

# Field distributions and effective-medium approximation for weakly nonlinear media

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An effective-medium theory is proposed for random weakly nonlinear dielectric media. It is based on a new gaussian approximation for the probability distributions of the electric field in each component of a multi-phase composite. These distributions are computed to linear order from a Bruggeman-like self-consistent formula. The resulting effective-medium formula for the nonlinear medium reduces to Bruggeman's in the linear case. It is exact up to second order in a weak-disorder expansion, and close to the exact result in the dilute limit (in particular, it is exact for  $d = 1$  and  $d = \infty$ ). In a high contrast situation, the noise exponents are  $\kappa = \kappa' = 0$  near the percolation threshold. Numerical results are provided for different weak nonlinearities. The use of the Bruggeman formula as a starting point for nonlinear homogenization theories in dimensions  $d > 2$  is questioned on the basis of known exact bounds on the noise exponents.

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## I. INTRODUCTION

Numerous papers have been devoted to the problem of determining the effective constitutive law of random nonlinear composites in electrostatics<sup>1-16</sup>, of importance for understanding various nonlinear phenomena, either entering the class of “weakly nonlinear” (WNL) phenomena, or that of “strongly nonlinear” phenomena such as random fuse-type or dielectric breakdown-type failure of materials<sup>17</sup>.

A convenient approach to the homogenization problem is the “energetic” one<sup>20,21</sup>, which can be summarized as follows. Let  $w_x(E)$  be the local potential (the energy density) from which derives the local nonlinear constitutive law. The homogenization step consists in computing the effective potential  $W$  as the volume average

$$W = \langle w_x(E) \rangle, \quad (1)$$

from which the effective constitutive law can be deduced (see below). Denoting by  $p_\alpha$  the volume proportion of component (“phase”)  $\alpha$  in a composite medium where  $w_x = w_\alpha$  if  $x$  is in phase  $\alpha$ , and introducing the volume averages *per phase*  $\langle \cdot \rangle_\alpha$ ,  $W$  can be written

$$W = \sum_{\alpha} p_{\alpha} \langle w_{\alpha}(E) \rangle_{\alpha}. \quad (2)$$

A class of effective-medium theories, mostly concerned with weak power-law nonlinearities, use our ability to compute the second moment of the electric field  $E$  in the various phases of the composite<sup>18</sup>, completed by a decoupling approximation<sup>3,19</sup>

$$\langle w_{\alpha}(E) \rangle_{\alpha} \simeq w_{\alpha}(\langle E^2 \rangle_{\alpha}^{1/2}). \quad (3)$$

This approximation is exact only when  $w_{\alpha}$  is quadratic (the linear theory), or when the field is uniform, which occurs only if all the  $w_{\alpha}$  are equal, i.e. in a homogeneous medium.

The purpose of this paper is to present a means to overcome this approximation, starting from the following remark: actually, with an additional hypothesis of ergodicity *per phase*, a volume phase average can be reinterpreted as an average over some probability distribution for the electric field in the phase under consideration. In this paper, volume averages will henceforth be identified to statistical averages. Then, the problem of computing  $W$ , Equ. (2), reduces

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to computing the probability distribution  $\mathcal{P}_\alpha$  of the field in each phase, by means of which the phase averages  $\langle \cdot \rangle_\alpha$  can be carried out, without appealing to (3).

Note that the full probability distribution  $\mathcal{P}(\mathbf{E})$  of the electric field in the medium is then

$$\mathcal{P}(\mathbf{E}) = \sum_{\alpha} p_{\alpha} \mathcal{P}_{\alpha}(\mathbf{E}). \quad (4)$$

