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Critical behaviour of non-linear susceptibility in random non-linear resistor networks

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Abstract. The critical behaviour of non-linear susceptibility of a two-component composite is studied in this paper. The first component of fraction p is non-linear and obeys a current–field (J–E) characteristic of the form $J = g_1 v + \chi_1 v^\beta$ while the second component of fraction q is linear with $J = g_2 v$. Near the percolation threshold p_c or q_c, we examine the conductor–insulator (C–I) limit ($g_2 = 0$) and superconductor–conductor (S–C) limit ($g_2 = +\infty$). For the C–I limit and $p > p_c$, the effective linear and non-linear response functions behave as $g_e \approx (p - p_c)^t$ and $\chi_e(\beta) \approx (p - p_c)^{t_2(\beta)}$, respectively. For the S–C limit and $q < q_c$, g_e and $\chi_e(\beta)$ are found to diverge as $g_e \approx (q_c - q)^{-s}$ and $\chi_e(\beta) \approx (q_c - q)^{s_2(\beta)}$. Within the effective-medium approximation, the exponents are found to be $s = t = 1$ and $s_2(\beta) = t_2(\beta) = (\beta+1)/2$, $p_c = 1/d$ and $q_c = (d-1)/d$. By using a connection between the non-linear response of the random non-linear composite problem and the resistance or conductance fluctuations of the corresponding random linear composite problem, the exponents $t_2(\beta)$ and $s_2(\beta)$ are found to be $t_2(\beta) = -\kappa((\beta+1)/2) + [(\beta+1)/2]t[(3-\beta)/2]d\nu$, $s_2(\beta) = \kappa'((\beta+1)/2) + [(\beta+1)/2]s + [(3+\beta)/2]d\nu$, respectively, where $t(s)$ is the conductivity exponent in a C–I(S–C) composite, d is the dimension of the composite and ν is the correlation-length exponent in d dimensions, $\kappa((\beta+1)/2)$ and $\kappa'((\beta+1)/2)$ are given by $\Psi_R((\beta+1)/2) + [(\beta+1)/2](d\nu - \zeta_R) = \kappa((\beta+1)/2) + d\nu$, $\Psi_G((\beta+1)/2) + [(\beta+1)/2](d\nu - \zeta_G) = \kappa'((\beta+1)/2) + d\nu$, where $\Psi_R((\beta+1)/2)(\Psi_G((\beta+1)/2))$ characterizes the scaling of the $[(\beta+1)/2]$th cumulant of the global resistance (conductance) distribution due to local resistance (conductance) fluctuations in the corresponding linear C–I(S–C) composites, $\zeta_R = t - (d-2)\nu$ and $\zeta_G = s + (d-2)\nu$. We prove that $t_2(\beta)$ is a monotonically increasing function of β while $s_2(\beta)$ is a monotonically decreasing function of β, which have the following special values: $t_2(1^+) = t - d\nu < 0$ and $s_2(1^+) = \zeta_G + d\nu (d = 2)$; $t_2(3) = 2t - \kappa(2)$ and $s_2(3) = 2s + \kappa'(2)$; $t_2(+\infty) = +\infty$ and $s_2(+\infty) = 1 + d\nu (d = 2)$. The critical behaviour of the non-linear susceptibility in a C–I composite is very different from that of the non-linear susceptibility in a S–C composite and some unexpected results of the critical behaviour of non-linear susceptibility in a C–I network are reported in this paper.

1. Introduction

Composite materials have long been known to have electrical and optical properties very different from those of their constituents [1]. The differences are particularly marked near a percolation threshold, i.e. a point at which one of the two components of the composite first forms a closed connected path extending throughout the sample. For a composite comprised of two materials with conductivities g_1 and g_2, the effective conductivity g_e exhibits the power law (in the limit in which g_2 approaches zero) [2]

$$g_e \approx g_1(p - p_c)^t \qquad (p > p_c) \tag{1}$$

where p is the volume fraction of material 1 and p_c is the percolation threshold for material 1. Conversely, if $g_1 \ll g_2$, then, as q approaches q_c from below, it is believed that g_e diverges according to the law [3]

$$g_e \approx g_2(q_c - q)^{-s} \qquad (q < q_c) \tag{2}$$

where q is the volume fraction of material 2 and q_c is the percolation threshold for material 2. The exponents t and s are believed to depend on the dimensionality and possibly also on the composite microstructure [4], as in continuum percolation, where the distribution of resistances has power-law singularities. The dependence on 'composite microstructure' exists not only for s and t, but also for other exponents, as was pointed out by Tremblay *et al* [5]. For lattice models, $t \approx 1.3$ in two dimensions ($d = 2$) [6, 7] and $t \approx 1.958$ for $d = 3$ [7], while $s \approx 1.3$ for $d = 2$ [7] and $s \approx 0.76$ for $d = 3$ [7].

