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Critical properties of nonlinear susceptibilities for weakly nonlinear composites

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Abstract. The critical exponents of the effective nonlinear susceptibilities for two-component weakly nonlinear composites near the percolation threshold are studied. We consider a d-dimensional, two-component weakly nonlinear composite in which the volume fraction f of the first component obeys a nonlinear current-density–field ($\boldsymbol{J}$–$\boldsymbol{E}$) relation of the form $\boldsymbol{J} = \sigma_1\boldsymbol{E} + \chi_1|\boldsymbol{E}|^\beta\boldsymbol{E}$ and the volume fraction g of the second component exhibits the linear response $\boldsymbol{J} = \sigma_2\boldsymbol{E}$. Two important limits are examined: (1) the normal-conductor–insulator (N/I) mixture with $\sigma_2 = 0$; (2) the superconductor–normal-conductor (S/N) mixture in which the superconducting component has infinite conductivity, $\sigma_2 = \infty$. As the percolation threshold f_c or g_c is approached, the effective response χ_e is found to behave as $\chi_e \sim (f - f_c)^{u(\beta)}$ in the N/I limit and $\chi_e \sim (g_c - g)^{-v(\beta)}$ in the S/N limit. On the basis of the relation between the effective nonlinear susceptibility in random nonlinear composites and the resistance (or conductance) fluctuations in the corresponding linear composites, we give explicit expressions for $u(\beta)$ and $v(\beta)$ for arbitrary nonlinearity β and dimensionality d. Meanwhile we calculate numerically the critical exponents $u(\beta)$ and $v(\beta)$ as functions of β for different dimensions; anomalous critical behaviour of the effective nonlinear susceptibility for a two-dimensional N/I system is found.

1. Introduction

The physics of weakly nonlinear composite media has attracted much interest in recent years [1–4]. A typical system is that of a composite in which one component has a nonlinear current-density–electric field ($\boldsymbol{J}$–$\boldsymbol{E}$) relation of the form $\boldsymbol{J} = \sigma_1\boldsymbol{E} + \chi|\boldsymbol{E}|^\beta\boldsymbol{E}$ ($\sigma_1 \gg \chi_1|\boldsymbol{E}|^\beta$) randomly mixed with another component with the linear response $\boldsymbol{J} = \sigma_2\boldsymbol{E}$. For such a system, the effective linear conductivity σ_e and the $(\beta+1)$th-order nonlinear susceptibility $\chi_e(\beta)$ have been calculated [5–7], and a higher-order nonlinear response has also been proposed [8, 9]. More recently, another problem worth noting has been identified: that of the critical behaviour of the effective nonlinear susceptibility near the percolation threshold. There are two important limits to be considered:

(i) the weakly nonlinear normal-conductor–insulator (N/I) mixture in which the insulating component has no finite conductivity ($\sigma_2 = 0$); and

(ii) the superconductor–weakly nonlinear normal-conductor (S/N) mixture in which the superconducting component has infinite conductivity ($\sigma_2 = \infty$).

For linear transport properties, it is known that the general theory of percolation [10] has been extremely useful in describing the properties of a random mixture in the vicinity of

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the percolation threshold. Above (or below) the percolation threshold, a critical exponent t (or s) has been applied to describe the vanishing (or divergence) of the linear conductivity σ_e in the N/I (or S/N) system, i.e.,

$$\sigma_e \sim (f - f_c)^t \qquad \text{in the N/I limit} \tag{1}$$

or

$$\sigma_e \sim (g_c - g)^{-s} \qquad \text{in the S/N limit} \tag{2}$$

where f_c and g_c are the percolation thresholds of the first and second components, respectively.

For nonlinear transport properties, following equations (1) and (2), it is natural to define other exponents in order to consider the geometric effects on the nonlinear transport properties in random composites; that is,

$$\chi_e(\beta) \sim (f - f_c)^{u(\beta)} \qquad \text{in the N/I limit} \tag{3}$$

or

$$\chi_e(\beta) \sim (g_c - g)^{-v(\beta)} \qquad \text{in the S/N limit} \tag{4}$$

