Self-consistent effective-medium approximation for strongly nonlinear media

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A self-consistent effective-medium theory is proposed for random dielectric composites of arbitrary nonlinear constitutive law. It is based on a Gaussian approximation for the probability distributions of the electric field in each component, and on second-order Taylor expansions with an integral remainder of the local energies. The effective energy is exact to second order in contrast. With power-law media with constitutive relation $\mathbf{D}=\chi E^{\gamma-1}\mathbf{E}$, the critical exponents are $s=t=(\gamma+1)/2$. The theory reduces to Bruggeman's in the linear case $\gamma=1$, and its percolation threshold is independent of the nonlinearity.

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I. INTRODUCTION

Estimating the effective properties of nonlinear dielectric composites is a problem which has attracted wide attention in the past decades.[1–22] However, to deal with highly disordered, contrasted, and strongly nonlinear composites, a self-consistent theory extending the Bruggeman-Landauer effective-medium approach[23,24] to the nonlinear domain is still lacking. The same situation prevails in the mechanics of continuous media.[25] Indeed, none of the existing theories has passed all of the necessary tests: (i) exactness to second order in a weak-contrast perturbative expansion, (ii) existence of a nonlinearity-independent percolation threshold,[14] and (iii) the fulfillment of various bounds.

Recently theories have been constructed which obey criterion (i).[8,17,21,22,26] As to criterion (ii), only two theories leave the threshold unchanged, and reduce to the Bruggeman theory in the linear case: the first one is the self-consistent version[9] of the variational theory of Ponte Castañeda, deBotton, and Li[10] (PC), rediscovered under a different form by Wan and co-workers[14,15] (WLHY); the second one is the "second-order" approach of Ponte Castañeda and Kailasam[17](PCK).

The equivalence between the PC and WLHY approach was demonstrated by Yu, Hui, and Lee[15] on a particular example, and has since been established in full generality:[20] this theory in fact constitutes an exact lower bound in the class of self-consistent theories reducing to Bruggeman's in the linear case.[9,10,20] We shall refer to it as the PC-WLHY theory hereafter. It is interesting to note that the PC and WLHY theories first appeared in the context of nonlinear elasticity, under the name "variational"[27] and "secant" approaches,[28] their equivalence having been proven there as well.[28]

Both the PC-WLHY and PCK procedures aim at finding the "best" linear approximation to the nonlinear theory, but are somewhat orthogonal to each other: Whereas PCK use a nonlinear self-consistent closure condition on the average of the electric field in each component of the medium, PC-WLHY utilize a condition on the second moment of the field,[4,29] both these quantities being derived from the underlying linear effective-medium scheme. None of these approaches is completely satisfactory. The PC-WLHY result possesses an incorrect weak-contrast expansion. On the other hand, the PCK theory is exact to second order in the contrast. Unfortunately, it also incorrectly gives a nonlinear susceptibility lower than that of PC-WLHY, in the vicinity of the percolation threshold, as a consequence of possessing inadequate critical exponents.[30]

The purpose of this study is to partially reconcile both approaches. The theory presented below relies on simultaneous closure conditions on the average *and* the second moment of the field, thanks to a Gaussian approximation for the probability distribution of the electric field in each phase.[21] In this way, it optimizes the use of the linear underlying theory. In order to achieve this goal, a linearization based on the Taylor formula with integral remainder is adopted. This ensures the correct second-order expansion in the contrast (the PCK approach was based on a Taylor expansion with a Lagrange remainder), and the use of the second moment in the closure condition guarantees reasonable exponents, those of the PC-WLHY theory. This paper extends to strongly nonlinear media a previous work dealing with weakly nonlinear composites,[21] and focuses on the static properties only.

Sections II and III are devoted to setting up our notations and to presenting an integral formalism for the nonlinear theory which mimics that of a linear theory with spontaneous polarization. The theory of linear homogeneization in anisotropic media with spontaneous polarization, on which this study heavily relies, is then explained (Sec. IV). The material concerning the fluctuations in this theory is relegated to the Appendix for the sake of clarity. The theories of PCK and PC-WLHY are next reviewed in this framework (Sec. V). We adopt the WLHY presentation of the PC-WLHY approach, simpler to explain. Their combination is carried out in Sec. VI. The outcome of this new theory is discussed in Sec. VII, before we conclude in Sec. VIII.

II. GENERAL FRAMEWORK

We consider a nonlinear d-dimensional inhomogeneous dielectric medium of volume V characterized by a local electrostatic energy density $w_{\mathbf{x}}(\mathbf{E})$ depending on the electric field $\mathbf{E}$ at $\mathbf{x}$. From this function derives the local constitutive law for the electric displacement $\mathbf{D}$ (Refs. 31,9)

$$\mathbf{D}=\partial_{\mathbf{E}} w_{\mathbf{x}}(\mathbf{E}). \tag{1}$$

where $\partial_{\mathbf{E}}$ stands for $\partial/\partial\mathbf{E}$. When $w_{\mathbf{x}}$ is a quadratic polynomial, the constitutive relation is linear. Here $w_{\mathbf{x}}$ is not necessarily quadratic. In a composite medium, the function $w_{\mathbf{x}}$ is the same in regions V_α (called "phases") of V, ($\alpha=1,\ldots,m$), and takes the form $w_{\mathbf{x}}=w_\alpha$ if $x\in V_\alpha$. The phase volume fractions $p_\alpha=V_\alpha/V$ are such that $\Sigma_\alpha p_\alpha=1$.

Let $\overline{f(\mathbf{x})}=\int_V d^d\mathbf{x} f(\mathbf{x})/V$ stand for volume averages. The homogeneization problem consists in computing the macroscopic energy density of the system defined by

$$W(\mathbf{E}^0)=\overline{w_{\mathbf{x}}[\mathbf{E}(\mathbf{x})]}, \tag{2}$$

where the field $\mathbf{E}$ is the solution of the electrostatic equations $\boldsymbol{\nabla}\cdot\mathbf{D}=0$ and $\mathbf{E}=-\boldsymbol{\nabla}\phi$ completed by Eq. (1) and a boundary condition for the electrostatic potential $\phi=-\mathbf{E}_0\cdot\mathbf{x}$ for $\mathbf{x}\in\partial V$. Setting $\mathbf{D}^0=\overline{\mathbf{D}}$, this implies $\overline{\mathbf{E}}=\mathbf{E}^0$ and $\mathbf{E}^0\cdot\mathbf{D}^0=\overline{\mathbf{E}\cdot\mathbf{D}}$. The electric field $\mathbf{E}^0$ is a macroscopic state variable, and one shows that the effective constitutive law is

$$\mathbf{D}^0=\partial_{\mathbf{E}^0}W(\mathbf{E}^0). \tag{3}$$

For simplicity we deal hereafter with *site-disordered* systems where p_α also is the probability that $w_{\mathbf{x}}=w_\alpha$ at the point $\mathbf{x}$ ("site"), and where the material properties are uncorrelated from point to point. To each point is associated a surrounding "infinitesimal" matter element of volume v in which the potential is constant. In the thermodynamic limit $V\to\infty$ the energy is self-averaging so that volume averages and statistical averages over configurations denoted by $\langle\cdots\rangle$ coincide.

III. INTEGRAL FORMALISM

We develop here an integral formulation for the nonlinear electrostatic problem along the lines laid down by Stroud[32] in the linear case. This will illustrate the fact that the nonlinear problem has close formal bearings to the anisotropic linear problem with spontaneous polarization.[17] In this section, useful quantities are introduced for further use.

