# Strongly nonlinear response of fractal clusters

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We have developed a differential effective medium approximation (DEMA) for the effective nonlinear response due to clustering of a strongly nonlinear conducting material of a current-field (**J**-**E**) response of the form $\mathbf{J} = \chi|\mathbf{E}|^{2\beta}\mathbf{E}$ ($\beta > 0$) in a host medium, where $\chi$ is the nonlinear coefficient. The DEMA results are compared with numerical calculations in a deterministic fractal model. As a similar problem of a random medium, we further investigate the scaling behavior of the nonlinear response. It is shown that by choosing a relevant scaling variable properly, the nonlinear response function can be rescaled to collapse onto a universal curve.

## I. INTRODUCTION

The transport and optical properties of nonlinear inhomogeneous media have received much attention recently because of their potential applications in engineering and physics.[1–4] Established theories are available in the weakly nonlinear case in which the nonlinearity can be treated as a small perturbation.[5,6] Over the past few years, substantial progress has been made in calculating the effective nonlinear response of random nonlinear composites in which a small volume fraction of nonlinear material is *randomly* embedded in a host medium. An effective medium approximation[7] (EMA) for weakly nonlinear composites was proposed. Recently, the scaling form of the weakly nonlinear response has been extracted from the EMA.[8]

More recently, 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.[9] For this composite system, the inclusion and the host medium obey a local current-field (**J**-**E**) relation of the form $\mathbf{J} = \chi|\mathbf{E}|^{2\beta}\mathbf{E}$, and $\beta > 0$. For such a nonlinear relation, the conventional perturbation method[6] fails and we have recently developed a variational method to obtain the dilute-limit expressions for the effective response of a small volume fraction of spherical inclusions embedded in a host medium,[10] valid for $\beta = 1$.

Moreover, the approach is only valid for *truly* random composites in the dilute limit. In fact, many growth and fabrication processes may produce spatial correlations in realistic composites. In particular, a fractal clustering will be generated via various aggregation processes.[11–13] The fractal geometry should have an observable effect on the nonlinear as well as the linear properties.[14–16] In this work, we aim at developing a differential effective medium approximation (DEMA) for the effective nonlinear response of clustering strongly nonlinear materials in a host medium, in which case a similar approximation in weakly nonlinear systems[17] cannot be applied.

The plan of the paper is as follows. In the next section, we invoke a variational principle to obtain the dilute-limit expression for the effective nonlinear response for a small volume fraction of inclusions embedded in a host medium. Explicit asymptotic behaviors of the local field will be obtained. In Sec. III, we develop the differential effective medium approximation for the strongly nonlinear response of fractal clusters, by using the dilute-limit expression. We obtain results valid for both cases when the host is the better or poorer conductor. Then, in Sec. IV, in an attempt to verify the DEMA results, we perform a numerical simulation on a deterministic fractal cluster. In Sec. V, we propose scaling forms for the effective nonlinear response in the extreme dilute limit. The relevant scaling variables are identified and the scaling behaviors are extracted in the DEMA. Possible extensions of the present approach and relevance to recent experiments will be discussed.

## II. MODEL AND METHOD

We consider a class of strongly nonlinear composite media which obey a current-field response of the following form:[9,10]

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

where $\beta > 0$. The nonlinear coefficient $\chi$ will take on different values in the inclusion and in the host medium. An external electric field $\mathbf{E}_0$ is applied. The governing equations for electric conduction, $\nabla\cdot\mathbf{J} = 0$ and $\nabla\times\mathbf{E} = 0$, lead to the following differential equation:

$$\nabla\cdot\left[\chi(\mathbf{x})|\nabla\varphi(\mathbf{x})|^{2\beta}\nabla\varphi(\mathbf{x})\right] = 0, \tag{2}$$

where $\varphi(\mathbf{x})$ is the potential. Together with the boundary conditions on the surfaces of inclusions, Eq. (2) forms an electrostatic boundary-value problem, which cannot be solved exactly. In Ref. 10, we invoked a variational principle by minimizing the energy functional:

$$W[\varphi] = \int_V \mathbf{J}(\mathbf{x})\cdot\mathbf{E}(\mathbf{x})dV, \tag{3}$$

where the electric field is given by $\mathbf{E} = -\nabla\varphi$ and $V$ is the volume. When the minimum condition is satisfied by a potential $\tilde{\varphi}$, then by using Eqs. (1) and (3), the effective

