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LETTER TO THE EDITOR

Effective response in random mixtures of linear and nonlinear conductors

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Abstract. A simple self-consistent mean-field theory for the effective nonlinear response in random mixtures of linear and strongly nonlinear conductors is proposed. Results are in much better agreement with published simulation data than all the previously proposed approximations. The theory thus represents a far superior approximation to the Clausius–Mossotti approximation and effective-medium approximation previously proposed.

Recently, random composites consisting of two or more different kinds of nonlinear conductor have attracted much attention [1, 2]. In strongly nonlinear composites, components with J–E relations of the form $J = \chi|E|^{\beta}E$, where J is the current density and E is the local field, are considered. By suitably tuning the system parameters such as volume fraction and the nonlinear conductivities χ of the constituents and the external applied field, it is possible to control the effective nonlinear response of the nonlinear mixture. The idea of controlling the physical properties of a novel material by putting together two different materials to form an inhomogeneous medium has been repeatedly applied to different areas of condensed matter physics. Examples include semiconductor superlattices [3], linear [4] and weakly nonlinear random composites [5], magnetic granular materials in which giant magnetoresistances have been observed [6], and photonic band-gap materials in which two different dielectrics are arranged in a periodic structure [7].

Straley and Kenkel [8] studied the percolating effects in systems in which a strongly nonlinear conductor is mixed with an insulator. Using standard methods, such as scaling and real-space renormalization groups, in statistical physics, they studied the critical behaviour of the effective response near the percolation threshold. They also established the uniqueness of the solution to the problem of nonlinear response in such systems. Meir and co-workers [9] carried out similar studies using series analysis. For systems consisting of two kinds of material with the *same* nonlinearity but different conductivities, Blumenfeld and Bergman [10] developed a perturbative method, based on the difference of the conductivities, to calculate the effective response. Numerical simulations on random nonlinear resistor networks have also been performed [11]. Recently, Hui and co-workers [12] have developed a mean-field theory, which treats each component as a linear conductor with a self-consistently determined field-dependent conductivity, for the effective nonlinear response and results are in remarkable agreement with simulation data.

Levy and Bergman [13] have performed numerical simulations in systems consisting of one linear component and one strongly nonlinear component. Results are compared with

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the Clausius–Mossotti (CM) and effective-medium approximation (EMA). The comparisons between theories and simulation data are far from being satisfactory (see figures in [13]). The EMA, which works well in *linear* random composites, only captures the trend, while the CM approximation fails badly in high external fields or/and high concentrations of the nonlinear conductor. The aim of this letter is to propose a simple mean-field theory for the effective response, which gives remarkable improvement when compared with published simulation data.

Consider a binary composite consisting of a concentration p of strongly nonlinear conductor with a $\boldsymbol{J}$–$\boldsymbol{E}$ relation of the form $\boldsymbol{J} = \chi_a|\boldsymbol{E}|^\beta \boldsymbol{E}$, and concentration $1 - p$ of linear conductor with a $\boldsymbol{J}$–$\boldsymbol{E}$ relation $\boldsymbol{J} = \sigma_b \boldsymbol{E}$, where σ_b is the linear conductivity. The mean-field theory amounts to approximating the nonlinear component as conductors with the property

$$\boldsymbol{J} \approx \chi_a \langle |\boldsymbol{E}|^\beta \rangle_a \boldsymbol{E} \equiv \tilde{\sigma}_a \boldsymbol{E}, \tag{1}$$

where $\langle |\boldsymbol{E}|^\beta \rangle_a$ is an average of the local electric field taken over the volume occupied by the nonlinear component and will be determined self-consistently. In equation (1), $\tilde{\sigma}_a = \chi_a \langle |\boldsymbol{E}|^\beta \rangle_a$ and the nonlinear component is treated as a linear conductor with a field-dependent conductivity $\tilde{\sigma}_a$. The effective field-dependent conductivity σ_{eff} is defined by treating the inhomogeneous medium as a uniform system with a $\boldsymbol{J}$–$\boldsymbol{E}$ relation of the form

$$\boldsymbol{J} = \sigma_{eff}(\boldsymbol{E}_0)\boldsymbol{E}_0 \tag{2}$$

where $\boldsymbol{E}$ is the external applied field. In general, $\sigma_{eff}(\boldsymbol{E}_0)$ depends on the concentrations of the constituents and the microgeometry within the composite, as well as the external applied field.