We start from the remark that the nonlinear electrostatic problem is a locally anisotropic problem where the anisotropy depends on the electric field. With $\mathbf{E}=-\boldsymbol{\nabla}\phi$ and Eq. (1) indeed, we have in any homogeneous region with energy density $w(\mathbf{E})$,

$$\boldsymbol{\nabla}\cdot\mathbf{D}=-\varepsilon:\boldsymbol{\nabla}\boldsymbol{\nabla}\phi=0, \tag{4}$$

where the *tangent permittivity tensor* is

$$\varepsilon(\mathbf{E})=\partial_{\mathbf{E}}\partial_{\mathbf{E}}w(\mathbf{E}). \tag{5}$$

For isotropic media where $w(\mathbf{E})$ only depends on the squared modulus E^2, ε is a uniaxial tensor with direction $\hat{\mathbf{E}}=\mathbf{E}/E$, of the type

$$\varepsilon_{ij}=\varepsilon_\perp(\delta_{ij}-\hat{E}_i\hat{E}_j)+\varepsilon_\parallel\hat{E}_i\hat{E}_j. \tag{6}$$

It proves convenient to emphasize this induced anisotropy by formally writing the electric displacement in terms of ε, as

$$\mathbf{D}=\varepsilon\cdot\mathbf{E}+\mathbf{P}. \tag{7}$$

This equation features an effective local polarization vector defined by

$$\mathbf{P}(\mathbf{E})=\partial_{\mathbf{E}}w_{\mathbf{x}}(\mathbf{E})-\partial_{\mathbf{E}}\partial_{\mathbf{E}}w_{\mathbf{x}}(\mathbf{E})\cdot\mathbf{E}. \tag{8}$$

Introducing an arbitrary anisotropic homogeneous medium with constant anisotropic permittivity tensor ε^0[32] and setting $\Delta\varepsilon=\varepsilon-\varepsilon^0$, we rewrite the equation for the potential as

$$\frac{\partial}{\partial x_i}\varepsilon^0_{ij}\frac{\partial}{\partial x_j}\phi=-\frac{\partial}{\partial x_i}(\Delta\varepsilon_{ij}E_i+P_i). \tag{9}$$

Using the dipolar Green tensor for the electric field in the homogeneous medium

$$G_{ij}(\mathbf{r})=-\lim_{\kappa\to 0}\int_{k>\kappa}\frac{d^dk}{(2\pi)^d}\frac{k_ik_j}{k_l\varepsilon^0_{lm}k_m}e^{i\mathbf{k}\cdot\mathbf{r}} \tag{10}$$

and taking care of the "boundary conditon"

$$\overline{\mathbf{E}}=\mathbf{E}^0, \tag{11}$$

Eq. (9) is recast into

$$\mathbf{E}(\mathbf{x})=\mathbf{E}^0+\int d^dy\,G(\mathbf{x}-\mathbf{y})[\Delta\varepsilon_{ij}(\mathbf{y})E_i(\mathbf{y})+P_i(\mathbf{y})]. \tag{12}$$

The limiting prescription in Eq. (10) ensures that $\int d^dx G(\mathbf{x})=0$ so that Eq. (11) is satisfied. As is well known $G(\mathbf{r})$ possesses an infinite number of representations of the type[33]

$$G_{ij}(\mathbf{r})=g_{ij}\delta(\mathbf{r})+H_{ij}(\mathbf{r}), \tag{13}$$

where the nonlocal part H is nonzero only outside some infinitesimal exclusion volume of arbitrary shape (generalized Lorentz cavity) and where the local part $g\delta$, represents the action of polarization charges on the surface of the cavity at its center. The value of g is shape dependent. For a spherical cavity we have

$$g_{ij}=-\int\frac{d\Omega_{\hat{\mathbf{k}}}}{S_d}\frac{\hat{k}_i\hat{k}_j}{\hat{k}_l\varepsilon^0_{lm}\hat{k}_m}, \tag{14}$$

where $S_d=2\pi^{d/2}/\Gamma(d/2)$ is the area of the unit d-dimensional sphere. The integration is carried out over all the directions of the unit vector $\hat{\mathbf{k}}$. For site disorder, the cavity is identified to the microscopic matter element with volume v and the theory is tantamount to a theory of interacting point dipoles. Dealing with randomly oriented nonspherical matter elements[34] would introduce unnecessary technical complications (among which a position-dependent g), so that only spheres are considered hereafter. Expression (14) is equivalent to a more complicated one usually used in the effective-medium literature,[32,34,35] thanks to a suitable change of variables.

Extracting g out of the integral in Eq. (12) gives a multiple-scattering expression for the *local field* impinging on $\mathbf{x}$. Setting (1 represents the identity matrix)

$$\mu=[1-g\cdot\Delta\varepsilon]^{-1}, \tag{15}$$

the local field is defined by

$$\mathbf{E}^l = \mu^{-1}\cdot\mathbf{E} - g\cdot\mathbf{P} \tag{16}$$

and obeys the integral equation

$$\mathbf{E}^l = \mathbf{E}^0 + ⨍ d^d y H(\Delta\varepsilon\cdot\mu\cdot\mathbf{E}^l + \mathbf{P}\cdot\mu), \tag{17}$$

where the notation $⨍ dy$ stands for the principal value $\lim_{\eta\to 0}\int_{|\mathbf{y}-\mathbf{x}|>\eta} d^d y$. In arriving at (17), use was made of the identity

$$1 + g\cdot\Delta\varepsilon\cdot\mu = 1 + \mu\cdot g\cdot\Delta\varepsilon = \mu. \tag{18}$$

Note for further use that $\mu\cdot g$ and g are symmetric tensors, so that (superscript t denoting the transpose)

$$\mu\cdot g = g\cdot{}^t\mu. \tag{19}$$

IV. LINEAR HOMOGENEIZATION WITH SPONTANEOUS POLARIZATION

A. Effective-medium conditions

Before carrying out the homogeneization of the nonlinear problem, we first have to examine the anisotropic *linear* theory with spontaneous polarization, a particular instance to which the integral formalism of Sec. III can be applied. We thus consider here quadratic local potentials of the form

$$w_{\mathbf{x}}(\mathbf{E}) = (1/2)\varepsilon^\alpha{:}\mathbf{E}\mathbf{E} + \mathbf{P}^\alpha\cdot\mathbf{E} + c^\alpha, \quad \mathbf{x}\in V_\alpha, \tag{20}$$

where ε, $\mathbf{P}$, and the scalar c are *constants* in each phase. Equations (5) and (8) now become tautologies. The local electric displacement is

$$\mathbf{D} = \partial_{\mathbf{E}} w_\alpha(\mathbf{E}) = \varepsilon^\alpha\cdot\mathbf{E} + \mathbf{P}^\alpha, \quad \mathbf{x}\in V_\alpha. \tag{21}$$

The problem being linear, the effective energy $W(\mathbf{E}^0) = \langle w_{\mathbf{x}}(\mathbf{E})\rangle$ is sought for under the form

$$W(\mathbf{E}^0) = (1/2)\varepsilon^0{:}\mathbf{E}^0\mathbf{E}^0 + \mathbf{P}^0\cdot\mathbf{E}^0 + c^0, \tag{22}$$

whereby Eq. (3) implies that

$$\mathbf{D}^0 = \varepsilon^0\cdot\mathbf{E}^0 + \mathbf{P}^0. \tag{23}$$

The effective-medium conditions can be derived from an assumption of statistical independence between the local field $\mathbf{E}^l(\mathbf{x})$ and the material properties at the same point $\mathbf{x}$. Hence, for instance, $\langle\mu(\mathbf{x})\mathbf{E}^l(\mathbf{x})\rangle = \langle\mu\rangle\cdot\langle\mathbf{E}^l\rangle$. It is clear that this (over)simplifying assumption, which may actually be physically reasonable for a particular choice of ε^0 and $\mathbf{P}^0$, and particular microstructures (such as Milton's hierarchical composite for which Bruggeman's theory is exact,[36] and where each element is surrounded by a smaller-scale self-averaging environment which acts as a homogeneous one), is definitely inexact for nonlinear media where material properties are intrinsically field dependent.