The problem is further complicated by the fact that, for realistic composites, the non-linearity may have an important role in the electrical transport phenomena [8–23]. A typical example consists of studying a non-linear composite medium in which an inclusion with non-linear current–field (I–V) characteristics

$$J = g_1 v + \chi_1 v|v|^{\beta-1} \qquad (\beta > 1) \tag{3}$$

where $\chi_1|v|^{\beta-1} \ll g_1$, is randomly embedded in a host with linear response

$$J = g_2 v \tag{4}$$

where g_1 and g_2 are linear conductivities and χ_1 is the non-linear susceptibility. The volume fractions of the two components are p and q, respectively. We have $p + q = 1$. For such a system, substantial theoretical progress in studies of the effective non-linear response has been made for the cubic non-linearity $\beta = 3$ case in the past few years. Stroud and Hui [10] studied the dilute limit in which a small concentration of non-linear material is embedded in a linear host. They demonstrated a relation between the random non-linear composite problem and the noise problem in the corresponding random linear composite. An effective-medium approximation (EMA) [11] for a non-linear composite is proposed for calculating the effective non-linear response of a random mixture at higher concentrations. A systematic perturbative approach [16, 17] has also been developed to evaluate the effective response in mixtures in which both the inclusions and the host materials may be non-linear. For the case of arbitrary non-linearity, it has been studied by Hui [13] within the framework of the Maxwell–Garnett formula in the dilute limit of the non-linear component. Such composites are of practical interest because they may have a large non-linear susceptibility in two different circumstances: near the percolation threshold and near a surface plasmon resonance. In this paper we concentrate on the first. Near the percolation threshold p_c, we examine the conductor–insulator (C–I) limit ($g_2 = 0$) and the superconductor–conductor (S–C) limit ($g_2 = +\infty$). For the C–I limit and $p > p_c$, the behaviour of the effective linear response g_e is given by (1) and the non-linear-response function $\chi_e(\beta)$ behaves as [20, 21]

$$\chi_e(\beta) \approx (p - p_c)^{t_2(\beta)}. \tag{5}$$

For the S–C limit and $q < q_c$, the behaviour of the effective linear response g_e is given by (2) and the non-linear-response function $\chi_e(\beta)$ behaves as

$$\chi_e(\beta) \approx (q_c - q)^{-s_2(\beta)}. \tag{6}$$

The critical exponents $t_2(\beta)$ and $s_2(\beta)$ are novel universal characteristics of non-linear electrical transport in self-similar or percolating networks. Indeed they are not related in any simple way to the geometrical exponents and the linear transport exponents t and s. Although a wide variety of approaches [23] has been applied to the determination of the exponents s and t in the past few years, few efforts have been made to determine the exponents $t_2(\beta)$ and $s_2(\beta)$ and to study systematically the effect of the strength of the non-linear exponent β on the non-linear transport exponents $t_2(\beta)$ and $s_2(\beta)$. For cubic non-linearity $\beta = 3$, the EMA predicts [21] that $t_2(3) = 2$ and $s_2(3) = 2$ in all dimensions. By relating the cubic non-linear composite problem to the noise problem [20] in linear composites, the exponents $t_2(3)$ and $s_2(3)$ are found to be $t_2(3) = 2t - \kappa$ and $s_2(3) = 2s + \kappa'$, where $\kappa(\kappa')$ measures the divergence of relative resistance (conductance) fluctuations. It is the purpose of the present investigation to extend the results [20, 21] which are only applicable to cubic non-linearity $\beta = 3$ to the case of arbitrary non-linearity. We deal with this problem by two methods. Firstly we employ the generalized EMA which will be discussed in section 2.1 to estimate the exponents $t_2(\beta)$ and $s_2(\beta)$, and we find that $t_2(\beta) = s_2(\beta) = (\beta + 1)/2$ in all spatial dimensions. Secondly by using a connection between the non-linear response of the random non-linear composite problem and the resistance or conductance fluctuations of the corresponding random linear composite problem, the exponents $t_2(\beta)$ and $s_2(\beta)$ are found to be

$$t_2(\beta) = -\kappa\left(\frac{\beta+1}{2}\right) + \frac{\beta+1}{2}t - \frac{3-\beta}{2}d\nu$$

$$s_2(\beta) = \kappa'\left(\frac{\beta+1}{2}\right) + \frac{\beta+1}{2}s + \frac{3-\beta}{2}d\nu$$