The effective response of a binary system consisting of materials with conductivities $\tilde{\sigma}_a$ and σ_b can be calculated using standard approximations [2], such as the Maxwell–Garnett approximation (MGA) and the effective-medium approximation (EMA), developed in linear random composites. Within EMA in two dimensions, σ_{eff} is given by [2]

$$\sigma_{eff} = \tfrac{1}{2}(1-2p)(\sigma_b - \chi_a\langle |\boldsymbol{E}|^\beta \rangle_a) + \tfrac{1}{2}\sqrt{(1-2p)^2(\sigma_b - \chi_a\langle |\boldsymbol{E}|^\beta \rangle_a)^2 + 4\sigma_b\chi_a\langle |\boldsymbol{E}|^\beta \rangle_a}. \tag{3}$$

To determine the averaged local field $\langle |\boldsymbol{E}|^\beta \rangle_a$ self-consistently, we invoke a decoupling scheme developed in weakly nonlinear composites [14, 15, 16]:

$$\langle |\boldsymbol{E}|^\beta \rangle_a \approx (\langle |\boldsymbol{E}|^2 \rangle_a)^{\beta/2} = \frac{1}{p^{\beta/2}} \left(\frac{\partial \sigma_{eff}}{\partial \tilde{\sigma}_a} \right)^{\beta/2} E_0^\beta. \tag{4}$$

The last equality follows from an established result in linear random composites giving σ_{eff} in terms of the local fields:

$$\sigma_{eff} = \frac{1}{VE_0^2} \int_v \sigma(\boldsymbol{x}) |\boldsymbol{E}(\boldsymbol{x})|^2 \, \mathrm{d}^3 x \tag{5}$$

where $\sigma(\boldsymbol{x})$ takes on the value $\tilde{\sigma}_a$ (σ_b) for $\boldsymbol{x}$ in regions occupied by the nonlinear (linear) component and V is the size of the composite. Working out the derivative using equation (3) for σ_{eff}, $\langle |\boldsymbol{E}|^2 \rangle_a$ can then be determined by solving the self-consistent equation:

$$\langle |\boldsymbol{E}|^2 \rangle_a = \frac{E_0^2}{2p} \left\{ -(1-2p) + \frac{2\sigma_b - (1-2p)^2(\sigma_b - \chi_a(\langle |\boldsymbol{E}|^2 \rangle_a)^{\beta/2}}{\sqrt{(1-2p)^2(\sigma_b - \chi_a(\langle |\boldsymbol{E}|^2 \rangle_a)^{\beta/2})^2 + 4\sigma_b\chi_a(\langle |\boldsymbol{E}|^2 \rangle_a)^{\beta/2}}} \right\}. \tag{6}$$

For given σ_b, χ_a and β, equation (6) can be solved, at least numerically, for $\langle |\boldsymbol{E}|^2 \rangle_a$ as a function of E_0 and the concentration p. Substituting the result back into equation (3), σ_{eff}

can be obtained as a function of p and E_0. Equations (3), (4) and (6) thus provide a simple self-consistent scheme for estimating the effective nonlinear response in composites of one linear and one nonlinear conductor.

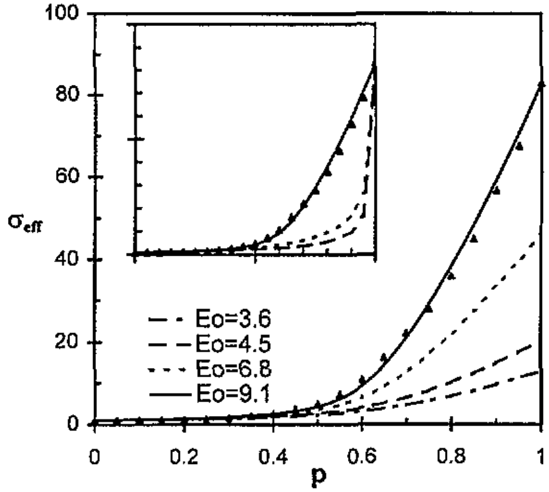

Figure 1. The effective nonlinear response σ_{eff} in the case of cubic nonlinearity is plotted as a function of the concentration of the nonlinear component for four different values of E_0 in the high-field regime. The parameters are chosen to be $\chi_a = 1$ and $\sigma_b = 1$ and are identical to those in [13]. The triangles are simulation data for the case $E_0 = 9.1$ taken from [13] and are reproduced here for comparison. The inset shows a comparison among the simulation data (triangles), results obtained from CM (dashed line) and EMA (dotted line) expressions given by [13], and the results of the present theory (solid line), for $E_0 = 9.1$.