Under the above assumption and imposing the condition (11), we find from Eqs. (16), (19)

$$\mathbf{E}^0 = \langle\mu\rangle\cdot\langle\mathbf{E}^l\rangle + \langle\mu\cdot g\cdot\mathbf{P}\rangle = \langle\mu\rangle\cdot\langle\mathbf{E}^l\rangle + g\cdot\langle\mathbf{P}\cdot\mu\rangle. \tag{24}$$

Were the medium homogeneous ($\Delta\varepsilon = 0$, $\mathbf{P} = \text{const.}$), the above relation between the field and the local field would read

$$\mathbf{E} = \mathbf{E}^l + g\cdot\mathbf{P}. \tag{25}$$

Comparing to Eq. (24) shows that the effective-medium conditions are

$$\langle\mu\rangle = 1, \tag{26}$$

$$\mathbf{P}^0 = \langle\mathbf{P}\cdot\mu\rangle \tag{27}$$

so that

$$\langle\mathbf{E}^l\rangle = \mathbf{E}^0 - g\cdot\mathbf{P}^0. \tag{28}$$

Condition (26) determines the effective permittivity tensor ε_0, and is equivalent to the equation[32]

$$\langle\Delta\varepsilon\cdot\mu\rangle = 0, \tag{29}$$

as can be seen from Eq. (18). Applied to isotropic media, it reduces to Bruggeman's condition.[23,24] As a consequence, we note that $\varepsilon^0 = \langle\varepsilon\cdot\mu\rangle$. In practice, ε_0 can in most cases be computed by iterating the equation

$$\varepsilon_0 = \langle\varepsilon\cdot\mu\rangle\cdot\langle\mu\rangle^{-1} \tag{30}$$

until convergence starting, e.g., from the upper bound approximation $\varepsilon_0 \simeq \langle\varepsilon\rangle$. Eq. (27) defines the effective macroscopic polarization $\mathbf{P}^0$.

Remark in passing the consistency of Eq. (28) with the result of a volume average of the integral equation (17): Because of the boundary constraint $\int d^d x G = 0$, we have $\int d^d x H = -g$ and Eq. (28) is recovered.

B. Macroscopic energy

An expression for the macroscopic energy density consistent with the above homogeneization procedure is obtained as follows. Starting from Eq. (20), we have, with $\mathbf{D} = \varepsilon\cdot\mathbf{E} + \mathbf{P}$,

$$\begin{aligned} W(\mathbf{E}^0) &= \langle w_{\mathbf{x}}(\mathbf{E})\rangle = (1/2)\langle\mathbf{E}\cdot\mathbf{D}\rangle + (1/2)\langle\mathbf{E}\cdot\mathbf{P}\rangle + \langle c\rangle \\ &= (1/2)\langle\mathbf{E}\rangle\cdot\langle\mathbf{D}\rangle + (1/2)\langle\mathbf{E}\cdot\mathbf{P}\rangle + \langle c\rangle, \end{aligned} \tag{31}$$

where the last equality stems from the boundary conditions (see Sec. II). Moreover,

$$\begin{aligned} \langle\mathbf{P}\cdot\mathbf{E}\rangle &= \langle\mathbf{P}\cdot\mu\cdot\mathbf{E}^l\rangle + \langle\mathbf{P}\cdot\mu\cdot g\cdot\mathbf{P}\rangle \\ &= \langle\mathbf{P}\cdot\mu\rangle\cdot\langle\mathbf{E}^l\rangle + \langle\mathbf{P}\cdot\mu\cdot g\cdot\mathbf{P}\rangle \\ &= \mathbf{P}^0\cdot\mathbf{E}^0 - \mathbf{P}^0\cdot g\cdot\mathbf{P}^0 + \langle\mathbf{P}\cdot\mu\cdot g\cdot\mathbf{P}\rangle \\ &= \mathbf{P}^0\cdot\mathbf{E}^0 + \langle\Delta\mathbf{P}\cdot\mu\cdot g\cdot\Delta\mathbf{P}\rangle, \end{aligned} \tag{32}$$

where

$$\Delta\mathbf{P} = \mathbf{P} - \mathbf{P}^0. \tag{33}$$

Combining these results with Eq. (23) finally yields an effective energy $W(\mathbf{E}^0)$ of the form (22) with $\mathbf{P}^0$ given by Eq. (27) and

$$c^0 = \langle c\rangle + \frac{1}{2}\langle\Delta\mathbf{P}\cdot\mu\cdot g\cdot\Delta\mathbf{P}\rangle. \tag{34}$$

The last term in Eq. (34) does not depend on the macroscopic electric field $\mathbf{E}^0$, and represents, within the effective-medium approximation, the interaction energy between the random spontaneous dipoles.

C. Averages and fluctuations

For a n-ary composite the *mean field in each phase* is

$$\mathbf{M}^\alpha \equiv \langle \mathbf{E} \rangle_\alpha = \frac{1}{p_\alpha} \frac{\partial W(\mathbf{E}^0)}{\partial \mathbf{P}^\alpha} = \mu^\alpha \cdot (\mathbf{E}^0 + g \cdot \Delta \mathbf{P}^\alpha), \quad (35)$$

where the notation $\langle \cdots \rangle_\alpha$ indicates an average over the electric field probability distribution in the phase α (equivalent to a volume average in the phase). We check that $\langle \mathbf{E} \rangle = \Sigma_\alpha p_\alpha \langle \mathbf{E} \rangle_\alpha \equiv \langle \mathbf{M} \rangle = \mathbf{E}^0$, and that

$$\begin{aligned}\langle \mathbf{D} \rangle &= \sum_\alpha p_\alpha \varepsilon^\alpha \cdot \langle \mathbf{E} \rangle_\alpha + \langle \mathbf{P} \rangle \\ &= \langle \varepsilon \cdot \mu \rangle \cdot \mathbf{E}^0 + \langle \varepsilon \cdot \mu \cdot g \cdot \mathbf{P} \rangle - \langle \varepsilon \cdot \mu \rangle \cdot g \cdot \langle {}^t\mu \cdot \mathbf{P} \rangle + \langle \mathbf{P} \rangle \\ &= \varepsilon^0 \cdot \mathbf{E}^0 + \langle \Delta \varepsilon \cdot \mu \cdot g \cdot \mathbf{P} \rangle + \langle \mathbf{P} \rangle \\ &= \varepsilon^0 \cdot \mathbf{E}^0 + \langle \mathbf{P} \cdot (\Delta \varepsilon \cdot \mu \cdot g + 1) \rangle \\ &= \varepsilon^0 \cdot \mathbf{E}^0 + \langle \mathbf{P} \cdot \mu \rangle = \varepsilon^0 \cdot \mathbf{E}^0 + \mathbf{P}^0 = \mathbf{D}^0, \end{aligned} \quad (36)$$

as must be. Likewise, the *covariance matrix of the electric field in each phase* reads[37]

$$C^\alpha \equiv \langle \mathbf{E}\mathbf{E} \rangle_\alpha - \langle \mathbf{E} \rangle_\alpha \langle \mathbf{E} \rangle_\alpha \quad (37a)$$

$$= \frac{2}{p_\alpha} \frac{\partial W(\mathbf{E}^0)}{\partial \varepsilon^\alpha} - \mathbf{M}^\alpha \mathbf{M}^\alpha. \quad (37b)$$

For purely isotropic media without spontaneous polarization, the expression of C is known explicitly.[21,37] Deriving an closed-form expression for C^α similar to that found for $\mathbf{M}^\alpha$, however, is a difficult task in the general case. For nonlinear homogeneization, we need to consider the case of uniaxial anisotropy where the anisotropy direction is that of the macroscopic field $\hat{\mathbf{E}}^0 = \mathbf{E}^0 / E^0$. Then any vector $\mathbf{A}$ is of the form $A_i = A \hat{E}_i^0$ and any second-order tensor A is of the form $A_{ij} = A_\parallel \hat{E}_i^0 \hat{E}_j^0 + A_\perp (\delta_{ij} - \hat{E}_i^0 \hat{E}_j^0)$. The shorthand notation $A = (A_\parallel, A_\perp)$ is used hereafter, in terms of which

$$\langle \mu_\parallel \rangle = \langle \mu_\perp \rangle = 1, \quad (38)$$

$$P^0 = \langle \mu_\parallel P \rangle. \quad (39)$$

Thus, with $\Delta P = P - P^0$

$$W(\mathbf{E}_0) = \frac{1}{2} \varepsilon_\parallel^0 E^{02} + P^0 E^0 + \frac{1}{2} g_\parallel \langle \mu_\parallel \Delta P^2 \rangle + \langle c \rangle \quad (40)$$

and

$$M^\alpha = \mu_\parallel^\alpha [E^0 + g_\parallel (P^\alpha - P^0)]. \quad (41)$$

Moreover,

$$C_\parallel^\alpha = \frac{2}{p^\alpha} \frac{\partial W}{\partial \varepsilon_\parallel^\alpha} - M^{\alpha 2}, \quad (42a)$$

$$C_\perp^\alpha = \frac{1}{d-1} \frac{2}{p^\alpha} \frac{\partial W}{\partial \varepsilon_\perp^\alpha}. \quad (42b)$$

Expression (40) allows for a straightforward computation of the derivatives in Eqs. (42). The sole difficulty consists in obtaining the variations of $\varepsilon_\parallel^0$ and $g_\parallel$ in a manageable form. The necessary results are presented in the Appendix, where explicit expressions for g in dimensions of interest are also recalled.

