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The enhanced nonlinear response of composite wires: crossover from one- to three-dimensional behaviour

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Abstract. The effective response is calculated for nonlinear composite wires which are modelled as random nonlinear conductance networks consisting of two different kinds of conductor, with lateral size L and square cross section of width $\delta \ll L$. The first kind is nonlinear and obeys a current–voltage (I–V) characteristic of the form $I = \sigma_1 V + \chi_1 V^3$, while the second one is linear with $I = \sigma_2 V$. We invoke a renormalization-group (RG) analysis to rescale the wire repeatedly by small-cell transformations to obtain a chain of nonlinear conductors, for which exact formulas for the effective linear response σ_e and nonlinear response χ_e are available. We calculate σ_e and χ_e as functions of the volume fraction and examine the dependence on δ. We observe a large enhancement in the nonlinear response as well as an interesting crossover from one- to three-dimensional behaviour as δ increases. Numerical simulations are performed to validate the RG calculations.

1. Introduction

The physics of nonlinear inhomogeneous media, in which nonlinearity and inhomogeneity are present simultaneously, has been a subject of much interest because of their potential applications [1–6]. During the past few years, much effort has been devoted to calculations of the effective response in nonlinear composite media consisting of two or more materials [1, 7–13]. In the weakly nonlinear regime in which the nonlinearity can be treated as a small perturbation, various methods have been established [7–9]. Recently much attention has been concentrated on strongly nonlinear composites [10–13].

It is observed that the effective response of composite media can differ drastically from that of their constituents [1]. The widely varying constitutive properties may lead to large fluctuations in the local electric fields and to large enhancements in the effective properties. Such an enhancement effect may be more pronounced in nonlinear composites [14]. It has been shown that the effective nonlinear response can be enormously enhanced near the percolation threshold under appropriate conditions [15]. Recently, large enhancements in the effective nonlinear response have also been found in fractal clustering in nonlinear composites [16]. It is therefore believed that the effective nonlinear response may depend strongly on the microgeometry and dimensionality of the composite systems.

Recently, significant advances have been made in materials fabrication techniques. By means of molecular beam epitaxial techniques, samples of various materials with desired geometry, size, interface and surface conditions have been made available. In this work, we use a simple nonlinear conductance network model to study the effective nonlinear response of composite wires. We apply a renormalization-group (RG) analysis for calculating the

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effective linear and nonlinear response. We find large enhancements in the effective nonlinear response under certain conditions and a crossover from one- to three-dimensional behaviour as the width increases. Numerical simulations are performed to validate the RG calculations.

The paper is organized as follows. In the next section, we describe the model for weakly nonlinear composites and present the established formulas for the effective linear and nonlinear response. In section 3, we derive the exact formulas for the effective linear and nonlinear response of a chain of nonlinear conductors. In section 4, we use the RG analysis to calculate the effective linear and nonlinear response of composite wires. We shall perform numerical simulations to validate the RG calculations. Possible generalizations of the present work will be discussed.

2. Model and method

Consider a d-dimensional (dD), two-component hypercubic random nonlinear conductance network which consists of two kinds of conductor. The first kind is assumed to be nonlinear and obeys a current–voltage (I–V) characteristic of the form

$$I = \sigma_1 V + \chi_1 V^3 \tag{1}$$

where σ_1 and χ_1 are the linear and nonlinear conductance respectively. Throughout this work, the nonlinearity is assumed to be weak so that $\chi_1 V^2/\sigma_1 \ll 1$ and we restrict ourselves to cubic nonlinearity. Generalizations to arbitrary nonlinearity are possible. The second component is ohmic with

$$I = \sigma_2 V \tag{2}$$

where σ_2 is the linear conductance. The nonlinear conductors are randomly assigned to the network with a volume fraction p while the volume fraction of the second component is $q = 1 - p$. We aim at calculating the effective linear and nonlinear response of the network, represented by a homogeneous network of identical conductors, each of which has an I–V characteristic of the form

$$I = \sigma_e V + \chi_e V^3 \tag{3}$$

where σ_e and χ_e are the effective linear and nonlinear response respectively and are given by the voltage-summation formulas [8, 9, 15]