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nonlinear response $\chi_e$ can be obtained:

$$\chi_e E_0^{2+2\beta} V = \tilde{W} = \int_V \chi(\mathbf{x}) |\tilde{\mathbf{E}}(\mathbf{x})|^{2+2\beta} dV, \tag{4}$$

where $\tilde{\mathbf{E}} = -\nabla\tilde{\varphi}$. The trial potential function will be taken as the solution of the linear problem.[10] Hence, Eq. (4) allows us to obtain the dilute-limit expression for the effective nonlinear response.

Let us consider a problem in $d$ dimensions, i.e., of spherical inclusions in three dimensions (3D) and cylindrical inclusions in two dimensions (2D) of radius $\rho$ and nonlinear coefficient $\chi_i$ suspended in a host medium of $\chi_m$, with the application of an external uniform field $\mathbf{E}_0$. We choose the following trial function for the potential:[10]

$$\varphi_i(r,\theta) = -(1-b)E_0 r\cos\theta, \qquad r < \rho, \tag{5}$$

$$\varphi_m(r,\theta) = -E_0(r - b\rho^d r^{1-d})\cos\theta, \qquad r > \rho, \tag{6}$$

where $b$ is a variational parameter as yet to be determined. With these trial functions, the energy functional is given by:[18]

$$\tilde{W}_\beta(b) = \left[\chi_m + p\chi_m Q_\beta(b) + p\chi_i(1-b)^{2+2\beta}\right] V E_0^{2+2\beta}, \tag{7}$$

where $p$ is the volume fraction of the inclusion, and $Q_\beta(b)$ is given by

$$Q_\beta(b) = \sum_{i=0}^{\infty} q_i(\beta) b^i. \tag{8}$$

Similar trial functions have been proposed in a variational treatment of weakly nonlinear composites.[19] Due to the progressively lengthy expressions of $q_i(\beta)$ for large $i$, we shall present the first few terms up to $i = 5$: $q_i(\beta) = -1$, $2(1+\beta)$, $(1+\beta)(d-1)(2+4\beta+d)/(2+d)$, $2\beta(1+\beta)(d-2)(d-1)(4+8\beta+3d)/3(2+d)(4+d)$, $\beta(1+\beta)(d-1)(-48+192\beta^2+4d+96\beta d-208\beta^2 d+32d^2-104\beta d^2+80\beta^2 d^2-17d^3+40\beta d^3+3d^4)/6(2+d)(4+d)(6+d)$, $(\beta-1)\beta(1+\beta)(d-2)(d-1)(-192+768\beta^2-8d+480\beta d-928\beta^2 d+154d^2-580\beta d^2+544\beta^2 d^2-125d^3+340\beta d^3+45d^4)/15(2+d)(4+d)(6+d)(8+d),\ldots$ for $i = 0,1,2,3,4,5,\ldots$, respectively. The results are valid for arbitrary $\beta$ and $d$. One can check from the coefficients that when $\beta$ is an integer, $q_i(\beta)$ vanishes identically for all $i > 2(1+\beta)$, and $Q_\beta(b)$ is a polynomial in $b$, while for a nonintegral $\beta$, $Q_\beta(b)$ is an infinite series of $b$. When $\beta = 1$, we recover the result for cubic nonlinearity:[10]

$$\tilde{W}_1(b) = \left[\chi_m + p\chi_m\left(-1 + 4b + \frac{2(d-1)(d+6)}{(d+2)}b^2 + \frac{4(d-1)(d-2)}{(d+2)}b^3 + \frac{(d-1)}{(d+2)}(d^2 - \tfrac{7}{3}d + 2)b^4\right) + p\chi_i(1-b)^4\right] V E_0^4, \tag{9}$$

valid for arbitrary $d$.

