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1995 J. Phys.: Condens. Matter 7 8785

(http://iopscience.iop.org/0953-8984/7/46/008)

J. Phys.: Condens. Matter 7 (1995) 8785–8790. Printed in the UK

Effective-medium theory for strongly nonlinear composites: improved formalism

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Received 12 June 1995, in final form 22 August 1995

Abstract. In this work, we have attempted to improve an effective-medium approximation previously developed by us to study the effective response of a class of strongly nonlinear composite media, which obey a current–field ($\boldsymbol{J}$–$\boldsymbol{E}$) relation of the form $\boldsymbol{J} = \chi|\boldsymbol{E}|^2\boldsymbol{E}$. The improved calculations are compared with numerical simulations and with the recently established bounds for strongly nonlinear composites. We find very good improvement over the previous work especially at high contrast between the components.

1. Introduction

Recently, attention has been concentrated on a class of strongly nonlinear composite media in which the nonlinear behaviour occurs when a strong electric field is applied to condensed matter. The study of nonlinear composite media has potential applications in engineering and physics [1–4]. In this work, we study a class of nonlinear conducting composite media in which the inclusion and the host medium obey a local current–field ($\boldsymbol{J}$–$\boldsymbol{E}$) relation of the form $\boldsymbol{J} = \chi|\boldsymbol{E}|^2\boldsymbol{E}$. The effective response of strongly nonlinear composite media is extremely difficult to calculate. As the nonlinearity appears as the leading form of the behaviour rather than as a small correction to a predominant linear response, the conventional perturbation method [5, 6] fails. Nevertheless, substantial progress has been made with the aid of various approximate analytic and numerical methods over the past few years [7–11].

Blumenfeld and Bergman [7] developed a small-contrast expansion for the effective dielectric response of strongly nonlinear composites. Ponte Castaneda [8] proposed a dual variational method for establishing optimal bounds and estimates for the effective response of nonlinear composites. Recently Yu and Gu [9] adopted a simple variational approach for obtaining explicit dilute-limit expressions for a small concentration of spherical inclusions embedded in a host medium. In an attempt to extend the validity of the dilute-limit expressions to larger volume fractions, Lee and Yu [10] re-examined a self-consistent Bruggeman-type effective-medium approximation (EMA) for strongly nonlinear composites. The EMA results were compared with numerical simulations in nonlinear conductance networks with reasonable agreement at low contrast. More recently, the scaling form of the strongly nonlinear EMA was extracted near the percolation threshold [11].

The object of the present investigation is twofold. First, it is instructive to improve the EMA developed by Lee and Yu [10] for strongly nonlinear composite materials by

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adding more parameters to the trial functions. Second, it is instructive to compare the improved calculations with the recently established bounds and estimates [8] and with numerical simulations [10]. The paper is organized as follows. In the next section, we shall reinvestigate a simple one-parameter EMA for strongly nonlinear composites, to establish notation. In section 3, we develop an improved formalism for the EMA. In section 4, results obtained with the improved EMA will be compared with numerical simulation data published in the literature [10], and with the recently proposed Hashin–Shtrikman bound for strongly nonlinear composites [12].

2. Effective-medium approximation for strongly nonlinear composites

We consider strongly nonlinear composite media which obey a current–field response of the form

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

The nonlinear coefficient χ takes on a value χ_i in the inclusion, present at volume fraction p, and χ_m in the host medium, present at volume fraction $1-p$. We shall restrict ourselves to two dimensions, i.e. the inclusions are long cylinders. In order to establish notation, we reinvestigate a fully self-consistent, Bruggeman-type EMA [10] by considering a cylindrical inclusion of radius a and nonlinear coefficient χ_α ($\alpha = m, i$) embedded in an effective homogeneous medium with nonlinear coefficient χ_e. The volume Ω of the effective medium is much larger than that of the inclusion. If a uniform far field E_0 is applied, the local field is given by E_α for the type-α inclusion. The EMA requires that the *volume average* of the local electric field coincides with the uniform applied field [10], or

$$\langle E_\alpha \rangle = E_0. \tag{2}$$

Hence the problem reduces to finding the local electric field in the inclusion by solving the electrostatic boundary-value problems of strongly nonlinear media. The governing equations for electric conduction $\nabla \cdot J = 0$ and $\nabla \times E = 0$ lead to the following differential equation:

$$\nabla \cdot [\chi(x)|\nabla\varphi(x)|^2 \nabla\varphi(x)] = 0 \tag{3}$$

where $\varphi(x)$ is the potential. Together with the boundary conditions for the continuity of φ and J on the surfaces of inclusions, equation (3) forms an electrostatic boundary-value problem, which cannot be solved exactly for E_α. To find the best approximate solution for E_α, we resort to using a variational principle by minimizing the energy functional [9]:

$$W_\alpha[\varphi] = \int_V J(x) \cdot E(x)\, \mathrm{d}V \tag{4}$$

with respect to an arbitrary $\delta\varphi(x)$ away from the solution of equation (3) provided that $\delta\varphi$ vanishes at the surfaces of inclusions. The electric field is then given by $E = \nabla\varphi$.