V. NONLINEAR THEORIES

The previous section has provided us with the material and notations needed in what follows. We now briefly review the two nonlinear effective-medium theories that this work unifies.

A. The PCK theory

The above theory for linear media has been utilized to homogenize nonlinear media in the following way.[17] The idea put forward by PCK is to start from a second-order Taylor expansion with Lagrange remainder of the microscopic nonlinear potential $w_\alpha(\mathbf{E})$, around the yet unknown phase average $\mathbf{M}^\alpha$ (35), assumed to be representative of the electric field in phase α. Thus

$$\begin{aligned} w_\alpha(\mathbf{E}) = {} & w_\alpha(\mathbf{M}^\alpha) + \partial_{\mathbf{E}} w_\alpha(\mathbf{M}^\alpha) \cdot (\mathbf{E} - \mathbf{M}^\alpha) \\ & + (1/2) \partial_{\mathbf{E}} \partial_{\mathbf{E}} w_\alpha(\tilde{\mathbf{M}}^\alpha) : (\mathbf{E} - \mathbf{M}^\alpha)(\mathbf{E} - \mathbf{M}^\alpha), \end{aligned} \quad (43)$$

where $(0 < \lambda < 1)$

$$\tilde{\mathbf{M}}^\alpha = \mathbf{M}^\alpha + \lambda (\mathbf{E} - \mathbf{M}^\alpha). \quad (44)$$

The Lagrange parameter λ depends on $\mathbf{E}$, and PCK make the further approximation $\lambda = 0$ which truncates the Taylor expansion at second order, so that finally

$$w_\alpha(\mathbf{E}) \simeq w_\alpha^{\mathrm{lin}}(\mathbf{E}) \equiv c^\alpha + \mathbf{P}^\alpha \cdot \mathbf{E} + (1/2) \varepsilon^\alpha : \mathbf{E}\mathbf{E}, \quad (45)$$

where ε^α, $\mathbf{P}^\alpha$ are defined by Eqs. (5), (8) at $\mathbf{E} = \mathbf{M}^\alpha$, and

$$c_\alpha = w_\alpha(\mathbf{M}^\alpha) - \mathbf{P}^\alpha \cdot \mathbf{M}^\alpha - (1/2) \varepsilon^\alpha : \mathbf{M}^\alpha \mathbf{M}^\alpha. \quad (46)$$

The nonlinear homogeneization procedure then consists in applying the previous linear homogeneization theory to the approximated potentials (45). The mean fields $\mathbf{M}^\alpha$ on which the constants c^α, $\mathbf{P}^\alpha$, and ε^α depend are computed self-consistently by means of Eq. (35). This last set of equations acts as closure relations for the nonlinear EMA. We refer the reader to the original paper for practical details about the resolution of these equations. We emphasize that the permittivity used here is an anisotropic tensor.

B. The PC-WLHY theory

The PC-WLHY theory stems from a completely different approach, where one only deals with scalar permittivities. It can be simply summarized as follows (showing that it constitutes a lower bound requires a more elaborate presentation[9]). An effective local *scalar* permittivity ε is defined via the relation

$$\mathbf{D}\equiv\varepsilon\mathbf{E}. \tag{47}$$

Thus, in terms of the potential w_x (assumed to depend on the modulus E only),

$$\varepsilon=\mathbf{E}\cdot\partial_{\mathbf{E}}w_x/E^2\equiv\varepsilon(E^2). \tag{48}$$

This is the so-called "secant" permittivity. WLHY assume that in the phase α, $\mathbf{D}$ can be approximated ("decoupling approximation") by $\mathbf{D}=\varepsilon^{\alpha}\mathbf{E}$, where

$$\varepsilon^{\alpha}=\langle\varepsilon(E^2)\rangle_{\alpha}\simeq\varepsilon(\langle E^2\rangle_{\alpha}^{1/2}). \tag{49}$$

This approximation therefore amounts to replacing the local potential w_α by

$$w_{\alpha}^{\text{lin}}=(1/2)\varepsilon^{\alpha}E^2. \tag{50}$$

This problem is homogenized with Bruggeman's *isotropic* EMT, which yields an effective scalar permittivity ε^0. The self-consistent closure relations now concern the second moment, obtained via a particularization of Eq. (37b) (Refs. 29,38,39)

$$\langle E^2\rangle_{\alpha}=\frac{1}{p_{\alpha}}\frac{\partial\varepsilon^0}{\partial\varepsilon_{\alpha}}E_0^2. \tag{51}$$

Solving the system (49), (51) then allows one to compute the effective nonlinear response under the form of a relation $\mathbf{D}^0=\varepsilon^0(E^0)\mathbf{E}^0$.

VI. COMBINING BOTH APPROACHES

So to speak, the previous theories are "orthogonal" to one another, and use complementary information about the moments of the electric field in each phase borrowed from the linear theory. We now present a combination of the previous approaches which utilizes both the means and the variances. It consists in approximating the probability distribution of the electric field in each phase by the Gaussian[21]

$$\mathcal{P}_{\alpha}(\mathbf{E})=\frac{1}{[(2\pi)^d\det(C^{\alpha})]^{1/2}}e^{-1/2\Delta\mathbf{E}^{\alpha}\cdot C^{\alpha-1}\cdot\Delta\mathbf{E}^{\alpha}}, \tag{52}$$

where $\Delta\mathbf{E}^{\alpha}=\mathbf{E}-\mathbf{M}^{\alpha}$. The parameters $\mathbf{M}^{\alpha}$ and C^{α} are, by definition of the Gaussian distribution, the mean and variance of the field as defined in Eqs. (35) and (37a). This approximation is discussed below, in Sec. VII E.

A property of the Gaussian distribution (easily demonstrated by integration by parts) is the following one. Let $\mathbf{X}$ be a random vector variable, distributed according to a centered (zero-mean) Gaussian law of variance the symmetric second-order tensor C. Denoting—in this paragraph only—an average with this Gaussian by $\langle\cdots\rangle$, one has for any function $f(\mathbf{X})$ the identity

$$\langle f(\mathbf{X})\mathbf{X}\rangle=C\cdot\langle\nabla f(\mathbf{X})\rangle. \tag{53}$$

From this property derives Wick's theorem (which essentially states that the average of an even moment of the variable is obtained as a sum of products of the variance tensor which exhausts the possible pairings of the indices; the average of an odd moment is zero): for instance, applying it twice to $\langle X^4\rangle$ yields

$$\langle X^4\rangle=\langle X_iX_iX_jX_j\rangle=C_{ii}C_{jj}+2C_{ij}C_{ij}. \tag{54}$$

Identity (53), however, permits one to extend Wick's decomposition beyond mere integer powers of the random vector.