Let us define a contrast $z = \chi_m/\chi_i$ between the components. Minimizing Eq. (7) with respect to $b$, we obtain the following asymptotic behaviors of $b(z)$:[20]

$$b(z) = b(0) - a_0 z^{1/(1+2\beta)} + \cdots \qquad (z \ll 1)$$

$$= b(\infty) + a_\infty/z + \cdots \qquad (z \gg 1), \tag{10}$$

valid for arbitrary $\beta$ in the limits of small and large $z$. These asymptotic forms have been discussed by Bergman[21] with yet undetermined coefficients. We should remark that in Ref. 21, a self-consistent, Bruggeman-type effective medium approximation for strongly nonlinear composites is derived, which is similar to the approximation to be considered in Sec. III. By the variational method, $b(0)$ is unity for all $\beta$ and $d$; other coefficients $a_0, b(\infty), a_\infty$ can be calculated explicitly. In subsequent discussions, we shall restrict ourselves to $\beta = 1$. We find $a_0^3 = Q_1'(1)/4 = d(4+2d+3d^2)/3(2+d)$, while $a_\infty$ satisfies the equation $Q_1'(b(\infty)) = 0$ and $a_\infty = 4[1-b(\infty)]^3/Q_1''(b(\infty))$. We report numerical values as follows: In 2D, $a_0 = 1.494$, $b(\infty) = -0.4814$, and $a_\infty = 1.457$ while in 3D, $a_0 = 1.949$, $b(\infty) = -0.2954$, and $a_\infty = 0.6568$, respectively.

## III. DIFFERENTIAL EFFECTIVE MEDIUM APPROXIMATION

Here we develop the differential effective medium approximation (DEMA) for the strongly nonlinear response $\chi_e$ of fractal clusters, modifying a similar approximation for a weakly nonlinear response.[17]

We shall use the dilute-limit expression [Eq. (9)] to obtain an approximate expression for the effective nonlinear response of a cluster. 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 $\rho$, at which $p_1 = 1$ and $p_2 = 0$. The volume fraction $p_2$ of host medium is increased by adding type-2 material at the expense of volume fraction $p_1$ of type-1 materials. Let the cluster at radius $L$ have an effective response $\chi_e(L)$. According to Ref. 17, the effective response of a cluster at radius $L + \delta L$ can be obtained by adding a volume fraction $\delta\eta = -\delta p_1/p_1$ of host material to a medium with an effective response $\chi_e(L)$. Then from Eqs. (4) and (9), we find

$$\delta\chi_e = -\frac{\delta p_1}{p_1}[Q_1(b)\chi_e + (1-b)^4\chi_2], \tag{11}$$

which is an ordinary differential equation for $\chi_e(p_1)$. It is more convenient to define the dimensionless quantities

$x = \chi_2/\chi_1$ and $x_e = \chi_e/\chi_1$. Note that $b$ is the solution of the cubic equation

$$Q_1'(b)x_e - 4(1-b)^3 x = 0 \qquad (12)$$

and is a function of $x_e/x$. Upon integrating Eq. (11), we find

$$\exp\left[-\int_1^{x_e} \frac{\delta x_e}{Q_1(b)x_e + (1-b)^4 x}\right] = p_1 \equiv f, \qquad (13)$$

where we have defined $f = p_1$ as the volume fraction of the cluster; $f$ decreases from unity towards zero while $\chi_e$ varies from $\chi_1$ to $\chi_2$ as the cluster size increases. It is instructive to examine the corresponding result for the linear problem ($\beta = 0$), which can be derived in essentially the same way:

$$\exp\left[-\int_1^{x_e} \frac{\delta x_e}{Q_0(b)x_e + (1-b)^2 x}\right] = f. \qquad (14)$$

By using Eq. (8), we find $Q_0(b) = -1 + 2b + (d-1)b^2$; hence $b = (x - x_e)/[x + (d-1)x_e]$. Equation (14) can be readily integrated to give

$$\frac{\chi_e}{\chi_1}\left(\frac{\chi_2 - \chi_1}{\chi_2 - \chi_e}\right)^d = f^{-d}, \qquad (15)$$

which coincides with the linear result of a similar approximation in weakly nonlinear composites.[17] Equation (13) can usually be solved numerically; however, an asymptotic form can be obtained for small and large $x$ as we shall see below.