respectively, where $t(s)$ is the conductivity exponent in a C–I(S–C) composite, d is the dimension of the composite and ν is the correlation-length exponent in d dimensions. $\kappa((\beta+1)/2)$ and $\kappa'((\beta+1)/2)$ are given by $\psi_R((\beta+1)/2) + [(\beta+1)/2](d\nu - \zeta_R) = \kappa((\beta+1)/2) + d\nu$, $\psi_G((\beta+1)/2) + [(\beta+1)/2](d\nu - \zeta_G) = \kappa'((\beta+1)/2) + d\nu$, where $\psi_R((\beta+1)/2)(\psi_G((\beta+1)/2))$ characterizes the scaling of the $[(\beta+1)/2]$th cumulant of the global resistance (conductance) distribution due to local resistance (conductance) fluctuations in the corresponding linear C–I(S–C) composites, $\zeta_R = t - (d-2)\nu$ and $\zeta_G = s + (d-2)\nu$. We prove that $t_2(\beta)$ is a monotonically increasing function of β while $s_2(\beta)$ is a monotonically decreasing function of β, which have the following special values: $t_2(1^+) = t - d\nu < 0$ and $s_2(1^+) = \zeta_G + d\nu$ $(d = 2)$; $t_2(3) = 2t - \kappa(2)$ and $s_2(3) = 2s + \kappa'(2)$; $t_2(+\infty) = +\infty$ and $s_2(+\infty) = 1 + d\nu$ $(d = 2)$. The critical behaviour of the non-linear susceptibility in a C–I composite is very different from that of the non-linear susceptibility in a S–C composite; the non-linear susceptibility of S–C near the percolation threshold will always diverge while $\chi_e(\beta)$ of the C–I composite can diverge or vanish depending on non-linearity.

2. Estimates of exponents $t_2(\beta)$ and $s_2(\beta)$

Consider a d-dimensional hypercubic non-linear conductor network which consists of two types of bond. We shall consider only bond percolation because it is more convenient for modelling transport properties. The first type of conductor is assumed to be non-linear and obeys a current–voltage $(I–V)$ characteristic of the form given by equation (3). Throughout this work, the non-linearity is assumed to be weak, so that $\chi_1 v^{\beta-1}/g_1 \ll 1$. The second component is assumed to be linear with the $I–V$ response given by equation (4). The volume fractions of the first and the second components are p and q, respectively. We have $p + q = 1$. We are interested in calculating the effective response of the network, representing by a homogeneous networks of identical conductors, each of which has an $I–V$ characteristic of the form [13]

$$i = g_e v + \chi_e(\beta)v|v|^{\beta-1} + \cdots \tag{7}$$

where g_e and $\chi_e(\beta)$ are the effective linear and leading-order non-linear responses, respectively, and are given by [10, 22]

$$g_e = \frac{1}{L^d} \sum_\alpha g_\alpha v_\alpha^2 \tag{8}$$

$$\chi_e(\beta) = \frac{1}{L^d} \sum_\alpha \chi_\alpha v_\alpha^{\beta+1} \tag{9}$$

where g_α and χ_α are the linear conductivity and non-linear susceptibility of the αth conductor, v_α is the voltage difference across the αth conductor in the *linear* random problem (i.e. obtained by solving the same random network problem with all $\chi_\alpha = 0$), and L is the size of the network. The summation is performed over all conductors in the network. In this paper, higher-order non-linear responses are not taken into account.

We consider the C–I limit in which the first component is percolating ($p > p_c$) while the second component is insulating, i.e. $g_2 = 0$, and the S–C limit in which the second component is simultaneously non-percolating and superconducting, i.e. $q < q_c$ and $g_2 = +\infty$. The critical behaviour of $\chi_e(\beta)$ is given by (5) and (6) in a C–I and S–C component, respectively.

2.1. Effective-medium approximation

The EMA is an old approach to transport properties of inhomogeneous materials. The EMA has attracted renewed interest with the development of percolation theory. As mentioned in [5], while there is a variational principle justifying the EMA for the exponents t and s, there is no such principle for other exponents, but this does not mean that the EMA cannot be used in other situations. In fact, the EMA in the context of exponents other than t and s was used first by Rammal [24] and also by Rammal *et al* [25]. The EMA, originally proposed for the cubic non-linearity $\beta = 3$ [11], can be readily generalized to the case of arbitrary non-linearity. The basis of our method is an exact result given by several workers [11, 22] that the effective non-linear response $\chi_e(\beta)$ can be calculated, to first order in the non-linearity, by the $\beta + 1$ moments of the local field distribution in a linear composite that has the same linear conductivity and the same microgeometry [23], namely [5, 10, 11]

$$\chi_e(\beta) = \frac{p\chi_1 \langle E^{\beta+1}\rangle_{lin}}{E_0^{\beta+1}} \tag{10}$$

where $\langle \ldots \rangle_{lin}$ denotes a volume average over the volume of the non-linear component in the linear limit (i.e. when $\chi_1 = 0$), p is the volume fraction of the non-linear component and E_0 is the space-averaged field within the composite.