This identity allows one to build a quadratic approximation to the local energy densities w_x as follows. Contrary to PCK who expand w_x around the mean field $\mathbf{M}$ by using a second-order Taylor formula with Lagrange remainder, we apply here the Taylor formula with *integral* remainder

$$f(1)=f(0)+f'(0)+\int_0^1 dt(1-t)f^{(2)}(t) \tag{55}$$

to the function

$$f(t)=w_{\alpha}(\mathbf{M}^{\alpha}+t\Delta\mathbf{E}^{\alpha}). \tag{56}$$

This yields

$$w_{\alpha}(\mathbf{E})=w_{\alpha}(\mathbf{M}^{\alpha})+\partial_i w_{\alpha}(\mathbf{M}^{\alpha})\Delta E_i^{\alpha}+\int_0^1 dt(1-t)\times\Delta E_i^{\alpha}\Delta E_j^{\alpha}\partial_{ij}^2 w_{\alpha}(\mathbf{M}^{\alpha}+t\Delta\mathbf{E}^{\alpha}). \tag{57}$$

Now, considered as a function of $\Delta\mathbf{E}^{\alpha}$, the distribution (52) is centered, and property (53) applies. Under the Gaussian approximation, averaging the last term of Eq. (57) therefore leads to

$$\begin{aligned}\int_0^1 dt(1-t)\langle\Delta E_i^{\alpha}\Delta E_j^{\alpha}\partial_{ij}^2 w_{\alpha}\rangle_{\alpha}&=C_{ik}^{\alpha}\int_0^1 dt(1-t)[\langle\partial_{ik}^2 w_{\alpha}\rangle_{\alpha}+t\langle\Delta E_j^{\alpha}\partial_{ijk}^3 w_{\alpha}\rangle_{\alpha}]\\&=C_{ik}^{\alpha}\int_0^1 dt(1-t)\left(1+t\frac{\partial}{\partial t}\right)\langle\partial_{ik}^2 w_{\alpha}\rangle_{\alpha}\\&=C_{ik}^{\alpha}\int_0^1 dt\,t\langle\partial_{ik}^2 w_{\alpha}(\mathbf{M}^{\alpha}+t\Delta\mathbf{E}^{\alpha})\rangle_{\alpha}.\end{aligned} \tag{58}$$

This expression, where we recognize a decoupling of the fluctuations, allows for a linearization of w_α. Setting

$$\varepsilon_{ij}^{\alpha}=2\int_0^1 dt\,t\langle\partial_{ij}^2 w_{\alpha}(\mathbf{M}^{\alpha}+t\Delta\mathbf{E}^{\alpha})\rangle_{\alpha}, \tag{59}$$

suggests that we approximate w_α by

$$w_\alpha^{\rm lin}(\mathbf{E}) = w_\alpha(\mathbf{M}^\alpha) + \partial_i w_\alpha(\mathbf{M}^\alpha)\Delta E_i^\alpha + \tfrac{1}{2}\,\varepsilon_{ij}^\alpha \Delta E_i \Delta E_j$$
$$\equiv c^\alpha + \mathbf{P}^\alpha \cdot \mathbf{E} + \tfrac{1}{2}\,\mathbf{E}\cdot\varepsilon^\alpha\cdot\mathbf{E}, \tag{60}$$

where c^α and $\mathbf{P}^\alpha$ are again defined by

$$\mathbf{P}^\alpha = \partial_{\mathbf{E}} w_\alpha(\mathbf{M}^\alpha) - \varepsilon^\alpha \cdot \mathbf{M}^\alpha, \tag{61a}$$

$$c_\alpha = w_\alpha(\mathbf{M}^\alpha) - \mathbf{P}^\alpha\cdot\mathbf{M}^\alpha - (1/2)\varepsilon^\alpha\!:\!\mathbf{M}^\alpha\mathbf{M}^\alpha. \tag{61b}$$

This approximation is such that both the original expressions (57) and its linearization (60) give the same result when averaged with Eq. (52), namely,

$$\langle w_\alpha\rangle_\alpha = \langle w_\alpha^{\rm lin}\rangle_\alpha = w_\alpha(\mathbf{M}^\alpha) + \tfrac{1}{2}\,C_{ij}^\alpha \varepsilon_{ij}^\alpha. \tag{62}$$

Apart from a change in the definition of ε_{ij}^α expression (60) is the same as that used in the PCK theory. Therefore, as in the PCK theory, homogeneization follows from using the linear theory of Sec. IV, the effective energy of which being used as the nonlinear effective potential. It is stressed that in Eq. (59), the permittivity tensor now directly depends on $\mathbf{M}^\alpha$ *and* the covariance tensors C^α, since the average entering ε^α is carried out by means of the Gaussian distribution (52). We now have to use *simultaneous* self-consistent closure relations on the averages $\mathbf{M}^\alpha$ and on the covariance tensors C^α. The numerical solution is obtained, e.g., by the following iterative procedure: start from values of $\mathbf{P}^\alpha$, c^α, and ε^α, obtained by using the definitions (61) and (59) with $C^\alpha = 0$ and $\mathbf{M}^\alpha = \mathbf{E}^0$; iterate Eq. (30) to compute ε^0, then compute $\mathbf{P}^0$ by means of Eq. (27); deduce new values of $\mathbf{M}^\alpha$ and C^α by means of Eq. (35) and the equations of the Appendix, and iterate until convergence (not guaranteed, but obtained in the numerical examples presented below; an implicit alternative procedure may be necessary). Then compute the coefficients c^α, the energy, and deduce the effective nonlinear response.

VII. DISCUSSION

We now discuss the salient features of the above theory, explain its residual flaws, and offer perspectives for future work.

A. General remarks

As with the PCK or PC-WLHY theory, our theory is meant to apply to a large class of nonlinearities, as long as the phase constitutive laws derive from energy potentials, and does not require any prior knowledge of the analytical form of the effective potential. In this respect, it markedly differs from that of Bergman and co-workers,[8,16] analytically approximated by Barthélémy,[22] which relies on the fact that this analytical form is known in advance—for the special case of power-law potentials with the *same* exponents—in terms of only *one* scalar unknown. The authors can then determine the effective potential by means of a single self-consistent equation, in the spirit of Bruggeman's, but the method does not seem to be easily generalizable (and moreover gives, in a yet unexplained way, a nonlinearity-dependent percolation threshold).

Trying to be more versatile, the approach advocated here insists on a self-consistent treatment of the fluctuations of the electric field. The decoupling procedure of the fluctuations used to linearize the nonlinear potential is completely transparent, and rigorous [in the sense of the first equality in Eq. (62)] as long as Gaussian distributions are used. In this context, so are the self-consistency conditions on the first two moments of the electric field.

In this theory as well as in PCK's, the effective nonlinear energy is identified with the effective energy of the linear homogeneization theory of Sec. IV. We remark that the second equality in Eq. (62) always holds *independently of Gaussian averaging* in both theories, as a mere consequence of the form (61) assumed by c^α and $\mathbf{P}^\alpha$. The estimated nonlinear effective energy can therefore alternatively be written

$$W = \sum_\alpha p_\alpha\left[w_\alpha(\mathbf{M}^\alpha) + \frac{1}{2}C_{ij}^\alpha \varepsilon_{ij}^\alpha\right] \tag{63}$$

in both theories.

We also remark that the permittivity of the PCK theory $\varepsilon_{ij}^\alpha = \partial_{ij}^2 w_\alpha(\mathbf{M}^\alpha)$, is formally recovered from the new procedure in the limit where $C^\alpha \to 0$ in Eq. (59), since the Gaussian used for the averaging then degenerates into the zero-width Dirac distribution

$$\mathcal{P}_\alpha(\mathbf{E}) = \delta^{(d)}(\mathbf{E} - \mathbf{M}^\alpha). \tag{64}$$

This comparison enlightens the way the nonzero covariance matrix enters the computation of ε^α (the PCK theory possesses a nonzero covariance matrix as well which can always be computed from the effective energy, though it does no enter the linearization procedure).

The numerical calculations presented below have been carried out for binary composites in dimension $d=2$, with power-law potentials of the type

$$w_\alpha(E) = \frac{\chi_\alpha}{\gamma+1}E^{\gamma+1}, \tag{65}$$

when γ is constant in the medium and χ_α varies in each component. The effective potential then reads $W = \chi_0 E^{0\,\gamma+1}/(\gamma+1)$. The cases $\gamma = 3,\ 5$ are examined for various contrasts. These values are used because when $\gamma = 2k-1$, the tensor ε^α [Eq. (59)] can be computed analytically in terms of M^α, $C_\parallel^\alpha$, and $C_\perp^\alpha$ through a finite number of repeated applications of identity (53). Such an expansion, however, becomes rapidly unmanageable as γ increases. For nonodd exponent values, the vector Gaussian integrals entering the definition of ε^α have to be computed numerically, which complicates the task.[40]

B. Critical regime

When the dielectric contrast between the components is infinite, the binary composite displays percolative behavior.[41,42] The percolation threshold of the present theory is $p_c = 1/d$ (p denoting the volume fraction of the high susceptibility component). This value is that of the Bruggeman theory, and is shared by the PCK and the PC-WLHY theories as well. Indeed, the anisotropy of the phases is irrelevant to the value of threshold—a geometric quantity—and the linear theory is consistent with this observation.[43,44] Asymptotic expansions of the linear theory in the vicinity of p_c allow to compute the critical exponents in the nonlinear theory for general power-law potentials (65). Two cases must be studied.