Thus, the probability distributions of the field *per phase* are functions of great importance, since they carry all the information needed to build up the effective nonlinear properties. A few analyses of  $\mathcal{P}(\mathbf{E})$  are available<sup>22</sup>, mainly focused on its tail of relevance for breakdown phenomena<sup>23</sup>. However, up to my knowledge, none has been specifically aimed at the individual components  $\mathcal{P}_{\alpha}(\mathbf{E})$ . Chen and Sheng studied numerically the distribution of the *local* field in binary composites<sup>24</sup>. An analytical model of the same distribution was subsequently proposed by Barthélémy and Orland, with excellent agreement with the latter simulations<sup>22</sup>. Both works were limited to linear composites, in a weak-contrast situation. A first numerical attempt in the continuum, in a strong-contrast situation this time, is due to Cheng and Torquato<sup>25</sup>. They carried out two-dimensional numerical calculations on disordered systems with inclusions of various shapes. An exact calculation for  $\mathcal{P}$  in a *linear* medium with Hashin-Shtrikman (HS) geometry was recently made using a density of states approach<sup>26</sup>. In these works, the distribution  $\mathcal{P}$  was found to be essentially bimodal for a binary medium (save in the HS geometry where fine-structure peaks are not wiped out by positional disorder), especially at high dielectric contrast<sup>25</sup>. This is understandable in view of the decomposition (4).

Still, in parallel with the second moment of the field, the *first* moment in each phase (the average field) can be computed as well from any linear effective-medium theory, without requiring any *a priori* knowledge about  $\mathcal{P}_{\alpha}$ . Widely used in mechanics of random continuous media in the framework of the so-called “thermoelastic problem”<sup>27</sup>, this fact seems to have escaped most of the literature on random dielectric media, excepted in the work by Ponte Castañeda and Kailasam<sup>15</sup> where it is used to compute reference fields around which non-linear potentials are expanded prior to homogenization. Knowing the first two moments is enough to attempt a modelization of  $\mathcal{P}_{\alpha}$ . The proposal examined in the present paper is to approximate each  $\mathcal{P}_{\alpha}(\mathbf{E})$  by a gaussian *vector* distribution, and thereby, to estimate (2) for any potential, without appealing to the strong (and somewhat artificial) decoupling assumption (3).

The scope of the paper is limited however to WNL media, for which it is sufficient to compute the field distribution to linear order only<sup>1,2</sup>. An extension of these considerations to strongly nonlinear media will be examined elsewhere.

## II. GENERAL FRAMEWORK

Let us therefore consider a  $d$ -dimensional random medium described by WNL thermodynamic potentials of the type

$$w_x(E) = w_x^{\text{lin}}(E) + w_x^{\text{nl}}(E), \quad (5)$$

where

$$w_x^{\text{lin}}(E) = \frac{1}{2} \varepsilon E^2 \quad (6)$$

( $\varepsilon(x)$  being the local permittivity entering the linear part), and  $w_x^{\text{nl}}(E)$  is the nonlinear (i.e. non-quadratic) part of the potential, assumed to be small. The local constitutive law is

$$D_i = \frac{\partial w_x(E)}{\partial E_i}. \quad (7)$$

The potential is prescribed in each phase:  $w_x(E) = w_{\alpha}(E)$  if  $x \in \alpha$ . Under the ergodic hypothesis introduced above, and a “boundary” condition<sup>28</sup>  $\langle \mathbf{E} \rangle = \mathbf{E}^0$ , the effective potential reads

$$W(E^0) = W_{\text{lin}}(E^0) + W_{\text{nl}}(E^0), \quad (8)$$

where

$$W_{\text{lin}}(E^0) = \frac{1}{2} \sum_{\alpha} p_{\alpha} \varepsilon_{\alpha} \langle E^2 \rangle_{\alpha}, \quad (9)$$

$$W_{\text{nl}}(E^0) = \sum_{\alpha} p_{\alpha} \langle w_{\alpha}^{\text{nl}}(E) \rangle_{\alpha}. \quad (10)$$

The effective constitutive law in the homgeneized medium is then

$$D_i^0 = \frac{\partial W(E^0)}{\partial E_i^0}. \quad (11)$$

To first order in  $w_x^{\text{nl}}$ , the field distributions entering (9) and (10) need only be computed at the linear level<sup>1,2</sup>. The procedure is the following one. An extended linear *anisotropic* homogeneization problem is first considered, with an artificial ferroelectric-like part added to the potential<sup>15</sup>:

$$\tilde{w}_x^{\text{lin}}(E) = \frac{1}{2} E_i \varepsilon_{ij} E_j + P_i E_i, \quad (12)$$

where  $\varepsilon_{ij}(x) = \varepsilon_{ij}^\alpha$  and  $P_i(x) = P_i^\alpha$  if  $x \in \alpha$ . The corresponding local constitutive law is  $D_i = \varepsilon_{ij} E_j + P_i$ . Assuming one knows an estimate for

$$\tilde{W}_{\text{lin}}(E^0) = \langle \tilde{w}_x^{\text{lin}}(E) \rangle, \quad (13)$$