$$\sigma_e = \frac{1}{\Omega} \sum_\alpha \sigma_\alpha V_\alpha^2 \tag{4}$$

$$\chi_e = \frac{1}{\Omega} \sum_\alpha \chi_\alpha V_\alpha^4 \tag{5}$$

where σ_α and χ_α are the linear and nonlinear response of the αth conductor; and $\Omega = \prod_{i=1}^{d} L_i$, where L_i is the lateral size along the ith Cartesian direction. Without loss of generality, we apply a voltage along the x_1-direction and free boundary conditions in the remaining $(d-1)$ directions. We adopt the convention that the voltage across the two opposite $(d-1)$-dimensional hyperplanes is $V = L_1$ so that $V_\alpha = 1$ for bonds along x_1 in the limit of a homogeneous network. In equations (4) and (5), 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 $\chi_\alpha = 0$). The summation is performed over all conductors in the network.

In what follows, we shall consider a cylindrical wire of length $L_1 = L$ and of square cross section of width $L_2 = \delta \ll L$. We calculate σ_e and χ_e and examine their dependence on δ. An interesting crossover from one- to three-dimensional behaviour will be observed.

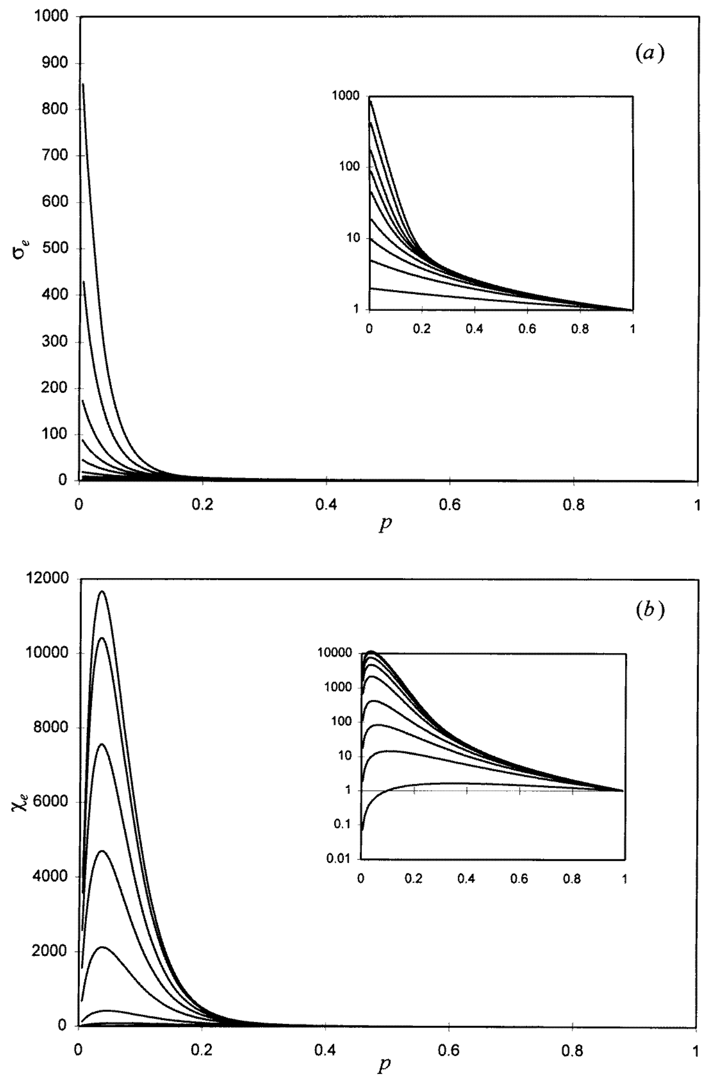

Figure 1. For $L = 32$ chains, the normalized effective (a) linear response (σ_e/σ_1) and (b) nonlinear response (χ_e/χ_1) are plotted as functions of the volume fraction p for various conductance ratios h in the S/N limit. We observe a large enhancement in the effective nonlinear response χ_e, which increases with h while the location of the maximum nonlinear response occurs at $p^* \approx 1/L$, roughly independently of h. For clarity, we show in the insets σ_e/σ_1 and χ_e/χ_1 in semi-logarithmic plots. From bottom to top in order of increasing conductance ratio: $h = 2$, 5, 10, 20, 50, 100, 200, 500 and 1000.

