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Effective medium theory for strongly nonlinear composites: fractal clusters

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Abstract

We have developed an effective medium approximation (EMA) for the effective nonlinear response due to clustering of a strongly nonlinear conducting material with a current-field (J–E) response of the form $J = \chi|E|^{2\beta}E$ ($\beta > 0$) in a host medium, where χ is the nonlinear coefficient. A cluster energy functional is constructed and a variational principle is invoked to obtain an expression for the effective nonlinear response of a fractal cluster. The EMA results are compared with numerical calculations in a deterministic fractal model and excellent agreement is found.

1. Introduction

The physics of nonlinear inhomogeneous media has received much attention in recent years [1,2]. In particular, attention has been paid to a class of strongly nonlinear conducting composite media with a power-law nonlinearity which occurs when a sufficiently strong field is applied to condensed matter [3–5]. For this composite system, the inclusion and the host medium obey a local current-field (J–E) relation of the form $J = \chi|E|^{2\beta}E$, and $\beta > 0$. For such a nonlinear relation, we have recently obtained the dilute-limit expressions for the effective response of a small volume fraction of spherical inclusions embedded in a host medium [3]. In order to extend the theory to a random composite at larger volume fractions, we develop an effective-medium approximation (EMA), which generally gives a better comparison with numerical simulations [4].

However, the approach is only valid for *truly* random composites in the dilute limit. Many growth and fabrication processes may indeed produce spatial correlations in realistic composites. In particular, a fractal clustering will be generated via various aggregation processes [6–8]. The fractal geometry should have an observable effect on the nonlinear properties [9,10] (for a recent review, see Ref. [11]). In this work, we aim at developing an EMA for the effective nonlinear response of clustering strongly nonlinear materials in a host medium, in which case a similar approximation in weakly nonlinear composites [12] cannot be applied.

2. Formalism

We consider a class of strongly nonlinear composite media which obey a current-field response of the following form [3–5],

$$J = \chi|E|^{2\beta}E, \tag{1}$$

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where $\beta > 0$. The nonlinear coefficient χ will take on different values in the inclusion and in the host medium. An external electric field $\boldsymbol{E}_0$ is applied. In Ref. [3], we invoke a variational principle by minimizing the energy functional,

$$W[\boldsymbol{E}] = \frac{1}{E_0^{2+2\beta} V} \int_V \boldsymbol{J}(\boldsymbol{x}) \cdot \boldsymbol{E}(\boldsymbol{x})\, \mathrm{d}V, \tag{2}$$

where V is the volume of the composite. When the minimum condition is satisfied by a trial electric field $\tilde{\boldsymbol{E}}$, then by using Eqs. (1) and (2), the effective nonlinear response χ_e can be obtained,

$$\chi_e = \tilde{W} = \frac{1}{E_0^{2+2\beta} V} \int_V \chi(\boldsymbol{x}) |\tilde{\boldsymbol{E}}(\boldsymbol{x})|^{2+2\beta} \mathrm{d}V. \tag{3}$$

The trial electric field $\tilde{\boldsymbol{E}}$ will be taken as the solution of the linear problem [3].

Let us consider a problem in d dimensions, i.e. of spherical inclusions in three dimensions and cylindrical inclusions in two dimensions (2D) of radius ρ and nonlinear coefficient χ_i suspended in a host medium of χ_m, the energy functional is given by [3]

$$W_\beta(b) = \chi_m + p\chi_m Q_\beta(b) + p\chi_i (1-b)^{2+2\beta}, \tag{4}$$

where b is a variational parameter as yet to be determined and p is the volume fraction of the inclusion. The quantity $Q_\beta(b)$ generally depends on β and d; $Q_\beta(b)$ is a polynomial in b for integral values of β, while for nonintegral values of β, $Q_\beta(b)$ is an infinite series of b [4]. Note that $W_\beta(b)$ has no direct relevance to the effective nonlinear response until we minimize it with respect to b and hence determine χ_e. Eq. (4) allows us to obtain the dilute-limit expression for the effective nonlinear response of a small volume fraction of strongly nonlinear materials embedded in a host medium [3].

We are now in a position to develop the EMA for the strongly nonlinear response χ_e of fractal clusters. We shall use Eq. (4) to derive an approximate expression for the effective nonlinear response of a cluster. We first construct an energy functional $C_\beta(L, b)$ for a cluster of radius L. We then minimize $C_\beta(L, b)$ with respect to b to find an expression for the effective nonlinear response $\chi_e(L)$ of fractal clusters. In order to describe a fractal cluster of type 1 embedded in a host medium of type 2, we start with a pure type 1 inclusion of radius ρ, at which $p_1 = 1$ and $p_2 = 0$. The volume fraction of host medium is increased by adding type 2 material. Now increase L by δL and the volume fraction by δp_2. The size dependence of volume fraction ($p_2(L)$) will be discussed below. Then from Eq. (4), we find

$$\delta C_\beta = \delta p_2 [Q_\beta(b) C_\beta + (1-b)^{2+2\beta} \chi_2], \tag{5}$$

which is an ordinary differential equation for $C_\beta(L, b)$ and can be solved to give

$$C_\beta = \chi_1 f^{-Q_\beta(b)} + \frac{\chi_2 (1-b)^{2+2\beta}}{Q_\beta(b)} (f^{-Q_\beta(b)} - 1), \tag{6}$$

where $f = 1 - p_2$ is the volume fraction of the cluster; we have explicitly used the initial condition that $C_\beta(L, b) = \chi_1$ at $L = \rho$ or $f = 1$. In this way, we obtain a cluster energy functional C_β for a cluster of radius L. An apparent divergence in C_β at the extremely dilute limit ($f \to 0$) will not occur in χ_e because in this limit $b \to 0$, and concomitantly $Q_\beta(b) \to -1$, hence $\chi_e \to \chi_2$.