where $u(\beta)$ and $v(\beta)$ are the critical exponents of the effective nonlinear susceptibility χ_e and are β-dependent in general. For the case where $\beta = 2$, these exponents have been studied in references [11, 12] on the basis of the effective-medium approximation (EMA), and in references [2, 12] on the basis of the relation to the noise exponents $k(2)$ (or $k'(2)$) which characterize the divergence of the relative resistance (or conductance) fluctuations in linear random composites. The qualitative results obtained using the EMA for arbitrary β have been reported [7, 13]. It is our aim here to determine the dependence of the exponents $u(\beta)$ and $v(\beta)$ on the nonlinear exponent β by relating the nonlinear response of the random composite problem to the resistance (or conductance) fluctuations of the corresponding problem. Our study emphasizes two points. Firstly, we analyse the relation between the size L of the composite system and the correlation length ξ. Secondly, it is necessary to give the critical exponents $v(\beta)$ for higher dimensions. We shall calculate $u(\beta)$ and $v(\beta)$ as functions of β for different dimensions. The effective nonlinear susceptibility χ_e for a two-dimensional N/I composite reveals anomalous critical behaviour.

2. Formalism

Let us consider a d-dimensional weakly nonlinear composite composed of two types of component. One, with the volume fraction f, obeys a weakly nonlinear current-density–electric field ($\boldsymbol{J}$–$\boldsymbol{E}$) response of the form

$$\boldsymbol{J} = \sigma_1 \boldsymbol{E} + \chi_1 |\boldsymbol{E}|^\beta \boldsymbol{E} \qquad (\beta > 0) \tag{5}$$

where σ_1, χ_1 and β are the linear conductivity, nonlinear susceptibility and nonlinear exponent respectively, and weak nonlinearity requires $\chi_1 |\boldsymbol{E}|^\beta \ll \sigma_1$. The other component, with the volume fraction g, has a linear characteristic relation of the form $\boldsymbol{J} = \sigma_2 \boldsymbol{E}$. We have $f + g = 1$. The effective response of the whole system can be defined in such a way that we use the following space-averaged current-density–space-averaged electric field ($\langle \boldsymbol{J} \rangle$–$\langle \boldsymbol{E} \rangle$) relation:

$$\langle \boldsymbol{J} \rangle = \sigma_e \langle \boldsymbol{E} \rangle + \chi_e \langle |\boldsymbol{E}|^\beta \rangle \langle \boldsymbol{E} \rangle \tag{6}$$

where σ_e and χ_e are the effective conductivity and nonlinear susceptibility, and are given by

$$\sigma_e E_0^2 = \frac{1}{V}\int_v \sigma_i |\boldsymbol{E}_i|_{lin}^2 \, \mathrm{d}V \tag{7}$$

$$\chi_e(\beta) E_0^{\beta+2} = \frac{1}{V}\int_v \chi_i |\boldsymbol{E}_i|_{lin}^{\beta+2} \, \mathrm{d}V \tag{8}$$

where σ_i and χ_i are the linear conductivity and nonlinear susceptibility of the ith component while E_0 and $|\boldsymbol{E}_i|_{lin}$ stand for the applied field and the local electric field in the ith component within linear composites (i.e., obtained from the same system but with $\chi_i = 0$), respectively.

With equations (7) and (8), the effective nonlinear response of σ_e and χ_e can be obtained. However, here we will derive the expressions for the critical behaviour $u(\beta)$ for a N/I composite and $v(\beta)$ for a S/N composite. In view of the similarity of the procedures followed in the discussion of these systems, the derivations will be presented in parallel.

The relation between the problem of weakly third-order nonlinearity ($\beta = 2$) and that of the resistance (or conductance) fluctuation has been formulated in reference [2]. It is found that

$$\chi_e(\beta = 2) \sim L^d \, \delta\sigma_e^2 \tag{9}$$

where $\delta\sigma_e^2$ is the mean square fluctuation of the effective linear conductivity σ_e. The above equation can be easily generalized to the effective nonlinear susceptibility $\chi_e(\beta)$ for arbitrary β [13, 14]:

$$\chi_e(\beta) \sim L^d \, \delta\sigma_e^{(\beta+2)/2} \tag{10}$$

where $\delta\sigma_e^{(\beta+2)/2}$ is defined as the higher-order cumulant. Consequently, the quantity $\delta\sigma_e^{(\beta+2)/2}/\sigma_e^{(\beta+2)/2}$, which characterizes the ratio of the higher-order cumulant to the mean $[(\beta+2)/2]$th order of the effective linear conductivity, is expected to be [14]

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} \sim L^{d(1-(\beta+2)/2)}(f - f_c)^{-\kappa((\beta+2)/2)} \qquad \text{with } f > f_c \text{ in the N/I limit} \tag{11}$$