Consider a chain ($\delta = 1$) of L conductors, k of which are of type 1 (σ_1, χ_1), while the remaining $(L - k)$ of which are of type 2 (σ_2, χ_2), subject to an applied voltage $V = L$ along the chain. For convenience, we denote the conductance ratio as $h = \sigma_2/\sigma_1$. For series combination of linear conductances, we obtain the voltages $V_1 = hL/(L-k+hk)$ and $V_2 = L/(L-k+hk)$ across type 1 and 2 conductors respectively. By using the voltage-summation formulas, together with simple combinatorial considerations, we arrive at the *exact* formulas for the effective linear and nonlinear response of a chain:

$$\sigma_e = \sum_{k=0}^{L} \binom{L}{k} p^k q^{L-k} \frac{L\sigma_1\sigma_2}{(L-k)\sigma_1 + k\sigma_2} \tag{6}$$

and

$$\chi_e = \sum_{k=0}^{L} \binom{L}{k} p^k q^{L-k} \frac{L^3[\chi_1 k h^4 + \chi_2(L-k)]}{(L-k+hk)^4}. \tag{7}$$

We study two limits: (1) the normal-conductor–insulator (N/I) case in which the second component conducts less well ($h \ll 1$) and (2) the superconductor–normal-conductor (S/N) case in which the second component conducts better ($h \gg 1$). The chain formulas can be used to calculate σ_e and χ_e for both cases. For the N/I case, we find a large decrease in the effective nonlinear response while a large enhancement occurs for the S/N case [15]. We shall present results for the S/N case for illustration. The length of wire $L = 32$. In figure 1, we plot the normalized effective linear response (σ_e/σ_1) and nonlinear response (χ_e/χ_1) as a function of the volume fraction p for various conductance ratios $h > 1$. We observe a large enhancement in the effective nonlinear response χ_e. The enhancement increases with the conductance ratio while the locations of maximum response of χ_e occur at $p^* \approx 1/L$. The location of peak p^* is determined numerically from equation (7) in the limit of large h. We obtain $p^* = 0.268$, 0.135, 0.067, 0.034 and 0.017 for $L = 4$, 8, 16, 32 and 64 respectively. The result is in reasonable agreement with the numerical calculations, as is evident in figure 1. For clarity, we also show the effective response in semi-logarithmic plots in the insets.

3. Linear and nonlinear response of composite wires

When inter-chain couplings are present, we may model the system with a nonlinear composite wire of a finite cross section of width δ. We resort to a renormalization-group (RG) analysis [17–19] because we believe that RG analysis is able to capture local field fluctuations better than the effective-medium approximation [7]. We start out with a wire of dimension $L \times \delta \times \delta$, with initial parameters $p, q, \sigma_1, \sigma_2, \chi_1$ and χ_2; $h = \sigma_2/\sigma_1$. The idea of RG analysis is to rescale the wire repeatedly via a simple small-cell transformation, to obtain a chain of a shorter length. In this way, we obtain a set of renormalized parameters $p', q', \sigma_1', \sigma_2', \chi_1'$ and χ_2'; $h' = \sigma_2'/\sigma_1'$. We perform a simple $2 \times 2 \times 2$ cell RG analysis to reduce the dimension of the wire to $(L/2) \times (\delta/2) \times (\delta/2)$; standard RG techniques are used to calculate the renormalized quantities [19]