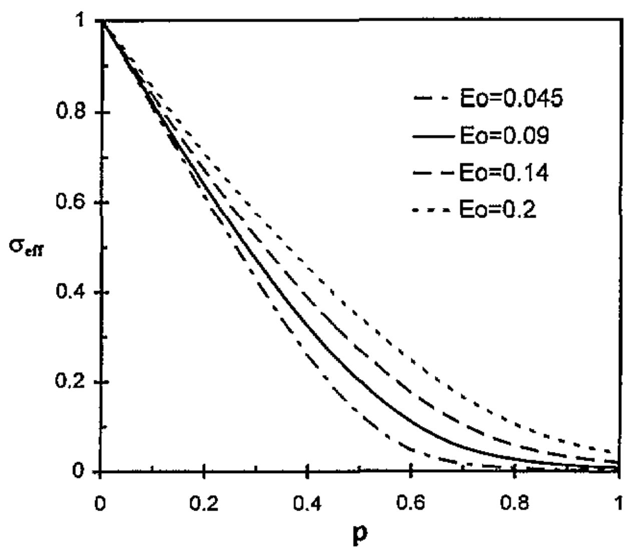

Figure 2. The effective nonlinear response σ_{eff} in the case of cubic nonlinearity is plotted as a function of the concentration of the nonlinear component for four different values of E_0 in the low-field regime. The parameters are the same as in figure 1.

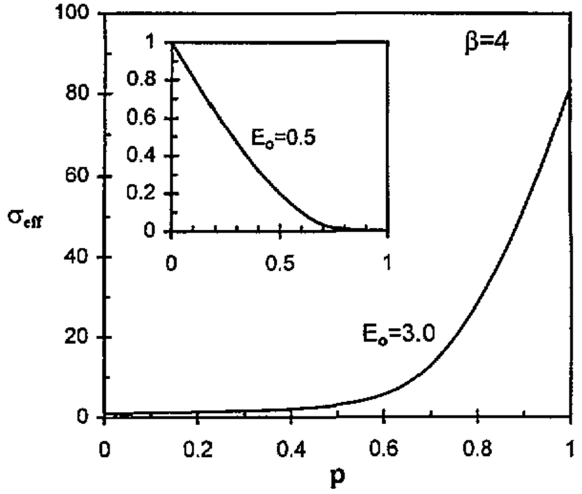

Figure 3. The effective nonlinear response σ_{eff} is plotted as a function of the concentration of the nonlinear component for the case of $\beta = 4$. The value $E_0 = 3.0$ corresponds to a high-field case and the inset with $E_0 = 0.5$ corresponds to a low-field case.

Tò compare with published simulation data, we performed model calculations based on our theory with parameters used by Levy and Bergman in their simulations [13] in two-dimensional random nonlinear resistor networks. The conductivities are chosen to be $\chi_a = 1$ and $\sigma_b = 1$, and cubic nonlinearity ($\beta = 2$) is assumed. In this way, the nonlinear component is the poorer (better) conductor between the two components in low (high) fields. Thus by tuning the field, the roles of the conductors can be interchanged. Their proposed theories fail in the limit of large contrast between the two components. Figure 1 shows the effective conductivity σ_{eff} as a function of p for four values of E_0 in the high-field regime. The simulation data for $E_0 = 9.1$, which represents the case of largest contrast and hence the worse agreement between previous theories and simulation data, are taken from [13] and reproduced here for comparison. Our results, when compared to the figures in [13], not only reproduce the trend of the data but also give values of σ_{eff} remarkably close to the simulation data for this case of highest contrast. For comparison with the approximations proposed in [13], the results obtains from the CM and EMA expressions given in [13] are plotted in the inset, together with simulation data and results of the present theory. The rapid increase in σ_{eff} near $p = 0.5$ is a percolating effect in which the better conductor, the nonlinear component in this case, forms a connected path across the system. Similar agreements are obtained for the other values of E_0, though not as surprising as for the highest-contrast case shown in the figure. Our theory thus represents a major improvement over the existing theories. This should be contrasted with the comparison between theories and simulations done in [13] in which the theories only reproduce the trend of the data qualitatively. The success of the present theory can be attributed to the more accurately determined local field within our self-consistency scheme.

Figure 2 shows results of similar model calculations in the low-field regime, again for four different values of E_0. When compared with published data (see figure 6 of [13]), the results give much better agreement than the other theories. The decrease in σ_{eff} as p increases is expected as the nonlinear conductor is the poorer conductor in this case. Through the decoupling scheme introduced in equation (4), we can carry out model calculations for arbitrary nonlinearity. Figure 3 shows typical results for the case of $\beta = 4$ for E_0 in both the high- and low-field (see inset) regimes. Levy and Bergman have also carried out some simulations for this case and our results describe their data reasonably well.

In summary, we have introduced a simple self-consistent mean-field theory for calculating the effective conductivity of random composites consisting of a linear component and a strongly nonlinear conductor with arbitrary nonlinearity. The theory represents a major improvement of the other existing theories such as the Clausius–Mossotti and effective-medium approximations. Our present theory can be readily generalized to systems with any spatial dimensions. It can also be applied to composites in which the constituents satisfy a general J–E relation of the form $J = \sigma E + \chi|E|^{\beta}E$, where both weak and strong nonlinearities are envisaged and β is arbitrary [17].

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