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} \sim L^{d(1-(\beta+2)/2)}(g_c - g)^{-\kappa'((\beta+2)/2)} \qquad \text{with } g < g_c \text{ in the S/N limit} \tag{12}$$

where $\kappa((\beta+2)/2)$ and $\kappa'((\beta+2)/2)$ denote the divergence of the high-order relative fluctuation and can be reduced to the noise exponents $\kappa(2)$ and $\kappa'(2)$ for $\beta = 2$. Combining equations (11), (12) and (10), we have

$$\chi_e(\beta) \sim L^{(2-\beta)d/2}(f - f_c)^{[\{(\beta+2)/2\}t - \kappa((\beta+2)/2)]} \qquad \text{in the N/I limit} \tag{13}$$

and

$$\chi_e(\beta) \sim L^{(2-\beta)d/2}(g_c - g)^{[-\{(\beta+2)/2\}s - \kappa'((\beta+2)/2)]} \qquad \text{in the S/N limit.} \tag{14}$$

The above relations are correct for $L > \xi$, i.e., in the Euclidean regime; thus they hold in the thermodynamic limit ($L = \infty$) and near the percolation threshold f_c (or g_c). Therefore, the critical exponents $u(\beta)$ and $v(\beta)$, which describe the dependence on $f - f_c$ (or $g_c - g$) of the effective nonlinear susceptibility $\chi_e(\beta)$ in the N/I (or the S/N) limit, are given as follows:

$$u(\beta) = \frac{\beta+2}{2}t - \kappa\left(\frac{\beta+2}{2}\right) \tag{15}$$

and

$$v(\beta) = \frac{\beta+2}{2}s + \kappa'\left(\frac{\beta+2}{2}\right). \tag{16}$$

Similar expressions for $u(\beta)$ and $v(\beta)$ have been obtained in [15]; when $\beta = 2$, equations (15) and (16) will be reduced to the well known results $u(2) = 2t - \kappa(2)$ and $v(2) = 2s + \kappa'(2)$, respectively [12].

Here we must emphasize that we are analysing the relation between L and the correlation length ξ ($\xi \sim (f - f_c)^{-\nu}$ or $\xi \sim (g_c - g)^{-\nu}$ near the percolation threshold, and ν is the correlation length exponent). It is known that L and ξ are two different physical parameters. In the vicinity of f_c (or g_c), ξ is a function of $f - f_c$ (or $g_c - g$), while L is a fixed quantity for a given system (in the thermodynamic limit, L is taken as ∞) and is independent of $f - f_c$ (or $g_c - g$). Thus, these physical parameters depend not only on L but also on $f - f_c$ (or $g_c - g$); however, at the percolation threshold, ξ diverges and equations (13) and (14) are not approached; the whole system is always in the fractal or self-similar regime. In this case, these physical parameters will only depend on L and can be obtained by putting $f - f_c \sim \xi^{-1/\nu} = L^{-1/\nu}$ (or $g_c - g \sim \xi^{-1/\nu} = L^{-1/\nu}$), i.e., by replacing ξ with L (not L with ξ). In short, L cannot be replaced by ξ in any case, and L and ξ must not be confused [13].

In order to investigate the properties of the critical exponents $u(\beta)$ and $v(\beta)$, we must look for expressions for $\kappa((\beta+2)/2)$ and $\kappa'((\beta+2)/2)$.

For finite L, in the fractal regime, $L < \xi$; thus $\xi \approx L$, and equations (11) and (12) can be written as

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} \sim L^{[d(1-(\beta+2)/2)+\kappa((\beta+2)/2)/\nu]} \tag{17}$$

and

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} \sim L^{[d(1-(\beta+2)/2)+\kappa'((\beta+2)/2)/\nu]}. \tag{18}$$

On the other hand, the above two equations can also be expressed as [14]

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} = \frac{\langle \delta R^{(\beta+2)/2}\rangle_c}{R^{(\beta+2)/2}} \sim L^{[\psi_R((\beta+2)/2)-((2+\beta)/2)\zeta_R]/\nu} \tag{19}$$

and

$$\frac{\langle \delta\sigma_e^{(\beta+2)/2}\rangle_c}{\sigma_e^{(\beta+2)/2}} = \frac{\langle \delta G^{(\beta+2)/2}\rangle_c}{G^{(\beta+2)/2}} \sim L^{[\psi_G((\beta+2)/2)-((2+\beta)/2)\zeta_G]/\nu}. \tag{20}$$