A useful approximation for an average in (10) is to make the approximate factorization [11]

$$\langle E^{\beta+1}\rangle_{lin} \approx \langle E^2\rangle_{lin}^{(\beta+1)/2}. \tag{11}$$

Equation (11) amounts to a denial of Jensen's inequality [26], which for $\beta > 1$ gives $\langle E^{\beta+1}\rangle_{lin} \geqslant \langle E^2\rangle_{lin}^{(\beta+1)/2}$, with equality only when the random variable E takes a single value with probability 1. The right-hand side therefore gives a rigorous lower boundary for $\chi_e(\beta)$, although not a directly computable bound since g_e is not known exactly. Equation (11) also corresponds in a sense to the assumption of 'gap scaling' which is precisely what does not occur for multifractal exponents [27–30].

Since $\langle E^2\rangle_{lin}$ is given exactly by [5, 12]

$$\frac{p\langle E^2\rangle}{E_0^2} = \frac{\partial g_e}{\partial g_1} \tag{12}$$

where g_e and g_1 are the effective linear conductivities of the composite and of the non-linear component, respectively. From (10)–(12), $\chi_e(\beta)$ can be written approximately as

$$\chi_e(\beta) = \frac{\chi_1}{p^{(\beta-1)/2}}\left(\frac{\partial g_e}{\partial g_1}\right)^{(\beta+1)/2}. \tag{13}$$

The EMA is completed by calculating g_e from some approximations. One possible approximation for g_e is of course the linear EMA, which in d dimensions is given by

$$\sum_i p_i \frac{g_i - g_e}{g_i + (d-1)g_e} = 0 \tag{14}$$

where g_i is the linear conductivity of the ith component and d is the dimensionality. The approximation will be accurate in geometries for which the electric field is nearly uniform within the non-linear component and less accurate when these fluctuations are large, as in a random mixture near the percolation threshold. Nevertheless, the EMA gives qualitatively correct critical behaviour, but it predicts incorrect exponents, a phenomenon common in treating second-order phase transitions by mean-field theory.

Let us consider the C–I limit first. Let $g_2 = 0$; the effective linear response g_e is given by [23]

$$g_e = \frac{g_1}{1-p_c}(p-p_c) \qquad p_c = \frac{1}{d}. \tag{15}$$

Using equations (13) and (15), we obtain

$$\chi_e(\beta) = \frac{\chi_1}{p^{(\beta-1)/2}}\frac{1}{(1-p_c)^{(\beta-1)/2}}(p-p_c)^{(\beta+1)/2}. \tag{16}$$

Thus $t = 1$ and $t_2(\beta) = (\beta+1)/2$ within the EMA. When $\beta = 3$ we recover the result of cubic non-linearity [20, 21]; $t_2(3) = 2$.

For the S–C limit, let $g_2 = +\infty$, the effective linear response g_e in this case is given by [23]

$$g_e = g_1 q_c (q_c - q)^{-1} \qquad q_c = \frac{d-1}{d} \tag{17}$$

and we find that

$$\chi_e(\beta) = \frac{\chi_1}{q^{(\beta-1)/2}} q^{(\beta-1)/2}(q_c - q)^{-(\beta+1)/2}. \tag{18}$$

Thus $s = 1$ and $s_2(\beta) = (\beta+1)/2$ within the EMA. Here again we recover the result of cubic non-linearity [20, 21]: $s_2(3) = 2$ when $\beta = 3$.

So a crude estimate $t_2(\beta) = s_2(\beta) = (\beta+1)/2$ is found for all spatial dimensions d in the EMA.

2.2. Relation to resistance (conductance) fluctuation

A formal relation between the problem of the non-linear effective response in a random composite with *cubic* non-linearity and the problem of relative fluctuations (noise) in a linear random composite has been demonstrated by Stoud and Hui [10]. Later Blumenfeld and Bergman [19] generalized the result of [10] and presented the relation between the problem of non-linear effective response in a random composite with *arbitrary* non-linearity and the problem of resistance fluctuations in a linear random composite; the result is [19]

$$\chi_e(\beta) \sim L^d \langle \delta g_e^{(\beta+1)/2} \rangle_c \tag{19}$$

where $\langle \delta g_e^{(\beta+1)/2} \rangle_c$ is the higher-order cumulant [24, 25].