$$q' = R_2(q) \tag{8}$$

$$\sigma_1' = \Phi_1(\sigma_1, \sigma_2, p) \tag{9}$$

$$\sigma_2' = \Phi_2(\sigma_1, \sigma_2, p) \tag{10}$$

$$\chi_1' = \Psi_1(\chi_1, \chi_2, h, p) \tag{11}$$

$$\chi_2' = \Psi_2(\chi_1, \chi_2, h, p) \tag{12}$$

where $R_2(q) = 4p^{10}q^2 + 48p^9q^3 + 238p^8q^4 + 616p^7q^5 + 856p^6q^6 + 776p^5q^7 + 493p^4q^8 + 220p^3q^9 + 66p^2q^{10} + 12pq^{11} + q^{12}$, $p' = 1 - q'$; Φ_1, Φ_2, Ψ_1 and Ψ_2 are transformations of their arguments. In obtaining equations (9)–(12), one has to invoke either a geometrical or arithmetic mean over 3330 spanning-superconductor and 766 nonspanning-superconductor configurations [18] respectively while equations (11) and (12) represent a generalization of the established RG analysis [17–19] to the nonlinear response. We then repeat the RG analysis several times until $\delta' = 1$. For a $\delta = 2$ wire, one RG procedure already meets the need, while for a $\delta = 4$ wire, two consecutive RG procedures are needed to obtain a chain of a shorter length L', with renormalized quantities $p', \sigma_1', \sigma_2', h', \chi_1'$ and χ_2'. The chain formulas (equations (6) and (7)) can therefore be used to obtain the effective linear and nonlinear response of composite wires.

We first present numerical RG results for a $\delta = 2, L = 32$ wire. In figure 2, we plot σ_e/σ_1 and χ_e/χ_1 as a function of the volume fraction p for various conductance ratios h. Again, we observe a large enhancement in the effective nonlinear response. The enhancement increases with the conductance ratio while the location of maximum response of χ_e has shifted to a larger p^*, an estimate of which will be discussed below. However, it is also noted that the strength of enhancement has decreased slightly.

4. Exact formulas for linear and nonlinear response of a chain of nonlinear conductors

We then present numerical RG results for a $\delta = 4, L = 32$ wire. In figure 3, we plot σ_e/σ_1 and χ_e/χ_1 as functions of the volume fraction p for various conductance ratios h. Similarly, we observe a large enhancement in the effective nonlinear response. The enhancement increases with the conductance ratio while the location of maximum response of χ_e has now shifted to an even larger p^*, indicating a crossover from 1D to 3D behaviour. It is also noted that the strength of enhancement has decreased substantially as compared to the small-δ cases due to to the reduced fluctuations of local electric fields present at larger δ.

It is instructive to establish a result for the location of peak via the RG analysis. For $\sigma_2 > \sigma_1$, a large enhancement of χ_e occurs in the vicinity of the percolation threshold of the better-conducting component [14, 15]. Since percolation of the better-conducting component occurs at $p_c = 0$ and 0.751 in 1D and 3D respectively [20], we expect a large enhancement of χ_e to occur somewhere between $p = 0$ and 0.751 for finite L and δ. In the language of RG analysis, the shift of p^* towards the 3D value as δ increases is attributed to the fact that the renormalized chain length decreases with δ. By using equation (8), we obtain for $32 \times \delta \times \delta$ wires, $p^* = 0.034$, 0.390, 0.643, and 0.740 for $\delta = 1$, 2, 4, and 8 respectively, values that are in good agreement with the numerical calculations presented in figures 2 and 3. These values can be interpreted as the effective percolation threshold of a wire of finite width [19, 21]. It is evident that the peak of response shifts towards the 3D percolation threshold $p_c = 0.792$ as δ increases. The change from 1D to 3D behaviour is quite rapid as is evident from the figures. The present RG analysis is noted to give an overestimate of $p_c = 0.751$ in 3D bond-percolation networks [20].

To confirm the RG result, it is instructive to perform numerical simulations in nonlinear conductance networks [15]. We have done simulations for $32 \times 2 \times 2$ and $32 \times 4 \times 4$ networks as a generalization of the previous simulation for 2D random nonlinear conductance networks [15]. Details of the numerical simulations can be found in [15]. Again, we present results for the S/N case only. In the insets of figures 2 and 3, we also show the normalized effective linear and nonlinear response of numerical simulation in a semi-logarithmic plot for comparison. As is evident from the figures, very good agreement between the RG and simulation data is obtained for the effective linear response. For the effective nonlinear response, while good agreement is obtained near the peaks, strong deviations are nevertheless observed at small p-values. This is attributed to the fact that