(i) Medium 1 has zero energy ($\chi_1 = 0$). This case is relevant to dc conducting composites, under a prescribed average electric field, assuming that medium 1 is insulating and that medium 2 is conducting. Then $\mathbf{D}$ must be interpreted as the current density $\mathbf{J}$, and χ_2, χ_0 represent nonlinear conductivities. Letting p denote the volume concentration of component 2, the effective conductivity is nonzero for $p > p_c$ only, and behaves as $\chi_0 \sim \chi_2 (p - p_c)^{t(\gamma)}$ in the vicinity of p_c.[45] Setting $y = P_2/(\varepsilon_{2\parallel} E^0)$ and $\delta p = p - p_c$, and assuming that $\varepsilon_{2\parallel}$ and $\varepsilon_{2\perp}$ are of the same order of magnitude, the linear theory gives $\varepsilon_{0\parallel} \sim \varepsilon_{0\perp} \sim \varepsilon_{2\parallel} \delta p$, $M_2 \sim (a\,\delta p - y) E^0$ (where a is an irrelevant d-dependent constant) and $C_{2\parallel} \sim C_{2\perp} \sim E^{02}(1 + y^2)\,\delta p$. We now apply these findings to the nonlinear theory. First, by Eq. (61a), $P_2/\varepsilon_{2\parallel} = \chi_2 M_2^{\gamma}/\varepsilon_{2\parallel} - M_2$. Using the definition of y and the expression for M_2, this implies $M_2^{\gamma} \sim E^0(\varepsilon_{2\parallel}/\chi_2)\,\delta p$. Next, from the definition (59) of ε^{α} and a suitable change of variables, we have

$$\varepsilon_{2\parallel} \sim \chi_2 \int_0^1 dt \int dz (M_2^2 + 2\sqrt{t}\mathbf{M}_2 \cdot C_2^{1/2} \cdot \mathbf{z} + t\mathbf{z} \cdot C_2 \cdot \mathbf{z})^{(\gamma-1)/2} e^{-z^2/2}. \qquad (66)$$

Assume now that the behavior of $\varepsilon_{2\parallel}$ is controlled by C_2: since M_2 must decrease to 0 in the conductor at the percolation threshold, $y \to 0$ so that $C_2 \sim \delta p$. Hence $\varepsilon_{2\parallel} \sim C_2^{(\gamma-1)/2} \sim \delta p^{(\gamma-1)/2}$, and $M_2^2 \sim \delta p^{(1+1/\gamma)} \ll C_2$, which is consistent with our assumption. Then, from (63), $W \sim w_2(M_2) + \varepsilon_{2\parallel} C_2 \sim \delta p^{(\gamma+1)/2}$, so that $t(\gamma) = (\gamma+1)/2$.

(i) *Medium 2 has zero energy* ($\chi_2 = \infty$). This occurs when $p < p_c$ in the presence of an infinitely susceptible ("superconducting") component 2. The electric field then identically vanishes in this component *below* the percolation threshold, and its corresponding contribution to the overall energy is zero. The effective dielectric susceptibility then behaves as $\chi_0 \sim \chi_1 (p_c - p)^{-s(\gamma)}$ for $p < p_c$ in the vicinity of p_c.[45] We now set $\delta p = p_c - p$. In the linear theory, the quantities relative to the first component now depend on the above-defined y, a quantity pertaining to component 2. More precisely, the linear theory gives here $M_1 \simeq (d + y)/(d-1) E^0$, $M_2 = -y E^0$, $C_{1\parallel} \simeq C_{1\perp} \simeq E^{02}(1+y)^2 \delta p^{-1}/[d(d-1)]$. Since the field must vanish in component 2 in the power-law case, we deduce that $y = 0$, and only component 1 needs to be considered; Hence M_1 is finite and C_1 diverges. Translating Eq. (66) to the first component, we deduce that $\varepsilon_{1\parallel} \sim \delta p^{-(\gamma-1)/2}$ and finally that $W \sim w_1 \sim \delta p^{-(\gamma+1)/2}$; hence $s(\gamma) = (\gamma+1)/2$.

The above exponents $s(\gamma)$ and $t(\gamma)$ have already been found in the PC-WLHY theory.[14] In opposition, the PCK theory features[30] $s = 1$, $t = \gamma$, these values having been observed in other theories as well.[8,13,16,22] They are easily recovered from the above arguments by ignoring the dependence of the phase permittivity tensors with respect to the variances C^{α}, and indicate that the approach to the critical point is controlled by the mean field rather than by fluctuations. Fluctuations thus play a major role near the percolation threshold in determining a satisfactory nonlinear overall susceptibility. To illustrate this point, the behaviors at high (but finite) contrast of the three theories are compared in Fig. 1 for potentials with $\gamma = 5$ and various contrasts $\chi_2/\chi_1 = 0.5$, $10^{-1}, \ldots, 10^{-6}$. Our new theory apparently admits the PC-WLHY curve as a lower bound, as required.[9,10] The violation of the bound by the PCK theory is due to its exponents,[30] and becomes conspicuous for high contrasts only.

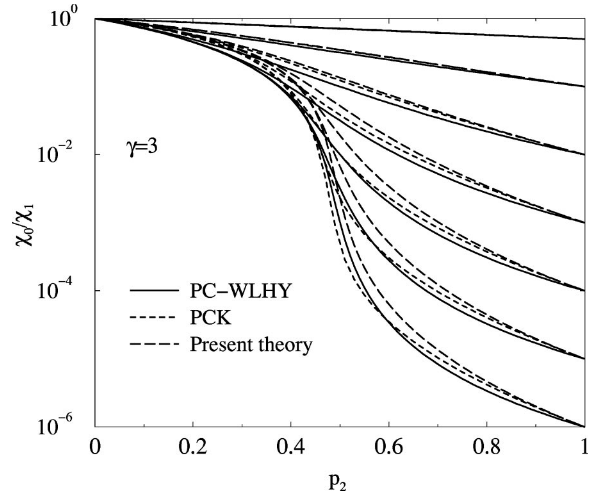

FIG. 1. Binary medium with power-law potentials ($\gamma = 5$) in two dimensions. Effective nonlinear susceptibility χ_0/χ_1 at increasing contrasts in the PC-WLHY, the PCK, and the present theories, vs p_2. The contrasts are $\chi_2/\chi_1 = 0.5$, $10^{-1}, \ldots, 10^{-6}$.

We found that dramatic crossover effects prevent an easy numerical determination of the critical exponents from our numerical procedure. Preliminary results where contrasts of order $10^{\pm 16}$ have to be reached for $\gamma = 5$ (not shown), are nonetheless fully consistent with the above asymptotic considerations (though convergence problems show up). The exponents are best observed in the case of truly infinite contrast, which will be discussed in detail elsewhere.[40]

C. Weak-contrast expansion

The new procedure is exact to second order in contrast, because it relies on a second-order Taylor expansion, and uses a permittivity tensor related to the tangent permittivity. The reader is reminded that in the weak-contrast expansion,[6,10,17,18,21,46] the local potentials $w_{\mathbf{x}}(\mathbf{y})$ are assumed to fluctuate weakly around their mean value. Introducing a bookkeeping parameter t to be set to 1 in the final results, the contrast $\delta w_{\mathbf{x}}(\mathbf{y})$ is defined by

$$w_{\mathbf{x}}(\mathbf{y}) = \langle w_{\mathbf{x}}(\mathbf{y}) \rangle + \delta w_{\mathbf{x}}(\mathbf{y}) t. \qquad (67)$$

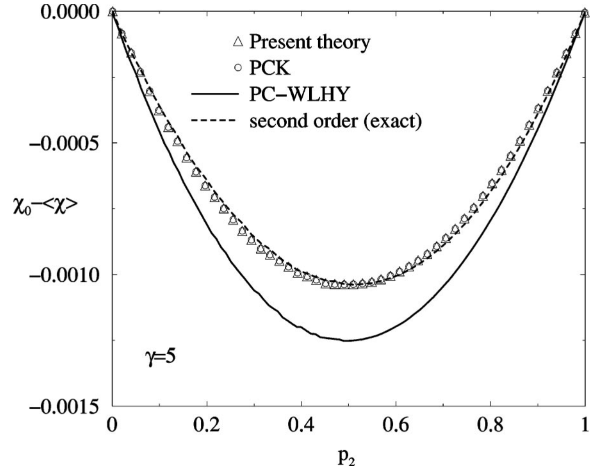

FIG. 2. Test of exactness to second order in the contrast. Binary medium with power-law potentials ($\gamma=5$) in two dimensions. Plot of $\chi_0-\langle\chi\rangle$ vs p_2, with $\chi_1=1.05$, $\chi_2=0.95$.