The quantity $(\langle \delta g_e^{(\beta+1)/2} \rangle_c)/(g_e^{(\beta+1)/2})$ is expected to be [19, 24, 25]

$$\frac{\langle \delta g_e^{(\beta+1)/2} \rangle_c}{g_e^{(\beta+1)/2}} \sim L^{d[1-(\beta+1)/2]}(p-p_c)^{-\kappa((\beta+1)/2)}. \tag{20}$$

Equation (20) defines the exponent $\kappa((\beta+1)/2)$ which reduces to the noise exponent $\kappa(2)$ when $\beta = 3$. As $p \to p_c$, $\xi = (p-p_c)^{-\nu} \to +\infty$ and, for a finite size system, $\xi \approx L$. Using (19), (20), $\xi \approx L$ and $g_e \sim (p-p_c)^t$, we obtain

$$\chi_e(\beta) \sim (p-p_c)^{t_2(\beta)}$$

where $t_2(\beta)$ is given by

$$t_2(\beta) = -\kappa\left(\frac{\beta+1}{2}\right) + \frac{\beta+1}{2}t - \frac{3-\beta}{2}d\nu. \tag{21}$$

Equation (20) can also be expressed by [19]

$$\frac{\langle \delta g_e^{(\beta+1)/2} \rangle_c}{g_e^{(\beta+1)/2}} \sim (p-p_c)^{-\{\psi_R((\beta+1)/2)-[(\beta+1)/2]\zeta_R\}}. \tag{22}$$

where $\psi_R((\beta+1)/2)$ characterizes the scaling of the $[(\beta+1)/2]$th cumulant of the global resistance distribution due to local resistance fluctuations [31] in a C–I network. The exponent $\psi((\beta+1)/2)$ is introduced under a different name and independently by both [33] and [29, 30]. Some analytic properties of the exponent $\psi((\beta+1)/2)$ have been discussed in [32]. Some of these exponents for integers $(\beta+1)/2$ have physical interpretations. For example, $\psi(0)$ gives the fractal dimensionality of the backbone D_B, $\psi(1)$ is equal to the resistance exponent ζ_R (N–I) or conductance exponent ζ_G (S–N), $\psi(2)$ is equal to the noise exponent κ (N–I) or κ' (S–N) [26, 29, 30], and $\psi(\infty)$ can be identified as the fractal dimension of the singly connected bonds (N–I) or singly disconnected bonds (S–N) [34].

When p approaches p_c from above, the correlation length ξ diverges as $\xi \sim (p-p_c)^{-\nu}$, and $L \approx \xi$, where L is the size of the system. From equations (20) and (22) we obtain the relation between $\kappa((\beta+1)/2)$ and $\psi_R((\beta+1)/2)$ [31, 32]:

$$\psi_R\left(\frac{\beta+1}{2}\right) + \frac{\beta+1}{2}(d\nu - \zeta_R) = \kappa\left(\frac{\beta+1}{2}\right) + d\nu \tag{23}$$

where ζ_R is defined by $\langle R \rangle \sim (p-p_c)^{-\zeta_R}$; $\langle R \rangle$ is the average resistance of the network and $\zeta_R = t-(d-2)\nu$ [34].

A similar result will be obtained by using a connection between the non-linear response of the random non-linear composite problem and the conductance fluctuations of the corresponding random linear composite problem [7, 12, 33]

$$\chi_e(\beta) \sim (q_c - q)^{-s_2(\beta)}$$

where $s_2(\beta)$ is given

$$s_2(\beta) = \kappa'\left(\frac{\beta+1}{2}\right) + \frac{\beta+1}{2}s + \frac{3-\beta}{2}d\nu. \tag{24}$$

The exponent $\kappa'((\beta+1)/2)$ is related to $\psi_G((\beta+1)/2)$ by

$$\psi_G\left(\frac{\beta+1}{2}\right)+\frac{\beta+1}{2}(d\nu-\zeta_G)=\kappa'\left(\frac{\beta+1}{2}\right)+d\nu \tag{25}$$

where ζ_G is defined by $\langle G\rangle\sim(q_c-q)^{-\zeta_G}$, with $\langle G\rangle$ the average conductance of the network and $\zeta_G=s+(d-2)\nu$ [34]. The physical meaning of $\psi_G((\beta+1)/2)$ is that $\psi_G((\beta+1)/2)$ characterizes the scaling of the $(\beta+1)/2$ cumulant of the global conductance distribution due to local conductance fluctuation in a S–C composite. Note that $t_2(\beta)$ and $s_2(\beta)$ can also be expressed in terms of $\psi_R((\beta+1)/2)$ or $\psi_G((\beta+1)/2)$ by using (23) and (25). The exponent $\psi_G((\beta+1)/2)$ has been discussed in [33].

Previous studies [12, 20, 21] on cubic non-linearity, $\beta=3$, give $t_2(3)=2t-\kappa(2)$ and $s_2(3)=2s+\kappa'(2)$. One can easily check that (21) and (24) will reduce to $2t-\kappa(2)$ and $2s+\kappa'(2)$ when $\beta=3$, respectively; by definition [32], $\kappa(2)=\kappa$ and $\kappa'(2)=\kappa'$. The values of the exponents are $t_2(3)\geqslant 0$ and $s_2(3)>0$ for $d=2$, $d=3$ and $d=6$ if one uses previously derived bounds on κ and κ' and estimates on t and s [5, 7]. For a summary of experiments on noise in percolating mixtures, we refer the reader to [30]. This implies that the cubic non-linear susceptibility $\chi_e(3)$ will have the same critical behaviour as the effective linear conductivity, namely $\chi_e(3)$ will vanish (diverge) in a C–I(S–C) network near the percolation threshold $p_c(q_c)$.

