, by assigning a resistance of the order R_{st} to each stick and a resistance of order R_j to each intersection. For $R_{\mathrm{st}} \gg R_j$, we have, as expected intuitively, that the sticks are both the geometrical bonds and the

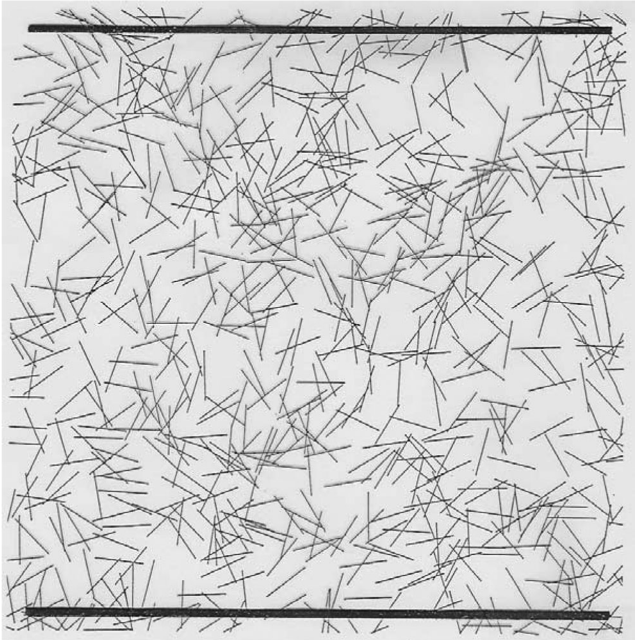

FIG. 3. A computer drawing of a two-dimensional system of sticks that can represent, e.g., a composite of nanotubes. The bus bars represent the electrodes that are used to monitor the sample's conductivity. In this system the resistance of each stick is R_{st} and the resistance of each intersection is R_j. Note that the condition $R_j > R_{st}$, is trivially suggesting that R_j is larger than the resistance of any segment of a stick. Of course, in real systems there can be a distribution of the lengths of the sticks and this illustration can describe a relevant average stick-length case.

electrical bonds of the system [11], while the intersections are the geometrical sites of the system. Hence, we expect [11,37], as in Eq. (2), the classical dependence of $\sigma \propto (1/R_{st})(n_{st} - n_{stc})^{\mu}$, which leads to our scenario (a). On the other hand if $R_j \gg R_{st}$ the dominant electrical bonds in the system are the intersection junctions [10] (as in the common intertube tunneling [7]) so that the conductivity is given, as in Eq. (4), by $\sigma \propto (1/R_j)(n_j - n_{jc})^{\mu}$, where n_{jc} is the corresponding conductivity threshold. Considering the above $n_j \propto n_{st}^2$ relation, the above analog and the fact that n_{st} is, in practice, the only available known (or "controllable") parameter, we have, that

$$\sigma \propto (1/R_j)(n_j - n_{jc})^{\mu} \propto (1/R_j)(n_{st}^2 - n_{stc}^2)^{\mu}. \quad (7)$$

In summary, while scenario (a) is expected for the $R_{st} \gg R_j$ case, for the combined case of $R_j \gg R_{st}$ [Eq. (7)] and the available $n_d \equiv (n_{st} - n_{stc})/n_{stc}) \gg 1$, we can expect one of the (b)–(e) scenarios. Indeed, simulations of the corresponding systems [19–22] (see SM6 in Ref. [14]) have shown the fulfillment of scenario (a) for the $R_{st} \gg R_j$ case and the increase of t with the increase of n_{st} beyond μ [19–22] (even up to $t = 2\mu$ [57]) for $R_j \gg R_{st}$. In fact, the continuous variation from scenario (a) to scenario (b) with the increase of the R_j/R_{st} ratio has been found in Refs. [20,21]. This suggests that the same conclusions apply to CNT and graphene based composites [42]. The important point to note here is that in all those simulations no g-value distribution [13–16], no change in system dimensions, and no anisotropy [23,24] were introduced and thus, the $t > \mu$ results cannot be attributed to either of those effects. In contrast, our present approach provides a general-wide framework since it accounts, by scenarios (b)–(e), for the majority of the observed $t > \mu$ values in the continuum, up to and including $2\mu = 2.6$ (in 2D) and $2\mu = 4.0$ (in 3D) values. That these are the majority t values, is proven by the fact that the statistics of all the above experimental [6,7] (see SM2 [14]) and computational data [19–22,57] (see SM6 [14]) exhibits the clustering of the values in the $\mu \le t \le 2\mu$ range, and that there is already available experimental evidence [9] for our prediction that the smaller the threshold the more likely the observation of scenarios (b) or (c). The fact that the same clustering applies to porous media [17] (as will be discussed elsewhere) provides then a firm support to our site-bond like models for the pseudononuniversal t values in many continuum systems.

In conclusion, combining the present results with the previously known universal and nonuniversal theories appears to provide now a wholesome framework for the understanding of the critical conductivity in numerous composite materials and porous media.

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