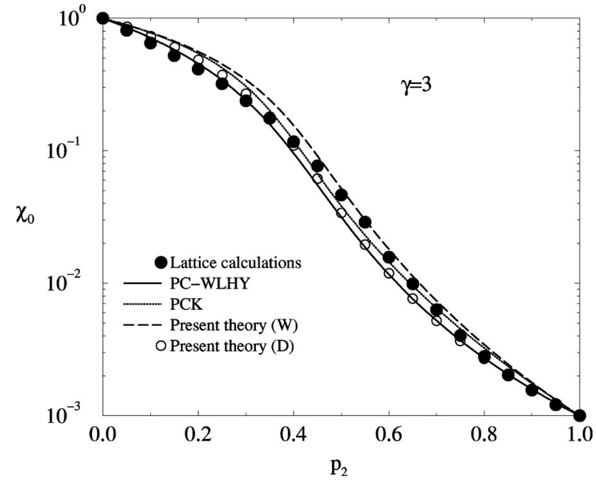

FIG. 3. Comparison between theoretical estimations of χ_0 and lattice numerical calculations (data borrowed from Ref. 14). Contrast $\chi_2/\chi_1=10^{-3}$. Power-law potentials with $\gamma=3$ in two dimensions (see text).

An expansion for the effective potential is sought for as a power series in t.[46] For a general potential, we have to second order[21]

$$W(\mathbf{E}^0)=\langle w(\mathbf{E}^0)\rangle+\frac{g_\parallel}{2}\langle[\partial_{\mathbf{E}}\delta w_{\mathbf{x}}(\mathbf{E}^0)\cdot\hat{\mathbf{E}}^0]^2\rangle t^2+O(t^3), \tag{68}$$

where $g_\parallel$ is computed from the Appendix with ε^0_{ij} replaced by

$$\varepsilon^0\equiv\langle\partial^2_{\mathbf{E}}w_{\mathbf{x}}(\mathbf{E}^0)\rangle. \tag{69}$$

This property can be checked analytically in the present theory at the price of tedious calculations which will not be reproduced here. Instead we illustrate it numerically for potentials of the type (65) and $\gamma=5$ in Fig. 2, where results from the PCK and PC-WLHY theories are also included.

D. Limits of the Gaussian approximation

The main shortcoming of the theory is the unconsistency between the values of the effective electric displacement obtained as

$$\mathbf{D}_0=\frac{\partial W}{\partial\mathbf{E}^0}\neq\left\langle\frac{\partial w_x}{\partial\mathbf{E}}\right\rangle, \tag{70}$$

when the average is carried out with the help of Gaussian averaging in each phase. A necessary condition for the theory to be fully consistent is that an equality instead holds. In order to decide which value of $\mathbf{D}^0$ must be retained here, we recall that only the value of the nonlinear susceptibility χ_0 obtained from the effective energy W is exact to second order in the contrast, and is therefore *a priori* acceptable.

In Figs. 3, 4, we display the predictions of the three theories at a contrast $\chi_2/\chi_1=10^{-3}$, for $\gamma=3$ and $\gamma=5$, respectively, along with the simulation results of Wan *et al.*[14] for power-law potentials on random resistor networks. The PC-WLHY curves lie closer to the simulation points than the other ones in both figures. The present theory apparently gives the worst results (the highest curves), as long as one extracts χ_0 from the energy. We also tested values of the nonlinear susceptibility extracted from the right-hand side (RHS) of expression (70). The results (not shown) were numerically higher than those extracted from W, which definitely rules out the use of the RHS in (70), as far as the present theory is concerned. More interestingly, however, the fourth curves (white circles) in Figs. 3,4 display values of χ_0 extracted from a "pragmatic" (and unjustified) identification of the nonlinear effective displacement to that of the linear underlying theory [see Eq. (23)], namely,

$$\mathbf{D}^0=\varepsilon^0\cdot\mathbf{E}^0+\mathbf{P}^0 \tag{71}$$

[the difference between this expression of $\mathbf{D}^0$ and that in the left-hand side of Eq. (70) is due, of course, to the dependence of c^0, $\mathbf{P}^0$, and ε^0 in $\mathbf{E}^0$]. We see that these last curves lie

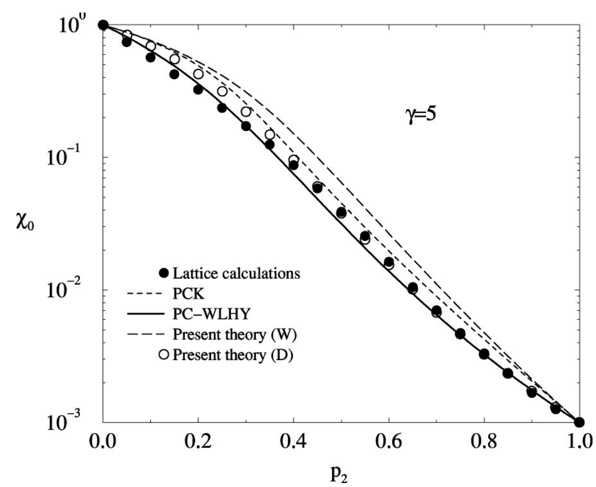

FIG. 4. Comparison between theoretical estimations of χ_0 and lattice numerical calculations (data borrowed from Ref. 14). Contrast $\chi_2/\chi_1=10^{-3}$. Power-law potentials with $\gamma=5$ in two dimensions (see text).

much closer to the simulation data, and resemble that of the PC-WLHY theory. These curves were obtained from the present theory, but similar observations can be made on the PCK theory as well (though the difference between the critical exponents of the two theories subsists). Note, however, that the corresponding values of χ_0 are *not* exact to second-order in the contrast. Because our theory is strictly valid for continuous media only, the relevance of comparisons to resistor networks data may be questioned. Since the extraneous dependence in E^0 due to nonlinearity—ignored in Eq. (71)—also lies at the crux of the mismatch in Eq. (70), however, it may well be that beyond the fact that the continuous theory cannot be expected to fully reproduce resistor networks results, curing the mismatch would improve the agreement to simulations in high contrast situations.

E. Possible improvements

In this perspective, modifications could be carried out along the following lines. In the author's mind, the theory presented here has to be understood as a first step towards a self-consistent calculation of the probability distribution of the electric field in each phase. At this stage, however, no self-consistency has been required on moments of degree higher than 2, the missing information being supplied trough the use of the Gaussian ansatz (chosen for simplicity reasons). From this point of view, the theory is not free of arbitrariness, which leaves room for further developments. In particular one could easily generalize[40] the central property (53) to a wider class of probability distributions (encompassing the Gaussian one), namely, distributions of the type $\mathcal{P}_\alpha(\mathbf{E})=g(\mathbf{\Delta E}\cdot C^{\alpha-1}\cdot\mathbf{\Delta E})$, where $g(x)$ is a suitably normalized function decaying sufficiently fast when $x\to\infty$. Note that neither the property of exactness to second-order in the contrast, nor the critical behavior discussed above, would be jeopardized by such an extension [Eq. (68) and the discussion of the critical regime are essentially independent of g]. Among additional self-consistent requirements to determine g, that of recovering an equality in Eq. (70) would certainly play a central role.

VIII. CONCLUSION

We have presented and discussed ideas (namely, the Gaussian approximation applied to strongly nonlinear composites, and the use of the second-order Taylor expansion with integral remainder) allowing for simultaneous self-consistency requirements on the first two moments of the probability distribution of the electric field in each phase. Thereby, we obtained a theory exact to second-order in the contrast, with a percolation threshold independent of the nonlinearity, which reduces to the Bruggeman theory in the linear case and which apparently admits the PC-WLHY theory as a lower bound, displaying in particular the same critical exponents.