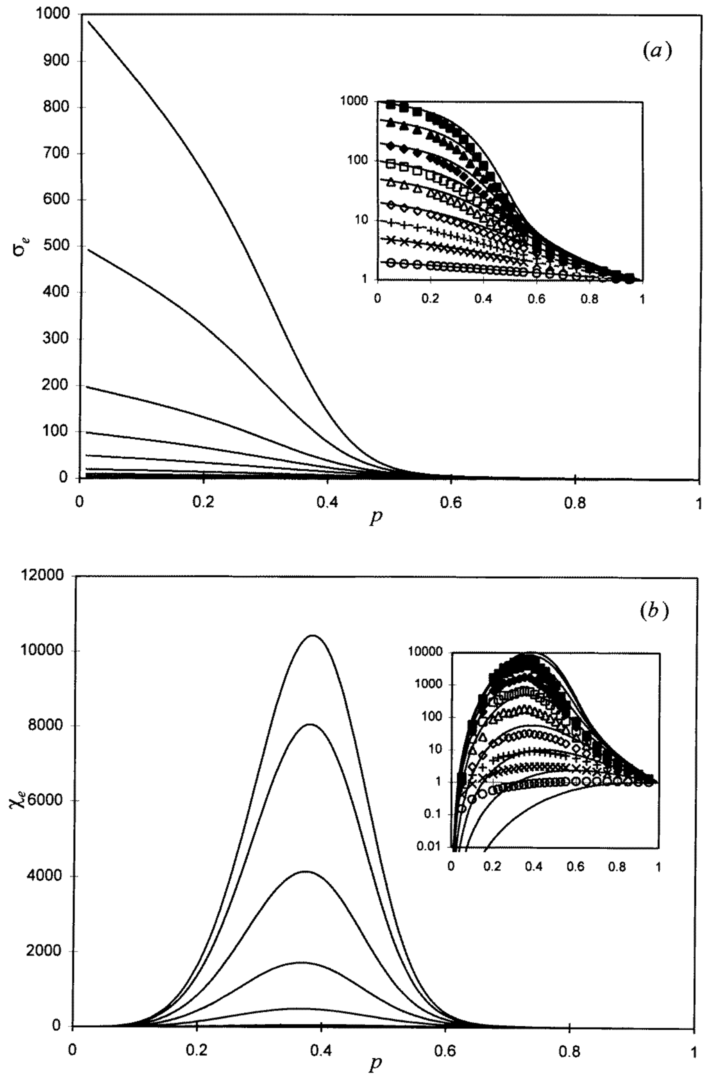

Figure 2. For $\delta = 2$ wire and $L = 32$, the (a) linear response σ_e/σ_1 and (b) nonlinear response χ_e/χ_1 are plotted as functions of the volume fraction p for various values of h. Again, we observe a large enhancement in χ_e. The location of the maximum nonlinear response has shifted to a larger p^*, the value of which coincides with the estimate from RG analysis. It is also noted that the strength of enhancement has decreased slightly. In the insets, we show the numerical simulation data (symbols) in semi-logarithmic plots. From bottom to top in order of increasing conductance ratio: $h = 2$, 5, 10, 20, 50, 100, 200, 500 and 1000. As is evident from the figures, a reasonable agreement between the RG and simulation data is obtained.

while the RG analysis generally gives reasonable critical behaviour near the percolation threshold p_c—namely, reasonable numerical values for p_c and critical exponents [19]—it may not capture the correct behaviour away from p_c.

In this connection, it is tempting to fit the simulation data via a simple effective-medium approximation (EMA) [7]. It has been found that while the EMA can be used to fit the