In equations (19) and (20), $\psi_{R(G)}$ characterizes the scaling of the $[(\beta+2)/2]$th cumulant of the global resistance (or conductance) distribution because of the local resistance (or conductance) fluctuations, i.e., $\langle \delta R^{(\beta+2)/2}(G^{(\beta+2)/2})\rangle_c \sim L^{[\psi_{R(G)}((\beta+2)/2)]/\nu}$ [16], while the average macroscopic resistance R (or conductance G) behaves as $R \sim L^{2-d}/\sigma_e \sim (f-f_c)^{-t}L^{2-d} \sim L^{t/\nu+2-d} \sim L^{\zeta_R/\nu}$ (or $G \sim \sigma_e L^{d-2} \sim (g_c-g)^{-s}L^{d-2} \sim L^{s/\nu+d-2} \sim L^{\zeta_G/\nu}$) for $L < \xi$, where $\zeta_R = t - (d-2)\nu$ (or $\zeta_G = s + (d-2)\nu$) [17].

Comparing equations (17) with (19) and equations (18) with (20), we can obtain the relation between $\kappa((\beta+2)/2)$ ($\kappa'((\beta+2)/2)$) and $\psi_R((\beta+2)/2)$ ($\psi_G((\beta+2)/2)$):

$$\kappa\left(\frac{\beta+2}{2}\right) = \psi_R\left(\frac{\beta+2}{2}\right) + \frac{\beta}{2}d\nu - \frac{\beta+2}{2}\zeta_R \tag{21}$$

and

$$\kappa'\left(\frac{\beta+2}{2}\right) = \psi_G\left(\frac{\beta+2}{2}\right) + \frac{\beta}{2}d\nu - \frac{\beta+2}{2}\zeta_G. \tag{22}$$

Substituting equations (21) and (22) into equations (15) and (16), we have

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

and

$$v(\beta) = \psi_G\left(\frac{\beta+2}{2}\right) + \frac{\beta+2}{2}(s - \zeta_G) + \frac{\beta}{2}d\nu. \tag{24}$$

These results are new and different from those given in previous work [13].

Analytic and numerical results for $\psi_R((\beta+2)/2)$ have been obtained for a d-dimensional random resistor network in the vicinity of the percolation threshold f_c or g_c [16]. One of the approximate expressions is

$$\phi_R\left(\frac{\beta+2}{2}\right) = 1 + (\nu D_B - 1)^{[1-(2+\beta)/2]}(\zeta_R - 1)^{(\beta+2)/2} \tag{25}$$

where D_B is the fractal dimensionality of the backbone [16, 17].

For $d = 2$, from duality considerations, it has been shown that $\psi_R((\beta+2)/2)$ for a random resistor network coincides with $\psi_G((\beta+2)/2)$ for a random superconducting network [18]. Thus equation (24) can be used to discuss the critical behaviour in two-dimensional S/N systems. The known parameters, such as ν, D_B, ζ_R (ζ_G) and t (s), for different dimensions, will be used in the following quantitative calculation. Their values are readily available in the literature and we refer the reader to [16].

Using a hierarchical model of the two-phase percolation structure near the percolation threshold, Morozovsky and Snarskii [19] proposed simple expressions:

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

and

$$\kappa'\left(\frac{\beta+2}{2}\right) = [2\nu - s]\frac{\beta}{2}.$$

Thus equations (15) and (16) can also be expressed simply as [15]

$$u(\beta) = t + \beta[t - \nu(d-1)] \tag{26}$$

and

$$v(\beta) = s + \nu\beta. \tag{27}$$

Equations (26) and (27) are simple and can also be used to describe the critical behaviour of the nonlinear response in the N/I or S/N system. More importantly, with equation (27), we can easily get information about $v(\beta)$ for larger-dimensional S/N systems.

Numerical results for $u(\beta)$ in the N/I limit and $v(\beta)$ in the S/N limit for different dimensions are shown in figures 1 and 2, respectively.