We should remark that the approach does *not* necessarily assume that the cluster is fractal. If, however, the cluster is indeed a fractal of fractal dimension $d_f$, then the volume fraction of nonlinear fractal inclusion is related to the cluster size as

$$f = (L/\rho)^{-(d-d_f)}. \qquad (16)$$

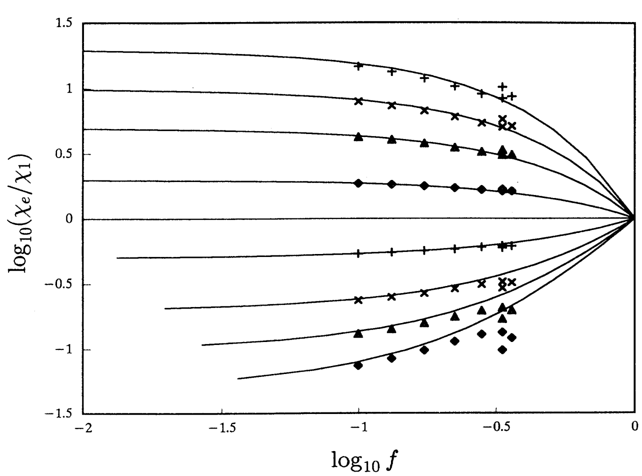

FIG. 1. Normalized effective nonlinear response $\chi_e/\chi_1$ in a DEMA (solid lines) and numerical simulations (symbols) plotted against the volume fraction $f$ for various ratios of conductivity $x$. From top and downwards in order of decreasing ratio of conductivity: $x$ = 20, 10, 5, 2, 0.5, 0.2, 0.1, and 0.05.

We are now in a position to obtain numerical results for the DEMA response. We distinguish two cases: (i) the host is the poorer conductor ($\chi_1 > \chi_2$ and $N/I$ limit) and (ii) the host is the better conductor ($\chi_1 < \chi_2$ and $S/N$ limit). We shall present results in two dimensions to compare with numerical simulations in a deterministic fractal cluster.[22] In Fig. 1, we plot the normalized effective nonlinear response $x_e/x$ as a function of volume fraction $f$ of fractal for various ratios $x$. As seen from Fig. 1, we confirm that as the cluster size increases and $f$ decreases from unity towards zero, $\chi_e$ varies from $\chi_1$ to $\chi_2$.

## IV. 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,[22] in order to compare with the DEMA results. The L-shape deterministic fractal cluster is constructed as follows. We begin with a square of lateral size of two units and divide it into four squares; the first generation is obtained when the upper right quadrant is removed. The second generation is obtained by putting together three units of the first generation. In this way, at the $n$th generation, we obtain a fractal cluster of size $L = 2^n$ embedded in the two-dimensional space. The cluster has a fractal dimension $d_f = \ln 3/\ln 2 = 1.585$ or equivalently the volume fraction of inclusion decreases with the increase of generation as $f = (3/4)^n$.[22] For convenience of numerical simulations, we then construct a fractal network by mapping the DFC onto a 2D square network — adjacent fractal squares are assigned a type-1 bond while the remaining squares assigned type-2 bonds; now the volume fraction of type-1 bonds is $f \approx (3/4)^n$ for large $n$. We associate each corresponding bond with two types of nonlinear conductors obeying a current-voltage ($I$-$V$) response of the form

$$I = \chi_i V^3, \qquad (17)$$

where $\chi_i$ ($i = 1, 2$) 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

$$I = \chi_e V^3. \qquad (18)$$

A unit voltage is applied across the top and bottom bars of the network. The nonlinear Kirchhoff's 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 DEMA [Eq. (13)] for the same $f$. The simulation is performed up to the eighth generation. In Fig. 1, we plot the normalized response $\chi_e/\chi_1$ against $f$. The simulation results are in excellent agreement with the DEMA results.