In [9], we invoked a one-parameter variational method and obtained a crude estimate of E_α. In order to improve the variational solution, we may include more parameters in the trial potential functions; we believe that trial potential functions with more parameters may generally be better than a simple one-parameter trial function [9, 10]. To this end, Yu and Gu [13] recently discussed the proper choice of trial functions and improved the approach by including more variational parameters. It was also shown that the proposed variational field distribution converges quite well when fifteen coefficients are included in the fields [13]. The improved formalism will be applied to the EMA in the next section.

3. Improved effective-medium approximation

The simple EMA is able to give a reasonably good description for the effective nonlinear response for moderate contrast, except for where the volume fraction is near the percolation threshold [9]. We attempt to improve the variational solution of electrostatic boundary-value problems of strongly nonlinear media. We choose trial functions with a total of 18 coefficients for the potential inside the inclusion (α) and the effective medium (e):

$$\begin{aligned}\varphi_\alpha(r,\theta) &= (c_{11}^\alpha r + c_{13}^\alpha a^{-2}r^3 + c_{15}^\alpha a^{-4}r^5)\cos\theta + (c_{31}^\alpha r + c_{33}^\alpha a^{-2}r^3 + c_{35}^\alpha a^{-4}r^5)\cos 3\theta \\ &\quad + (c_{51}^\alpha r + c_{53}^\alpha a^{-2}r^3 + c_{55}^\alpha a^{-4}r^5)\cos 5\theta + \cdots \qquad r < a \end{aligned} \tag{5}$$

$$\begin{aligned}\varphi_e(r,\theta) &= r\cos\theta + (b_{11}^\alpha a^2 r^{-1} + b_{13}^\alpha a^4 r^{-3} + b_{15}^\alpha a^6 r^{-5})\cos\theta \\ &\quad + (b_{31}^\alpha a^2 r^{-1} + b_{33}^\alpha a^4 r^{-3} + b_{35}^\alpha a^6 r^{-5})\cos 3\theta \\ &\quad + (b_{51}^\alpha a^2 r^{-1} + b_{53}^\alpha a^4 r^{-3} + b_{55}^\alpha a^6 r^{-5})\cos 5\theta + \cdots \qquad r > a. \end{aligned} \tag{6}$$

We have set $E_0 = 1$ for convenience. Hence we write the trial potential functions in the inclusion and host regions as infinite sums of functions that can be separated into radial and azimuthal parts, to which we usually refer as the mode functions. These functions have been chosen so as to satisfy the symmetry and the boundary conditions—for boundary-value problems with cylindrical symmetry, they are trigonometric functions [13]. We should remark that it is not the case that each mode of (5) and (6) solves the electrostatic boundary-value problem of strongly nonlinear media—in principle, one has to calculate the entire infinite series. It is hoped that the contribution from higher-order modes is not as important as that from the few lowest modes. This can be confirmed only in actual calculations.

By using the boundary condition for φ at $r = a$, we find three relations among the 18 coefficients:

$$\begin{aligned} c_{11}^\alpha + c_{13}^\alpha + c_{15}^\alpha &= 1 + b_{11}^\alpha + b_{13}^\alpha + b_{15}^\alpha \\ c_{31}^\alpha + c_{33}^\alpha + c_{35}^\alpha &= b_{31}^\alpha + b_{33}^\alpha + b_{35}^\alpha \\ c_{51}^\alpha + c_{53}^\alpha + c_{55}^\alpha &= b_{51}^\alpha + b_{53}^\alpha + b_{55}^\alpha. \end{aligned}$$