(cf. below), the following *exact* relations hold for the distribution  $\mathcal{P}_\alpha$  of the original linear problem (9):

$$\langle E_i \rangle_\alpha = \frac{1}{p^\alpha} \frac{\partial \tilde{W}_{\text{lin}}}{\partial P_i^\alpha} \bigg|_{\substack{\varepsilon_{ij} = \varepsilon \delta_{ij} \\ P_i = 0}}, \quad (14a)$$

$$\langle E_i E_j \rangle_\alpha = \frac{2}{p^\alpha} \frac{\partial \tilde{W}_{\text{lin}}}{\partial \varepsilon_{ij}^\alpha} \bigg|_{\substack{\varepsilon_{ij} = \varepsilon \delta_{ij} \\ P_i = 0}}. \quad (14b)$$

I emphasize that the derivatives are computed at the point  $P_i = 0$ ,  $\varepsilon_{ij}^\alpha = \varepsilon^\alpha \delta_{ij}$  where  $\varepsilon^\alpha$  is the scalar permittivity in (5) and (9). There,  $\tilde{w}_x^{\text{lin}} = w_x^{\text{lin}}$  and  $\tilde{W}_{\text{lin}} = W_{\text{lin}}$ . Note that (14b) is a slight generalization of the formula<sup>18</sup>

$$\langle E^2 \rangle_\alpha = \frac{2}{p^\alpha} \frac{\partial W_{\text{lin}}}{\partial \varepsilon^\alpha}. \quad (15)$$

Both (14a) and (14b) are particular cases of a more general theorem, the demonstration of which can be found in appendix B of the review by Ponte Castañeda and Suquet on nonlinear composites<sup>29</sup>. This theorem is a consequence of the equality  $\langle D_i E_i \rangle = \langle D_i \rangle \langle E_i \rangle$  which holds only for particular boundary conditions, or in the infinite-volume limit if there exists an effective permittivity.

Let us set

$$M_i^\alpha = \langle E_i \rangle_\alpha, \quad (16)$$

$$C_{ij}^\alpha = \langle E_i E_j \rangle_\alpha - M_i^\alpha M_j^\alpha, \quad (17)$$

with  $M_i^\alpha \equiv M^\alpha \hat{E}_i^0$  and  $C_{ij}^\alpha \equiv C_\parallel^\alpha \hat{E}_i^0 \hat{E}_j^0 + C_\perp^\alpha (\delta_{ij} - \hat{E}_i^0 \hat{E}_j^0)$ , since the averages only depend on the direction  $\hat{E}_i^0 = E_i^0 / E^0$  of the macroscopic field. I introduce now the gaussian approximation for the probability distributions  $\mathcal{P}_\alpha$

$$\mathcal{P}_\alpha(\mathbf{E}) = \frac{1}{[(2\pi)^d \det(C^\alpha)]^{1/2}} e^{-\frac{1}{2} (E_i - M_i^\alpha) C_{ij}^{\alpha^{-1}} (E_j - M_j^\alpha)}, \quad (18)$$

which constitutes the main ingredient of the theory.

There remains to compute an approximation for the quantities  $\langle E_i \rangle_\alpha$  and  $\langle E_i E_j \rangle_\alpha$ , in order to completely define the above gaussian distributions. Let us therefore estimate  $\tilde{W}_{\text{lin}}(E^0)$  through an extension of the Bruggeman self-consistent effective-medium approximation<sup>30</sup> to potentials of the form (12). The formula is given and discussed in Appendix A. Using the scalar permittivities and  $P_i = 0$ , this estimate reduces to