The analytic and numerical results concerning the $\psi_R((\beta+1)/2)$ have already appeared. Two approximate functions [32] both of which agree with the series results for all $(\beta+1)/2>1$ and with existing numerical simulations have been constructed in a randomly diluted resistor network on a dimensional hypercubic lattice at the percolation threshold p_c; one of the two approximate functions is

$$\psi_R\left(\frac{\beta+1}{2}\right)=1+(\nu D_B-1)^{1-((\beta+1)/2)}(\zeta_R-1)^{(\beta+1)/2} \tag{26}$$

where D_B is the fractal dimensionality of the backbone. $\nu=\frac{4}{3}$, $\zeta_R=1.297$ and $D_B=1.62$ [34] for $d=2$, and $\nu=0.89$, $\zeta_R=1.07$ and $D_B=1.74$ [34] for $d=3$.

For $d=2$, by duality considerations, de Arcangelis *et al* [33] have proven that $\psi_R((\beta+1)/2)$ of a random resistor network and $\psi_G((\beta+1)/2)$ of a random superconducting network coincide. So the approximate functions for $\psi_R((\beta+1)/2)$ in a random resistor network in $d=2$ can also be used to calculate $\psi_G((\beta+1)/2)$ in a S–C composite. Above two dimensions, the duality relation no longer holds. As a result, there appears to be no simple correspondence between $\psi_R((\beta+1)/2)$ of the random resistor network and $\psi_G((\beta+1)/2)$ of the random superconducting network. So for $d=3$ the approximate functions for $\psi_R((\beta+1)/2)$ in a random resistor network are not equal to $\psi_G((\beta+1)/2)$ in a random superconducting network. Until now the analytic form of $\psi_G((\beta+1)/2)$ above two dimensions in a random superconducting network has not been thoroughly investigated [26], despite the fact that some numerical values have been calculated [35–37]. So in this paper we present only our results of $s_2(\beta)$ for $d=2$.

Now we are in a position to discuss some properties which characterize the exponents $t_2(\beta)$ and $s_2(\beta)$. The properties of the exponents $t_2(\beta)$ and $s_2(\beta)$ will become very important when one wants to know how the magnitude of the non-linear susceptibility $\chi_e(\beta)$ depends upon the non-linear exponent β, as we now proceed to demonstrate. Let us first discuss the properties of the exponent $s_2(\beta)$ in $d=2$. We shall prove that

$$s_2(\beta)>0 \tag{27}$$

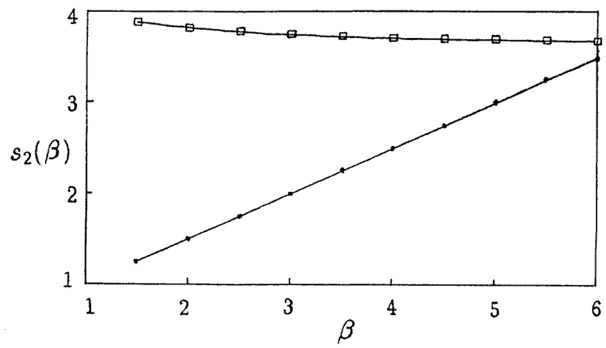

Figure 1. The exponent $s_2(\beta)$ as a function of β for a two-dimensional S–C network: ●, results given by the EMA; □, results calculated from equation (24). The values of $\psi((\beta+1)/2)$ used to construct the plot are from [32].

and

$$\frac{\mathrm{d}s_2(\beta)}{\mathrm{d}\beta} < 0 \tag{28}$$

for $\beta > 1$. The proof is very simple. By using equation (25), $s_2(\beta)$ can be written in terms of $\psi_G((\beta+1)/2)$, namely

$$s_2(\beta) = \psi_G\left(\frac{\beta+1}{2}\right) + d\nu + \frac{\beta+1}{2}(s - \zeta_G).$$

Since, for $d = 2$, $s = \zeta_G$ [34], $\psi(+\infty) = 1$ and $\psi(1) = \zeta_G$ [32], from the above equation, we obtain

$$\lim_{\beta\to+\infty}[s_2(\beta)] = 1 + 2\nu > 0 \tag{29}$$

$$\lim_{\beta\to1^+}[s_2(\beta)] = \zeta_G + 2\nu > 0 \tag{30}$$

and

$$\frac{\mathrm{d}s_2(\beta)}{\mathrm{d}\beta} = \frac{\mathrm{d}\psi_G((\beta+1)/2)}{\mathrm{d}((\beta+1)/2)} < 0. \tag{31}$$