In spite of the qualitative progress made, some problems remain, and the discrepancy with simulations data is in need for explanations. Further extensions of the theory, along the lines discussed above, might help improving the situation.

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APPENDIX: FLUCTUATIONS IN THE LINEAR THEORY

In this technical appendix, we complete our explicit solution for the EMT in a uniaxial medium with spontaneous polarization by giving the details required to compute the variances of the field (42), and we provide in particular expressions for the variations of $g=(g_\parallel,g_\perp)$ and of $\varepsilon^0=(\varepsilon^0_\parallel,\varepsilon^0_\perp)$ under variations $\delta\varepsilon^\alpha=(\varepsilon^\alpha_\parallel,\varepsilon^\alpha_\perp)$ of the uniaxial permittivity tensors in the phases.

We first recall the expression of g in a uniaxial medium.[48] It can be written

$$g=(g_\parallel,g_\perp)=(-n_\parallel/\varepsilon^0_\parallel,-n_\perp/\varepsilon^0_\perp), \tag{A1}$$

where we introduced the depolarization factors $n_{\parallel,\perp}$ of a sphere in a uniaxial anisotropic medium. Setting $r=\varepsilon^0_\parallel/\varepsilon^0_\perp$ and $u=\hat{\mathbf{k}}\cdot\hat{\mathbf{E}}^0$, these depolarization factors obey

$$n_\parallel+(d-1)n_\perp=1 \tag{A2}$$

with $(d>1,\ r<1)$

$$\begin{aligned} n_\parallel&=\int\frac{d\Omega_{\hat{\mathbf{k}}}}{S_d}\frac{ru^2}{1+(r-1)u^2}\\ &=\frac{S_{d-1}}{S_d}\int_{-1}^{1}du\,\frac{(1-u^2)^{(d-3)/2}ru^2}{1+(r-1)u^2}\\ &=\frac{r}{d}\,{}_2F_1(1,3/2;1+d/2;1-r). \end{aligned}\tag{A3}$$

The quantity $S_d=2\pi^{d/2}/\Gamma(d/2)$ is the area of the unit d-dimensional sphere, and ${}_2F_1$ is Gauss's hypergeometric function.[47] In physical dimensions, $n_\parallel$ has been computed under various equivalent forms.[17,32,35] We have

$$n_\parallel=1\ \ (d=1), \tag{A4a}$$

$$n_\parallel=\sqrt{r}/(1+\sqrt{r})\ \ (d=2), \tag{A4b}$$

$$n_\parallel=\frac{r}{1-r}\left(\frac{\operatorname{arctanh}\sqrt{1-r}}{\sqrt{1-r}}-1\right)\ \ (d=3). \tag{A4c}$$

Expression (A4c) is analytically continued to the region $r>1$. This remark also holds for $m_\times$, below.

Under variations $\delta\varepsilon^\alpha$ of the permittivities in the phases, ε^0 undergoes a variation $\delta\varepsilon^0=(\delta\varepsilon^0_\parallel,\delta\varepsilon^0_\perp)$ and the corresponding variation of g is $\delta g=(\delta g_\parallel,\delta g_\perp)$,

$$\delta g_\parallel=\Gamma_\parallel\delta\varepsilon^0_\parallel+(d-1)\Gamma_\times\delta\varepsilon^0_\perp, \tag{A5a}$$

$$\delta g_\perp=\Gamma_\times\delta\varepsilon^0_\parallel+(d-1)\Gamma_\perp\delta\varepsilon^0_\perp. \tag{A5b}$$

We introduced $\Gamma_\parallel = m_\parallel/\varepsilon_\parallel^{0\,2}$, $\Gamma_\times = m_\times/(\varepsilon_\parallel^0\varepsilon_\perp^0)$, $\Gamma_\perp = m_\perp/\varepsilon_\perp^{0\,2}$, and the "second order" depolarization factors $m_{\parallel,\times,\perp}$ defined by

$$m_\times = \frac{r}{d-1}\frac{dn_\parallel(r)}{dr} = -r\frac{dn_\perp(r)}{dr}, \tag{A6a}$$

$$m_\parallel + (d-1)m_\times = n_\parallel, \tag{A6b}$$

$$m_\times + (d-1)m_\perp = n_\perp. \tag{A6c}$$

For $d=1$, 2, 3, respectively, we find

$$m_\parallel = 1, \tag{A7a}$$

$$m_\times = \frac{1}{2}\frac{\sqrt{r}}{(1+\sqrt{r})^2}, \tag{A7b}$$

$$m_\times = \frac{1}{4}\frac{r}{(1-r)^2}\left[3-(2+r)\frac{\operatorname{arctanh}\sqrt{1-r}}{\sqrt{1-r}}\right]. \tag{A7c}$$

Next, the variations $\delta\varepsilon^0$ are determined by means of the effective-medium equations $\langle\mu_\parallel\rangle = \langle\mu_\perp\rangle = 1$, which lead to the system

$$\begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix}\begin{pmatrix} \delta\varepsilon_\parallel^0 \\ \delta\varepsilon_\perp^0 \end{pmatrix} = \begin{pmatrix} \delta b_1 \\ \delta b_2 \end{pmatrix}, \tag{A8}$$

where

$$a_{11} = g_\parallel\langle\mu_\parallel^2\rangle + (1-\langle\mu_\parallel^2\rangle)(\Gamma_\parallel/g_\parallel), \tag{A9a}$$

$$a_{12} = (d-1)(1-\langle\mu_\parallel^2\rangle)(\Gamma_\times/g_\parallel), \tag{A9b}$$

$$a_{21} = (1-\langle\mu_\perp^2\rangle)(\Gamma_\times/g_\perp), \tag{A9c}$$

$$a_{22} = g_\perp\langle\mu_\perp^2\rangle + (d-1)(1-\langle\mu_\perp^2\rangle)(\Gamma_\perp/g_\perp), \tag{A9d}$$

$$\delta b_1 = g_\parallel\langle\mu_\parallel^2\,\delta\varepsilon_\parallel\rangle = g_\parallel\sum_\alpha p_\alpha\mu_\parallel^{\alpha 2}\,\delta\varepsilon_\parallel^\alpha, \tag{A9e}$$

$$\delta b_2 = g_\perp\langle\mu_\perp^2\,\delta\varepsilon_\perp\rangle = g_\perp\sum_\alpha p_\alpha\mu_\perp^{\alpha 2}\,\delta\varepsilon_\perp^\alpha. \tag{A9f}$$

From these equations, the derivatives in Eqs. (42) are readily obtained by means of the explicit formulas

$$\frac{\partial W}{\partial\varepsilon_\parallel^\alpha} = \left[\frac{E^{02}}{2} + g_\parallel\mu_\parallel^{\alpha 2}\left(P^\alpha E^0 + \frac{g_\parallel}{2}\Delta P^{\alpha 2}\right)\right]\frac{\partial\varepsilon_\parallel^0}{\partial\varepsilon_\parallel^\alpha} - \frac{P^0E^0}{g_\parallel}\frac{\partial g_\parallel}{\partial\varepsilon_\parallel^\alpha} + \left(\frac{1}{2}\langle\mu_\parallel^2\Delta P^2\rangle + \frac{E^0}{g_\parallel}\langle\mu_\parallel^2 P\rangle\right)\left(\frac{\partial g_\parallel}{\partial\varepsilon_\parallel^\alpha} - g_\parallel^2\frac{\partial\varepsilon_\parallel^0}{\partial\varepsilon_\parallel^\alpha}\right), \tag{A10}$$

$$\frac{\partial W}{\partial\varepsilon_\perp^\alpha} = \frac{E^{02}}{2}\frac{\partial\varepsilon_\parallel^0}{\partial\varepsilon_\perp^\alpha} - \frac{P^0E^0}{g_\parallel}\frac{\partial g_\parallel}{\partial\varepsilon_\perp^\alpha} + \left(\frac{1}{2}\langle\mu_\parallel^2\Delta P^2\rangle + \frac{E^0}{g_\parallel}\langle\mu_\parallel^2 P\rangle\right)\times\left(\frac{\partial g_\parallel}{\partial\varepsilon_\perp^\alpha} - g_\parallel^2\frac{\partial\varepsilon_\parallel^0}{\partial\varepsilon_\perp^\alpha}\right). \tag{A11}$$

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