For a N/I composite, our approximations give reasonable results for $d > 2$ (see figure 1(a)). At first, whether based on equations (23) and (26) or the EMA [7, 13], $u(\beta)$ is always positive. Thus for $d > 2$, as f is close to the percolation threshold f_c, from the above, $\chi_e(\beta)$ vanishes for arbitrary β. Such behaviour is similar to that of the effective linear conductivity σ_e, as $f \to f_c^+$ for such N/I composites. Secondly, the larger the

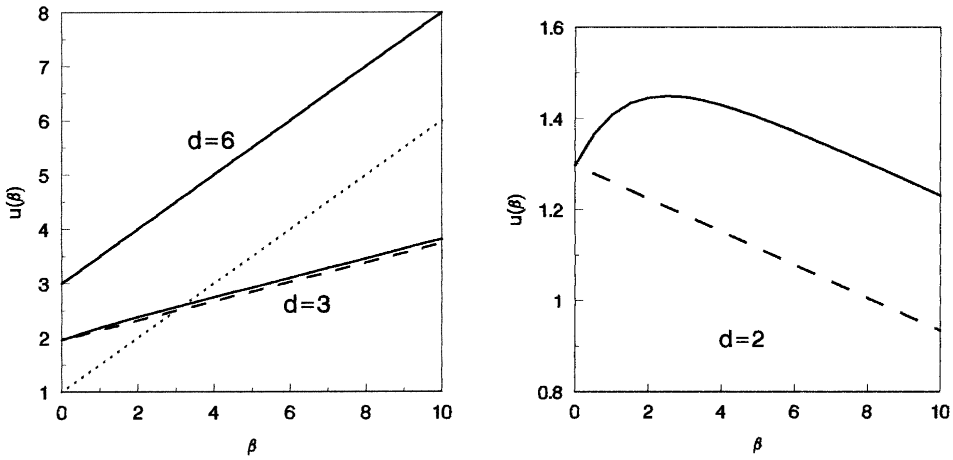
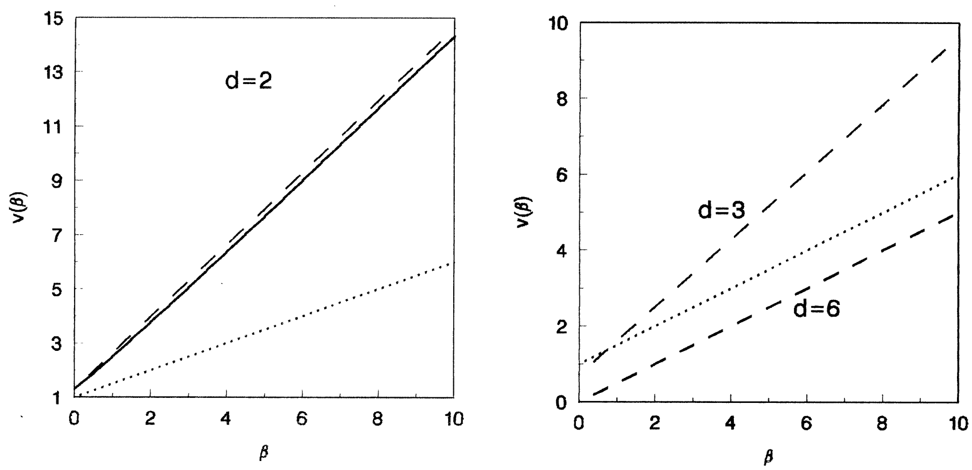


Figure 1. (a) For a N/I composite, the critical exponent $u(\beta)$ of the nonlinear susceptibility $\chi_e(\beta)$ as a function of the nonlinear exponent β for $d = 3$ and $d = 6$. The results come from equation (23) (solid line), equation (26) (dashed line) and the EMA (dotted line). Note that equations (23) and (26) predict same results for $d = 6$. (b) For a N/I composite, the critical exponent $u(\beta)$ of the effective nonlinear susceptibility $\chi_e(\beta)$ for $d = 2$ is plotted versus the nonlinear exponent β. The key to the curves is the same as for (a).

nonlinearity β, the bigger the critical exponent $u(\beta)$. This corresponds to the fast vanishing of $\chi_e(\beta)$ with the increase of β. Thirdly, $u(\beta)$ appears to be an increasing function of the dimensionality d, which implies that $\chi_e(\beta)$ vanishes quickly for larger d. We also notice that equations (23) and (26) predict reasonable agreement for $d = 3$ and give the same results for $d = 6$; therefore the representation is valid. It is known that the EMA gives the crude estimate $u(\beta) = v(\beta) = (\beta + 2)/2$ [7, 13], which is independent of d. For comparison, the results obtained using the EMA (the dotted line) are also depicted in figure 1; it is evident that the EMA gives the correct behaviour of $\chi_e(\beta)$ qualitatively, but predicts incorrect critical exponents near the percolation threshold [13].