This reduces the problem to 15 independent variational parameters for the type-α inclusion. With this choice of trial function, the energy functional in equation (4) is given by

$$\begin{aligned} W_\alpha &= \chi_e + p'\chi_e\left[-1 - 4b_{11}^\alpha + 4(b_{11}^\alpha)^2 + \frac{1}{3}(b_{11}^\alpha)^4 + \cdots\right] \\ &\quad + p'\chi_\alpha\left[(c_{11}^\alpha)^4 + 4(c_{11}^\alpha)^3 c_{13}^\alpha + \frac{28}{3}(c_{11}^\alpha)^2(c_{13}^\alpha)^2 + \cdots\right] \end{aligned} \tag{7}$$

where $p' = \pi a^2/\Omega$ is the volume fraction of the type-α inclusion. Minimizing equation (7) with respect to the variational parameters gives the following equations:

$$\begin{aligned} &\frac{\partial W_\alpha}{\partial b_{11}^\alpha} = 0 \qquad \frac{\partial W_\alpha}{\partial b_{13}^\alpha} = 0 \qquad \frac{\partial W_\alpha}{\partial b_{15}^\alpha} = 0 \qquad \cdots \\ &\qquad \frac{\partial W_\alpha}{\partial c_{11}^\alpha} = 0 \qquad \frac{\partial W_\alpha}{\partial c_{13}^\alpha} = 0 \qquad \frac{\partial W_\alpha}{\partial c_{31}^\alpha} = 0 \qquad \cdots. \end{aligned} \tag{8}$$

Hence, a system of 15 simultaneous equations has to be solved for the type-α inclusion. The nonlinear equations can be solved to give the coefficients as functions of χ_e. By examining the local-field distribution in the inclusions, we find that dominant contributions arise from coefficients c_{11}, $b_{11}, b_{13}, b_{31}, b_{33}, b_{53}$ and b_{55}. Other coefficients are relatively small in magnitude. In fact, the inclusion of just two parameters b_{11} and b_{13} already gives roughly the same results [13].