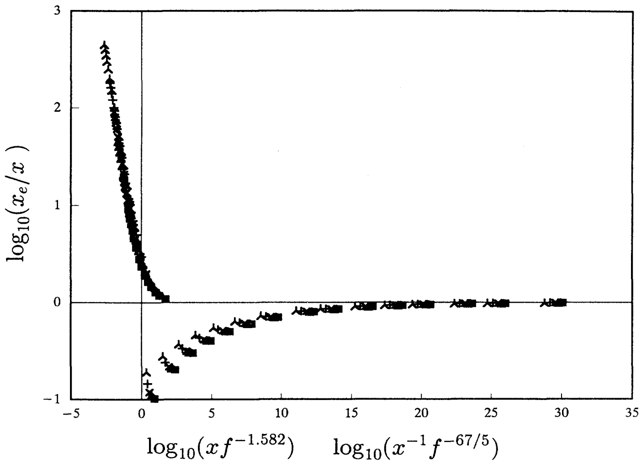


FIG. 2. Rescaled nonlinear response in a DEMA plotted against the corresponding scaling variable in 3D for the $S/N$ and $N/I$ limits. Upper curves ($N/I$ limit): $\log_{10}(x_e/x)$ is plotted against $\log_{10}(xf^{-1.582})$ for $x$ = 0.1, 0.05, 0.02, 0.01, 0.005, and 0.002. Lower curves ($S/N$ limit): $\log_{10}(x_e/x)$ is plotted against $\log_{10}(x^{-1}f^{-67/5})$ for $x$ = 500, 200, 100, 50, 20, and 10. Data collapse is evident for both limits.

## V. SCALING BEHAVIORS

As a similar problem of a random medium, we may expect the effective nonlinear response to exhibit a universal scaling behavior.[22] To this end, we propose the following scaling forms for the nonlinear response[23] in the limit of $f \ll 1$:

$$x_e = x^u \Phi(x^{-1}f^{-\phi}) \quad (x \gg 1) \tag{19}$$

$$= x^v \Psi(xf^{-\psi}) \quad (x \ll 1), \tag{20}$$

where $\Phi$ and $\Psi$ are universal scaling functions and $u$, $v$, $\phi$, and $\psi$ are exponents. By incorporating the asymptotic behaviors [Eq. (10)] into Eq. (13), we obtain $\phi = Q_1(1), \psi = Q_1(b(\infty)), u = v = 1$ in the DEMA. The numerical values are $\phi = 22/3$ and $67/5$ in 2D and 3D, respectively, while $\psi = 1.981$ and $1.582$ in 2D and 3D, respectively. It should be remarked that the exponent $\phi$ generally differs from $\psi$, indicating different scaling behaviors for the $S/N$ and $N/I$ limits. Hence a suspension of fractal clusters is not *exactly* analogous to the percolation problem (in which case the crossover exponent $\phi$ must coincide with $\psi$). We identify the relevant scaling variables $y = x^{-1}f^{-\phi}$ and $y = xf^{-\psi}$ for the $S/N$ and $N/I$ limits, respectively. To confirm the scaling form, we plot in Fig. 2 the rescaled nonlinear response $x_e/x$ as a function of the corresponding scaling variable both for the $S/N$ and $N/I$ limits; data collapse is evident for a wide range of $x$ and $f$.

## VI. DISCUSSIONS AND CONCLUSIONS

In this work, we report on several results of significance. The dilute-limit expression [Eqs. (7) and (8)] is valid for arbitrary nonlinear exponent $\beta$ and dimension $d$. With the use of its asymptotic behaviors at both small and large contrast, we should be able to develop various effective medium approximations, valid for arbitrary $\beta$ and $d$. Although the present approach deals with strongly nonlinear composites, with slight modifications, the variational method can be applied to arbitrary nonlinearity as well. However, 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[23,24] in the extremely dilute limit ($f \ll 1$), and therefore, the strongly nonlinear approach may be more applicable. Moreover, our results may have relevance to a recent experiment on laser-irradiated polymers,[25] where a power-law current-voltage characteristic of the form $I \approx V^2$ (which corresponds to $\beta = 1/2$) has been observed even in a small applied voltage $V$.

In conclusion, we have developed a DEMA for the effective nonlinear response of strongly nonlinear fractal clusters. The results are compared with numerical simulations in a deterministic fractal cluster. Very often in experimental situations, the conductivity ratio between the poor and good conducting components is finite. This leads us to examine the scaling behavior of the nonlinear response. By using the asymptotic behaviors of the field distribution, we are able to extract the exponents in the DEMA. Scaling forms for the response functions are proposed and confirmed in numerical calculations, and excellent agreements are found.

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