$$W_{\text{lin}}(E^0) = \frac{1}{2} \varepsilon_0 E^{02}, \quad (19)$$

where  $\varepsilon_0$  is the usual Bruggeman effective permittivity<sup>30</sup>. In addition, the derivatives (14a) and (14b) of the estimate for  $\tilde{W}_{\text{lin}}(E^0)$  yield

$$M^\alpha = \mu^\alpha E^0, \quad (20)$$

$$\langle E_i E_j \rangle_\alpha = z \mu^{\alpha 2} \left[ E_i^0 E_j^0 - \frac{E^{0 2}}{d+2} (1 - y \langle \mu^2 \rangle) \delta_{ij} \right], \quad (21)$$

where

$$\mu^\alpha = \frac{d \varepsilon^0}{\varepsilon^\alpha + (d-1) \varepsilon^0}, \quad (22)$$

$$y = \frac{\varepsilon_0}{\langle \mu^2 \varepsilon \rangle}, \quad (23)$$

$$z = \frac{d+2}{d \langle \mu^2 \rangle + 2/y}. \quad (24)$$

Note that the Bruggeman equation for  $\varepsilon_0$  reads  $\langle \mu \rangle = 1$ , or alternatively  $\langle \mu \varepsilon \rangle = \langle \mu \rangle \varepsilon_0 = \varepsilon_0$ . From these relations, we check that: (i)  $\langle \varepsilon E \rangle = \varepsilon^0 \langle E \rangle$ ; (ii)  $\langle \varepsilon E^2 \rangle = \varepsilon^0 \langle E \rangle^2$ , where  $\langle E \rangle = E^0$ , as must be. Expressions for  $C_\parallel^\alpha$  and  $C_\perp^\alpha$  are readily obtained from (21).

In Fig. 1 are displayed curves for the probability density  $\mathcal{P}_\parallel(E_\parallel/E^0)$  of the normalized longitudinal electric field,  $E_\parallel = \mathbf{E} \cdot \hat{\mathbf{E}}^0$ , for a binary medium with dielectric ratio  $\varepsilon_2/\varepsilon_1 = 5$ , for various fractions  $p_2$  of material 2. The space dimension is  $d = 2$ . With  $\mathcal{P}(E)$  given by (4), and  $\mathbf{E}$  written as  $\mathbf{E} = E_\parallel \hat{\mathbf{E}}^0 + \mathbf{E}_\perp$  (which defines  $\mathbf{E}_\perp$ ),  $\mathcal{P}_\parallel$  is obtained as

$$\mathcal{P}_\parallel(E_\parallel/E^0) = E^0 \int d^{d-1} E_\perp \mathcal{P}(\mathbf{E}) = E^0 \sum_\alpha p_\alpha \frac{1}{(2\pi C_\parallel^\alpha)^{1/2}} e^{-\frac{1}{2} \frac{(E_\parallel - M^\alpha)^2}{C_\parallel^\alpha}}, \quad (25)$$

where  $E_\perp$  is the transverse component of the field:  $\mathbf{E}_\perp = \mathbf{E} - E_\parallel \hat{\mathbf{E}}^0$ . The probability density tends towards a Dirac peak, centered at  $E_\parallel/E^0 = 1$  as  $p_2 \rightarrow 0$  or  $p_2 \rightarrow 1$ , since the field is then equal to the applied field  $\mathbf{E}^0$ . For intermediate volume fractions, the bimodal character of the probability distribution is evident.

Under the same conditions, Fig. 2 displays the probability density of the scaled modulus of the transverse electric field,  $E_\perp/E^0$ . This distribution is defined, for  $E_\perp > 0$ , by

$$\mathcal{P}_\perp(E_\perp/E^0) = E^{0 d-1} \int dE_\parallel d\Omega_{\mathbf{E}_\perp} \mathcal{P}(\mathbf{E}) = E^0 \sum_\alpha p_\alpha \frac{S_{d-1}}{(2\pi C_\perp^\alpha)^{(d-1)/2}} \left( \frac{E_\perp}{E^0} \right)^{d-2} e^{-\frac{1}{2} \frac{E_\perp^2}{C_\perp^\alpha}}, \quad (26)$$

where  $S_d = 2\pi^{d/2}/\Gamma(d/2)$  is the surface of a  $d$ -dimensional unit sphere. Unlike the previous one, this distribution is not multimodal since the mean value of the transverse field is  $\langle \mathbf{E}_\perp \rangle_\alpha = \mathbf{0}$  in each phase, whatever the volume fractions.