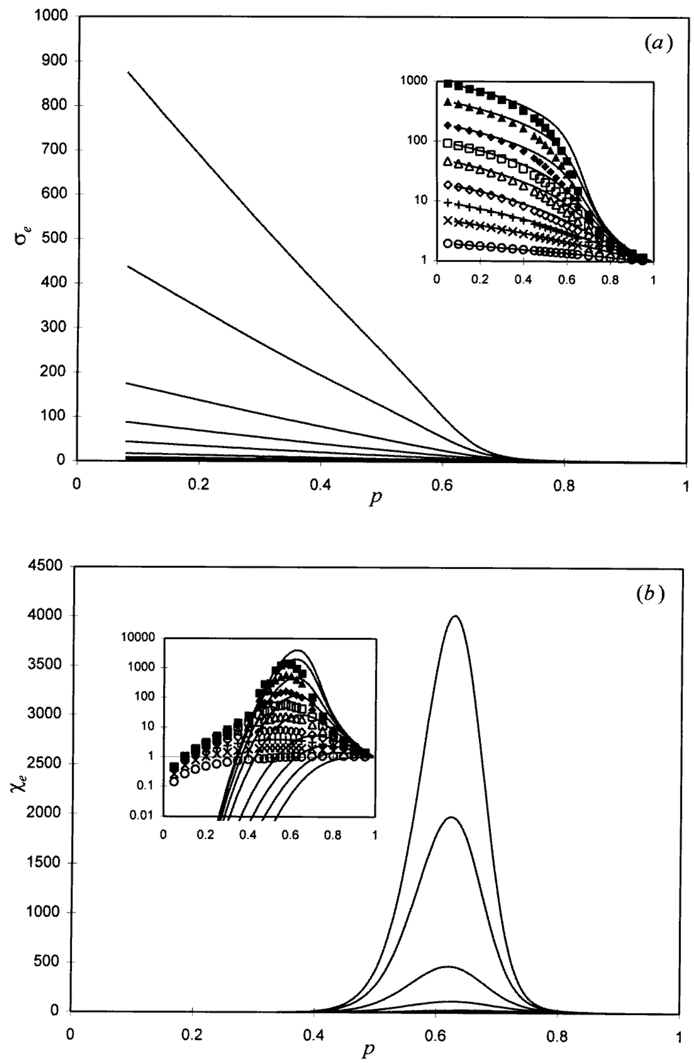

Figure 3. Similar to figure 2, but for a $\delta = 4$ wire. We observe a large enhancement in χ_e. The location of the maximum response of χ_e has now shifted to an even larger p^*, indicating a crossover from 1D to 3D behaviour. In the insets, we also show the simulation data (symbols). As is evident from the figures, a reasonable agreement between the RG and simulation data is obtained.

simulation data of σ_e reasonably well, strongly deviations are nevertheless observed for the effective nonlinear response χ_e. This is attributed to the fact that the EMA ignores local field fluctuations explicitly [22].

In fact, such an enhancement in the effective nonlinear response was found earlier by Levy and Bergman [23] in weakly nonlinear conductivity of a two-component composites. In [23], a scaling *ansatz* for χ_e was proposed, similar to that proposed for the scaling behaviour of flicker noise in two-component composites [24]. It should be remarked that a significant result of our calculations is the dependence of the enhancement on the small dimensions of a composite wire.

5. Discussions and conclusions

In conclusion, the effective response has been calculated in nonlinear composite wires and wires with lateral size L much larger than the width δ. We have used the renormalization-group analysis to calculate the effective linear and nonlinear responses as functions of the volume fraction p and examine their dependence on δ. We observe large enhancements in the nonlinear response under appropriate conditions, as well as interesting crossover from 1D to 3D behaviour as δ increases. Numerical simulations are performed and compare well with the RG calculations.

Here a few comments on the results are in order. Although the discussion has been limited to percolative conduction, generalization can readily be made to dielectric response at finite frequencies. The enhancement as well as dimensionality crossover effects may possibly be observed in experiments on electrorheological (ER) systems where an inherent nonlinear characteristic occurs due to the formation of columnar structures in the ER systems under the application of intense electric fields [25]. Possible experiments may also be done on the optical properties of nonlinear composite wires.

Moreover, generalization can also be made to composite thin films [26] of metallic particles embedded in dielectric hosts, in which case δ plays the role of the thickness of the film and a crossover from 2D to 3D behaviour can be observed. Possible experiments may also be done on the optical properties of nonlinear composite thin films. In this connection, a simple EMA is shown to give a reasonable fit of the simulation data [26] in linear composite thin films. However, we believe that the present RG analysis may fit the effective nonlinear response better [27]. In order to test the asymptotic scaling theory [21], pertaining to the 2D–3D dimensionality crossover, extensive simulation as well as RG analysis should be performed. Relevant studies include the extraction of the effective percolation threshold, critical exponents and universal scaling functions [15]. This work is left for future studies [27].

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