For two-dimensional N/I systems, more complex critical behaviour is shown in figure 1(b). On the basis of equation (23), we find that there is a critical value $\beta_c \approx 2.53$ at which $u(\beta_c)$ has a maximum value. Thus χ_e vanishes quickly in the region $0 < \beta < \beta_{c1} \approx 2.53$, then vanishes slowly with the increase of β. Equation (26) cannot give such interesting behaviour, but produces a monotonic decrease with the increase of the nonlinearity.

The critical behaviour $v(\beta)$ of the nonlinear susceptibility $\chi_e(\beta)$ in the S/N limit is plotted as a function of β in figure 2(a). Both equation (24) and equation (27) predict $v(\beta) > 0$ and, thus, divergence of χ_e as $g \to g_c^-$. As β increases, χ_e also diverges quickly. This means that the enhancement of the nonlinear susceptibility is greater for larger values of the nonlinearity β [7]. Because the expressions for $\psi_G((\beta + 2)/2)$ for larger dimensions are unknown, previous work cannot be used for discussing the critical behaviour in the larger-dimensional S/N case. Here we can only resort to equation (27) to analyse such a situation. Theoretical results obtained by using equation (27) are shown in figure 2(b) for the $d = 3$ and $d = 6$ cases. $v(\beta) > 0$ for arbitrary β; thus as $g \to g_c^-$, not only does σ_e diverge, but also χ_e diverges and it diverges faster for larger β. From figure 2, we also find, with the increase of d, that $v(\beta)$ diverges quickly. From a comparison to the EMA results, we find that the EMA also gives a qualitative description.



(a) (b)

Figure 2. (a) For a two-dimensional S/N composite, the critical exponent $v(\beta)$ of the effective nonlinear susceptibility χ_e versus β for $d = 2$. The key to the curves is the same as for figure 1(a). (b) For a higher-dimensional S/N system, $v(\beta)$ versus β for $d = 3$ and $d = 6$. The key to the curves is the same as for figure 1(a).

3. Conclusions

In the present work, we have analysed the relation between L and ξ in detail and thus established that L and ξ must not be confused. Furthermore, we have studied the critical behaviour of arbitrary $(\beta + 1)$th-order nonlinear random composites for the N/I and S/N limits by investigating the relationship between the effective nonlinear susceptibility in random nonlinear composites and the resistance (or conductance) fluctuations in the corresponding linear composites for arbitrary dimensions. The critical properties $u(\beta)$ and $v(\beta)$ are important and necessary for the investigation of nonlinear susceptibilities with arbitrary β. Our results are new and different from those given in previous work. Our conclusions are as follows.

(1) The critical exponent $u(\beta)$ for a three-dimensional N/I system is always positive and takes on the form of a monotonically increasing function of the nonlinear exponent β; the larger β, the faster the nonlinear susceptibility $\chi_e(\beta)$ vanishes as $f \to f_c^+$.

(2) The critical exponent $v(\beta)$ for a two-dimensional S/N system is also positive and shows a monotonic increase with the increase of β. Therefore, $\chi_e(\beta)$ diverges quickly as β increases.

(3) $u(\beta)$ for $d = 2$ in the N/I limit exhibits anomalous behaviour. On the basis of equation (23), we predict that there exists a critical value β_c (≈ 2.52) which characterizes the maximum of $u(\beta)$. This indicates that the effective nonlinear susceptibility in a N/I system exhibits a different critical behaviour with the increase of β, while on the basis of equation (26) we only obtain a monotonic decrease with the increase of β.

(4) $u(\beta)$ in the six-dimensional N/I limit and $v(\beta)$ in higher-dimensional $(d = 3, 6)$ S/N cases are always positive. They increase as the nonlinearity increases. Previous work cannot describe them.

The theoretical results presented in this paper are of potential practical use, especially for percolating S/N composites which have high conductivity and yet are highly nonlinear, and may be useful in extracting the critical behaviour from experimental data [20].

We mainly concentrate on the critical behaviour of the effective nonlinear properties; we can take a further step and discuss the crossover effect, for which the linear and nonlinear response become comparable. For realistic composites, the ratio of the conductivity of the poor conductor to that of the good conductor ($h \equiv \sigma_1/\sigma_2$) may not be zero. Such composites will also be worth studying, because h governs the crossover from fractal ($h = 0$) to homogeneous $h = 1$ behaviour [15]. This problem will be resolved in the future.

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