Fig. 3 displays  $\mathcal{P}_\parallel(E_\parallel/E^0)$  for a higher dielectric ratio  $\varepsilon_2/\varepsilon_1 = 1000$ . The concentrations are  $p_2 = 0.12, 0.24, 0.36$ . These plots can be compared to that obtained from hard disks simulations by Cheng and Torquato (CT) in Fig. 3 of Ref.<sup>25</sup>, for the same dielectric contrast and concentrations  $p_2 = 0.2, 0.4, 0.6$ . These concentrations differ from ours by a factor 0.6. Apart from these differences, the overall features (shape and heights) of the probability distributions are well rendered: a Dirac peak close to  $E_\parallel = 0$  indicates that the field is almost null in phase 2, and a widely spread contribution from phase 1 for the highest  $p_2$  indicates an enhancement of the fluctuations in this phase as the percolation threshold (for the Bruggeman theory), or the jamming threshold (for the CT simulations) is approached. The differences in the values for  $p_2$  can be explained by the fact that the present theory relies on the Bruggeman effective-medium formula, of relevance for cell-materials but not really adequate for hard disks. Moreover, the overall agreement between our distributions and that of CT is not so good at moderate dielectric contrast: Bruggeman's theory completed by the gaussian approximation somewhat overestimates, because of its percolating nature, the width of the probability distributions. Better adequation with the CT simulations would probably be obtained with an effective-medium theory for dielectric-coated inclusions, which would prevent percolation. Since our main objective however is to discuss an effective-medium theory for WNL composites, such an improvement will not be considered here.

With these results in hand, analytical investigations of the theory can be carried out in some limiting cases, as well as numerical ones in more complicated situations. The next section examines particular WNL potentials.

### III. WEAKLY NONLINEAR POWER-LAW POTENTIAL

As a first application, a WNL local potential of the form

$$w_x(E) = \frac{1}{2}\varepsilon E^2 + \frac{1}{4}\chi E^4 + O(E^6), \quad (27)$$

is considered, where the  $E^4$  term is the first in a series of corrections to the linear behaviour. The corresponding constitutive law is

$$D_i = [\varepsilon + \chi E^2 + O(E^4)]E_i. \quad (28)$$

This approximation actually is a weak-field one.

#### A. Effective-medium formula

The effective potential takes the form

$$W(E^0) = \frac{1}{2}\varepsilon^0 E^{02} + \frac{1}{4}\chi^0 E^{04} + O(E^{04}), \quad (29)$$

where  $\varepsilon^0$  is the Bruggeman result, and<sup>2,4</sup>

$$\chi^0 = \langle \chi E^4 \rangle / E^{04} = \sum_{\alpha} p_{\alpha} \chi_{\alpha} \langle E^4 \rangle_{\alpha} / E^{04}. \quad (30)$$

The phase averages  $\langle E^4 \rangle_{\alpha}$  are carried out with the help of Wick's theorem which allows to write down expressions for the integer moments of a *centered* vector gaussian distribution by mere inspection<sup>31</sup>: with  $A_i = E_i - M_i^{\alpha}$ ,

$$\langle A_i A_i A_j A_j \rangle_{\alpha} = \langle A_i A_i \rangle_{\alpha} \langle A_j A_j \rangle_{\alpha} + 2 \langle A_i A_j \rangle_{\alpha} \langle A_i A_j \rangle_{\alpha}. \quad (31)$$

We deduce

$$\begin{aligned} \langle E^4 \rangle_{\alpha} &= \langle E_i E_i E_j E_j \rangle_{\alpha} \\ &= C_{ii}^{\alpha} C_{jj}^{\alpha} + 2 C_{ij}^{\alpha} C_{ij}^{\alpha} + 2 M^{\alpha 2} C_{ii}^{\alpha} + 4 M_i^{\alpha} M_j^{\alpha} C_{ij}^{\alpha} + M^{\alpha 4} \\ &= \left[ C_{\parallel}^{\alpha} + (d-1) C_{\perp}^{\alpha} + M^{\alpha 2} \right]^2 + 2 \left[ C_{\parallel}^{\alpha 2} + (d-1) C_{\perp}^{\alpha 2} + 2 M^{\alpha 2} C_{\parallel}^{\alpha} \right]. \end{aligned} \quad (32)$$