In deriving equation (31), we have used $\mathrm{d}\psi_G((\beta+1)/2)/\mathrm{d}((\beta+1)/2) < 0$ [32]; this inequality is a consequence of the inequality $x_n > x_{n+1}$ that appears in the original work of Rammal *et al* [25]. From (27) we obtain $\chi_e(\beta) \to +\infty$ as $q \to q_c$ from below. So, as $q \to q_c$, not only g_e but also $\chi_e(\beta)$ diverge. From (28) we predict that $s_2(\beta)$ is a monotonically decreasing function of β. This implies that, as β increases, the non-linear susceptibility $\chi_e(\beta)$ will diverge more slowly.

Next we discuss the properties of the exponent $t_2(\beta)$. We shall prove that there exists a critical value β_c at which $t_2(\beta_c) = 0$; $t_2(\beta) < 0(1 < \beta < \beta_c)$ and $t_2(\beta) > 0(\beta > \beta_c)$.

By using equation (23), $t_2(\beta)$ can be written in terms of $\psi_R((\beta+1)/2)$:

$$t_2(\beta) = -\psi_R\left(\frac{\beta+1}{2}\right) - d\nu + \frac{\beta+1}{2}(\zeta_R + t). \tag{32}$$

Since $\psi_R(1) = \zeta_R$ [32], we obtain

$$\lim_{\beta\to1^+}[t_2(\beta)] = t - d\nu \leqslant 0. \tag{33}$$

By using $\psi_R(+\infty) = 1$ [32], we obtain

$$\lim_{\beta\to+\infty} [t_2(\beta)] \to +\infty. \tag{34}$$

Using $\mathrm{d}\psi_R((\beta+1)/2)/\mathrm{d}((\beta+1)/2) < 0$, we obtain

$$\frac{\mathrm{d}t_2(\beta)}{\mathrm{d}\beta} = -\frac{1}{2}\frac{\mathrm{d}\psi_R((\beta+1)/2)}{\mathrm{d}((\beta+1)/2)} + \tfrac{1}{2}(\zeta_R + t) > 0. \tag{35}$$

The inequality in (33) is obtained by direct calculation of $t_2(\beta)$ using the following: $t = 1.30$ and $\nu = \frac{4}{3}$ for $d = 2$; $t = 1.958$ and $\nu = 0.89$ for $d = 3$; $t = 6$ and $\nu = \frac{1}{2}$ for $d = 6$ [27]. Since the critical behaviour of $\chi_e(\beta)$ is given by (5) near p_c, the result of (35) implies that the non-linear susceptibility will vanish more quickly as β increases. From (33)–(35), we predict that there exists a critical value β_c at which $t_2(\beta_c) = 0$. While the exponent $s_2(\beta)$ is always positive, however, the exponent $t_2(\beta)$ may take positive values ($\beta > \beta_c$), a zero value ($\beta = \beta_c$) and negative values ($1 < \beta < \beta_c$). As a consequence, the magnitude of the non-linear susceptibility $\chi_e(\beta)$ in a C–I network may change dramatically as β increases. As $p \to p_c$ from above, we have

$$\chi_e(\beta) \to 0 \qquad \beta > \beta_c \qquad t_2(\beta) > 0 \tag{36}$$

$$\chi_e(\beta) \to \text{constant} \qquad \beta = \beta_c \qquad t_2(\beta) = 0 \tag{37}$$

$$\chi_e(\beta) \to +\infty \qquad 1 < \beta < \beta_c \qquad t_2(\beta) < 0. \tag{38}$$

We have calculated the critical value of β_c by solving (21), and we obtain $\beta_c = 1.947 (d = 2)$ and $\beta_c = 1.372 (d = 3)$.

The non-linear susceptibility $\chi_e(\beta)$ in a C–I network in the region $1 < \beta < \beta_c$ will show some anomalous behaviour. In this region, as $p \to p_c$, the linear response g_e will approach zero; however, $\chi_e(\beta)$ will diverge. Such a result is somewhat unexpected, because in a C–I composite both the linear response g_e and the non-linear response $\chi_e(\beta)$ are expected to approach zero as $p \to p_c$. However, we found that the non-linear susceptibility $\chi_e(\beta)$ will diverge, which usually occurs in a S–C composite, and this cannot be explained from the viewpoint of percolation theory. Unfortunately we cannot explain this anomalous phenomenon. If one assumes that this arises because, near the percolation p_c, not only the restricted geometry of the path through which current flows, but also the strength of the non-linear exponent β have effects on $\chi_e(\beta)$, the final behaviour has contributions from both; however, one cannot explain why the exponent $s_2(\beta)$ does not change sign as β increases.