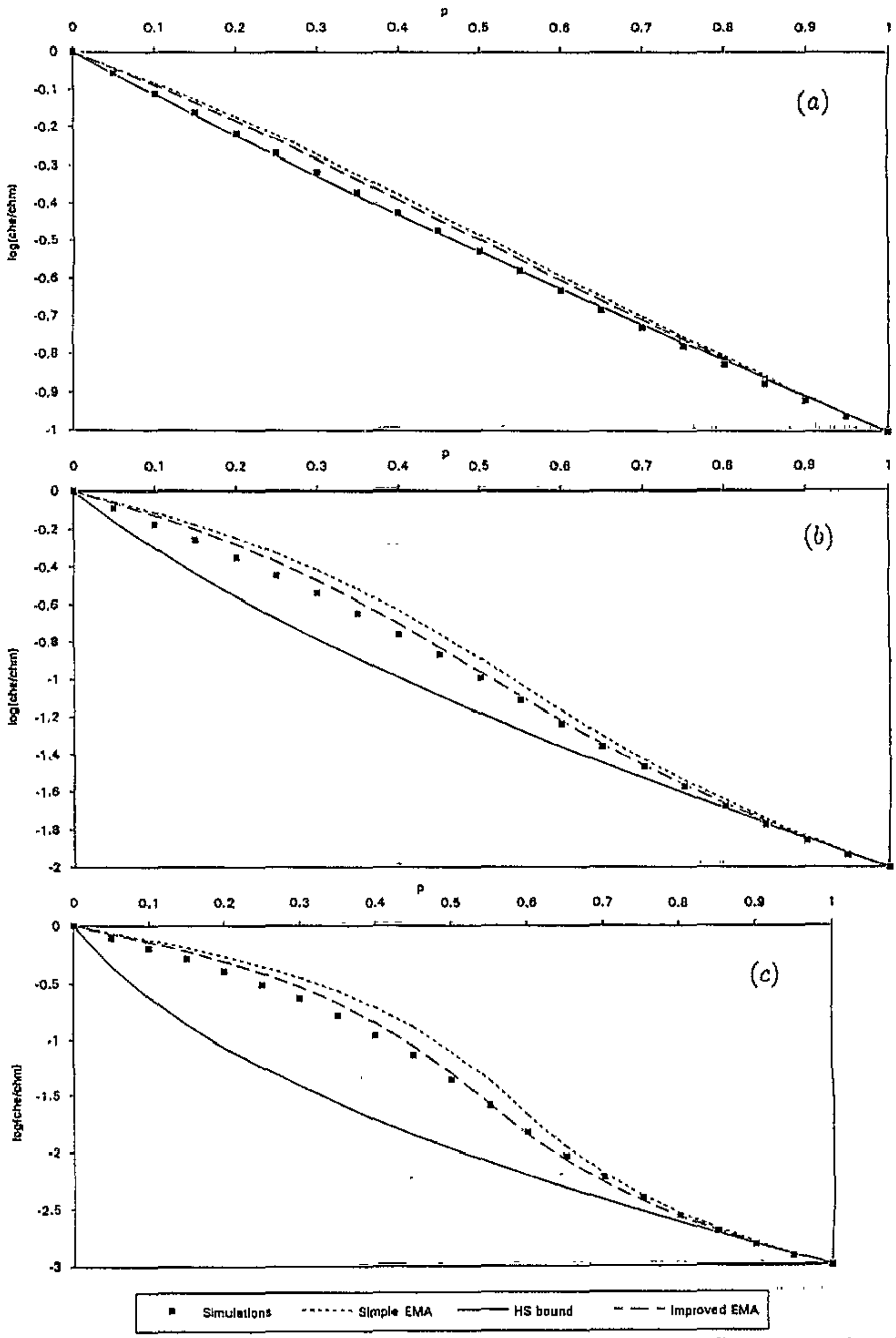

Figure 1. A semilogarithmic plot of the normalized effective nonlinear response $\log_{10}(\chi_e/\chi_m)$ against the volume fraction p for varying contrast between the components: (*a*) $\chi_i/\chi_m = 0.1$; (*b*) $\chi_i/\chi_m = 0.01$; and (*c*) $\chi_i/\chi_m = 0.001$. From the bottom upwards, we show results from: the HS bound study (solid lines); numerical simulations (symbols); the present work (long-dashed lines); and previous work [10] (short-dashed lines).

Let us consider a strongly nonlinear composite medium in which inclusions of nonlinear coefficient χ_i at a volume fraction p are randomly embedded in a host medium of coefficient χ_m at volume fraction $1-p$. The self-consistency condition (equation (2)) requires that

$$p(c_{11}^i + c_{13}^i + c_{15}^i) + (1-p)(c_{11}^m + c_{13}^m + c_{15}^m) = 1. \tag{9}$$

The right-hand side denotes the magnitude of the applied electric field ($E_0 = 1$) along the x direction. The effective response χ_e can be calculated numerically by solving (8) and (9). In figure 1, we plot the normalized effective nonlinear response χ_e/χ_m against the volume fraction p for varying contrast between the components: (a) $\chi_i/\chi_m = 0.1$; (b) $\chi_i/\chi_m = 0.01$; and (c) $\chi_i/\chi_m = 0.001$. The results of the present work will be compared with those of the Hashin–Shtrikman bound method [12] and with numerical simulations [10] in the next section.

4. Comparisons with numerical simulations and the Hashin–Shtrikman bound method

Numerical simulations were performed on two-dimensional nonlinear conductance networks as described in [10], with the network size increased to 30×30. It is instructive to compare the results with existing approximations. In analogy to the Hashin–Shtrikman (HS) bounds [12] for linear composites, Ponte Castaneda [8] proposed the following bound for strongly nonlinear composites:

$$\tilde{\chi}_e \geqslant \min_{\omega} \{p\chi_i S_i^2 + (1-p)\chi_m S_m^2\} \tag{10}$$

where ω is a variational parameter. If $\chi_i > \chi_m$, then

$$S_i = [1-(1-p)\omega]^2 \qquad S_m = (1+p\omega)^2 + p\omega^2$$

while if $\chi_i < \chi_m$, then

$$S_i = [1-(1-p)\omega]^2 + (1-p)\omega^2 \qquad S_m = (1+p\omega)^2.$$

In figure 1, we also plot the results from the simple EMA investigation [10], numerical simulations [10] and the HS lower-bound method against p for varying contrast χ_i/χ_m. We find from the plots that the improved calculations are in much better agreement with numerical simulations than the simple EMA results and the HS bound results are always below those obtained using the other approaches. We find very good improvement over the previous work, especially for high contrast between the components. We should remark that the HS bound is only a rigorous lower bound, rather than an exact result [8].

In conclusion, we have improved the recently developed EMA [10] in order to study the effective response of a class of strongly nonlinear composite media. The improved formalism is much better than the simple EMA approach when compared with numerical simulations, especially at high contrast χ_i/χ_m between the components. By using the improved formalism, we should be able to obtain a better estimate of the percolation threshold, p_c, and improved scaling laws near the percolation threshold [11].

Acknowledgments

The work was supported by the Research Grants Council of the Hong Kong Government under project numbers CUHK 78/93E and 461/95P. GQG acknowledges the support from the Chinese National Science Foundation, grant number 19374040.

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