It should be remarked that the approach does not necessarily assume that the cluster is fractal. If, however, the cluster is indeed a fractal of fractal dimen-

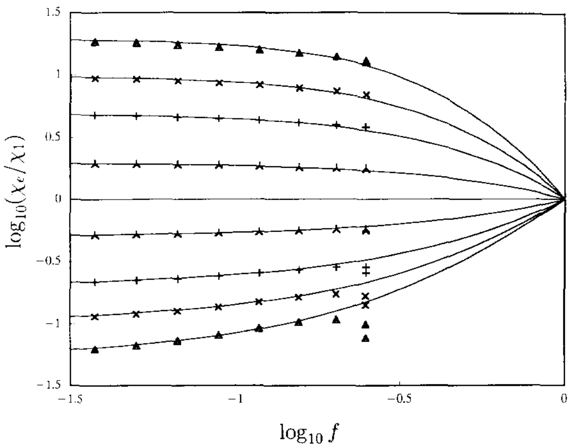

Fig. 1. Normalized effective nonlinear cluster response χ_e/χ_1 in EMA (solid lines) and numerical simulations (symbols) plotted against the volume fraction f for various ratios of conductivity χ_2/χ_1. From top and downwards in order of decreasing ratio of conductivity: χ_2/χ_1 = 20, 10, 5, 2, 0.5, 0.2, 0.1, and 0.05. Note the remarkable agreement between EMA and simulations results.

sion d_f, then the volume fraction of nonlinear fractal inclusion is related to the cluster size as

$$f = (L/\rho)^{-(d-d_f)}. \tag{7}$$

We are ready to obtain numerical results for the EMA cluster response. We shall present results in 2D to compare with numerical simulations in a deterministic fractal cluster [13]. In what follows, we restrict ourselves to a cubic nonlinearity ($\beta = 1$). The expression for $Q_1(b)$ is given by [4]

$$Q_1(b) = -1 + 4b + 4b^2 + \tfrac{1}{3}b^4, \tag{8}$$

valid in 2D. In Fig. 1, we plot the normalized effective nonlinear response χ_e/χ_1 as a function of the volume fraction f of fractal for various ratios χ_2/χ_1. As seen from Fig. 1, we confirm that as the cluster size increases and f decreases from unity towards zero, χ_e varies from χ_1 to χ_2.

3. Numerical calculations in deterministic fractal clusters

We attempt numerical calculations of the effective response of a deterministic fractal cluster (DFC) which is constructed recursively from a simple basic unit [13], in order to compare with the EMA results. The cluster has a fractal dimension $d_f = \log 3/\log 2 = 1.585$ and the volume fraction $f = (\frac{3}{4})^n$, where n is the generation [13]. We construct a fractal network by mapping the DFC onto a 2D square network (now $f \approx (\frac{3}{4})^n$ for large n) and associating each bond with two types of nonlinear conductors obeying a current–voltage (I–V) response of the form

$$I = \chi_i V^3, \tag{9}$$

where χ_i ($i = 1, 2$) is the nonlinear coefficient and V the voltage across the conductors. The effective response of the network is defined as that of a homogeneous network of identical conductors, each of which has a response of the form

$$I = \chi_e V^3. \tag{10}$$

A unit voltage is applied across the top and bottom bars of the network. The nonlinear Kirchhoff circuit equations for each node are solved by the relaxation method. When convergence is achieved, the current going into the top bar and that going out of the bottom bar will be the same. The effective nonlinear response of the network is used to compare with the EMA for the same f. The simulation is performed up to the ninth generation. In Fig. 1, we plot the normalized response χ_e/χ_1 against f. Except for large volume fractions, the simulation results are in excellent agreement with the EMA results. The relatively large deviation of the numerical data from the EMA is probably due to the fractal limit being not yet achieved for small generations ($n \leqslant 3$) of the DFC.

4. Discussions and conclusions

Here a few comments are in order. In this work, we develop an EMA for fractal clusters through the construction of a cluster energy functional $C_\beta(L, b)$. We may adopt an alternative approach and determine the cluster response directly from the dilute-limit expression $\tilde{W}_\beta(\tilde{b})$, where $\tilde{b}$ is found from minimizing Eq. (4). We prefer the present approach as it is computationally much simpler and it gives results at most a few percent different from those of the alternative consideration. More importantly, it compares better with numerical simulations. Although the present approach deals with strongly nonlinear composites, with slight modifications, the variational method can be applied to arbitrary nonlinearity as well. However, as it has been recently shown that even if one considers clustering of a weakly nonlinear material in a host medium, the effective nonlinear response can be largely enhanced [14, 15] in the extremely dilute limit ($f \ll 1$), the strongly nonlinear approach will be viable. Moreover, our results may have relevance to a recent experiment on laser-irradiated polymers [16], where a power-law current–voltage characteristic of the form $I \approx V^2$ (which corresponds to $\beta = \frac{1}{2}$) has been observed even in a small applied voltage V.

In conclusion, we have developed an EMA for the effective nonlinear response of strongly nonlinear fractal clusters. Based on the previously derived dilute-limit expression, we construct a cluster energy functional and invoke a variational principle to obtain an approximate expression for the effective nonlinear response of a fractal cluster. The EMA results are compared with numerical calculations in a deterministic fractal model; excellent agreement is found.

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