We also calculate the exponents $t_2(\beta)$ and $s_2(\beta)$ numerically. The results are given in figures 1 and 2. As mentioned above, the EMA gives incorrect exponents, especially for $s_2(\beta)$; the EMA predicts that $s_2(\beta)$ is an increasing linear function of β, while (24) predicts that $s_2(\beta)$ is a decreasing function of β. The EMA also predicts that $s_2(+\infty) = +\infty$, while (24) gives $s_2(+\infty) = 1 + d\nu$. For $t_2(\beta)$, in the region when β is very small, the EMA also gives incorrect exponents; for example, in this region, the EMA predicts that $t_2(\beta) > 0$ while (21) gives $t_2(\beta) < 0$. However, for $\beta \approx 6$, the EMA gives a somewhat correct exponent for $s_2(\beta)$ and, for $2 < \beta < 3$, the EMA also gives the correct exponent for $t_2(\beta)$. So the EMA can still be used as a first step towards estimates of critical exponents.

3. Conclusion

In this work the critical exponents $t_2(\beta)$ and $s_2(\beta)$ governing the behaviour of non-linear susceptibility $\chi_e(\beta)$ are obtained by two methods. Firstly, we employ the generalized EMA

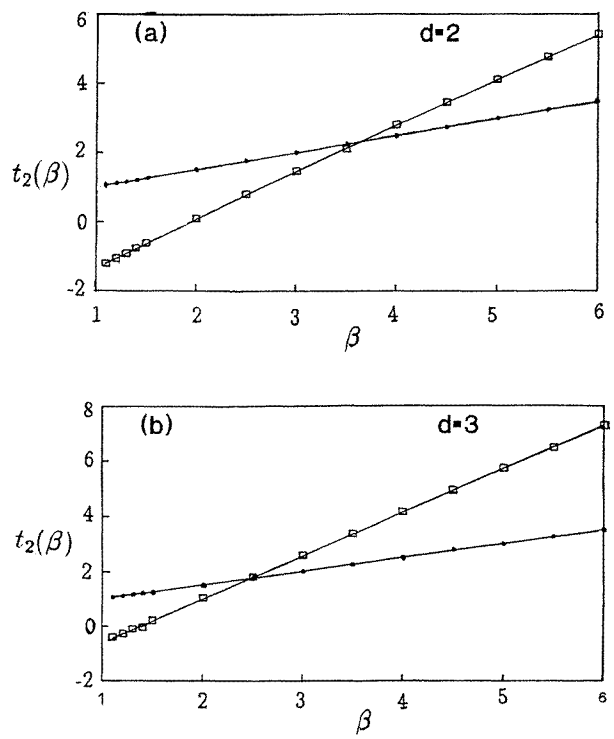

Figure 2. The exponent $t_2(\beta)$ as a function of β for a C–I network: (a) $d = 2$; (b) $d = 3$. The symbols are the same as in figure 1 except that the open squares are the data calculated from equation (21).

which is applicable to arbitrary non-linearity to estimate the exponents $t_2(\beta)$ and $s_2(\beta)$, and we find that $t_2(\beta) = s_2(\beta) = (\beta + 1)/2$ in all spatial dimensions. Secondly, by using a connection between the non-linear response of the random non-linear composite problem and the resistance or conductance fluctuations of the corresponding random linear composite problem, the exponents $t_2(\beta)$ and $s_2(\beta)$ are found to be $t_2(\beta) = -\kappa((\beta+1)/2) + [(\beta + 1)/2]t - [(3 - \beta)/2]d\nu$ and $s_2(\beta) = \kappa'((\beta + 1)/2) + [(\beta + 1)/2]s + [(3 - \beta)/2]d\nu$, respectively. We prove that $t_2(\beta)$ is a monotonically increasing function of β while $s_2(\beta)$ is a monotonically decreasing function of β, and the non-linear susceptibility $\chi_e(\beta)$ of the S–C network will diverge as q approaches q_c from below. The non-linear susceptibility $\chi_e(\beta)$ of a C–I network may show a complicated behaviour. There exists a critical value β_c at which $t_2(\beta_c) = 0$. When $\beta > \beta_c$, $t_2(\beta) > 0$, and $\chi_e(\beta)$ will vanish as p approaches p_c from above. However, when $1 < \beta < \beta_c$, $t_2(\beta) < 0$, and $\chi_e(\beta)$ will diverge as p approaches p_c. We cannot explain this anomalous behaviour and we hope that further investigation may be carried out along these lines.

Our predictions presented above should be compared with the results of numerical simulations and experiments and we hope that our paper may stimulate numerical simulations and experiment investigation on this problem. It would be worthwhile to study the exponent $s_2(\beta)$ above two dimensions where we can no longer exploit duality arguments to obtain a relation with the random resistor network.

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