A few simplifications yield the simple result

$$\chi^0 = \left[ (1 + 2/d)y^2 + (2 - 2/d)z^2 - 2 \right] \langle \mu^4 \chi \rangle. \quad (33)$$

As a first check, we remark that in a non-disordered situation where  $\varepsilon$  and  $\chi$  are constant in the medium, we have  $\varepsilon_0 = \varepsilon$  and  $\mu = 1$ , so that  $y = z = 1$ ; whence  $\chi_0 = \chi$ , as was expected.

#### B. Weak-contrast expansion

A further check for the self-consistent formula (33) consists in examining its weak-contrast limit. An exact expression is known for any nonlinear potential, which is first briefly reminded. In the weak-contrast expansion<sup>5,7,15,16</sup>, the local potentials  $w_x(y)$  are assumed to fluctuate weakly around their mean value  $w^{(0)}(y) = \langle w_x(y) \rangle$ . Introducing a bookkeeping parameter  $t$  to be set to 1 in the final results, the contrast,  $w_x^{(1)}(y)$ , is defined by

$$w_x(y) = w^{(0)}(y) + w_x^{(1)}(y)t, \quad (34)$$

so that  $\langle w_x^{(1)}(y) \rangle = 0$ . An expansion for the effective potential is sought for in the form:

$$W(E^0) = W^{(0)} + W^{(1)}t + W^{(2)}t^2 + \dots \quad (35)$$

Then<sup>15</sup>,

$$W(E^0) = \langle w(E^0) \rangle - \frac{1}{2} \frac{n_{\parallel}}{\varepsilon_{\parallel}} \left\langle \left[ \partial_i w_x^{(1)}(E^0) \hat{E}_i^0 \right]^2 \right\rangle t^2 + O(t^3), \quad (36)$$

where

$$n_{\parallel} = - \int \frac{d\Omega_{\hat{\mathbf{k}}}}{S_d} \frac{r(\hat{\mathbf{k}} \cdot \hat{\mathbf{E}}^0)^2}{1 + (r-1)(\hat{\mathbf{k}} \cdot \hat{\mathbf{E}}^0)^2}, \quad (37)$$

$$\partial_i \partial_j w^{(0)}(E^0) \equiv \varepsilon_{\parallel} \hat{E}_i^0 \hat{E}_j^0 + \varepsilon_{\perp} (\delta_{ij} - \hat{E}_i^0 \hat{E}_j^0), \quad (38)$$

$$r = \varepsilon_{\parallel} / \varepsilon_{\perp}. \quad (39)$$

This result is the exact one<sup>15</sup>. An analogous expansion for the complementary potential  $\tilde{W}(D^0)$  defined from the dual homogeneization problem can be written down<sup>15,16</sup>.

Let us therefore set  $\varepsilon = \langle \varepsilon \rangle + \delta \varepsilon t$  and  $\chi = \langle \chi \rangle + \delta \chi t$ . The Bruggeman permittivity  $\varepsilon^0$  is exact to second order in the contrast:

$$\varepsilon^0 = \langle \varepsilon \rangle \left[ 1 - \frac{\langle \delta \varepsilon^2 \rangle}{d \langle \varepsilon \rangle^2} t^2 + O(t^3) \right]. \quad (40)$$

Expanding (33) to second order in  $t$  entails<sup>16</sup>

$$\chi^0 = \langle \chi \rangle \left\{ 1 + \left[ \frac{2(d+8)}{d(d+2)} \frac{\langle \delta \varepsilon^2 \rangle}{\langle \varepsilon \rangle^2} - \frac{4}{d} \frac{\langle \delta \varepsilon \delta \chi \rangle}{\langle \varepsilon \rangle \langle \chi \rangle} \right] t^2 + O(t^3) \right\}, \quad (41)$$

which can directly be obtained from a first-order expansion of (36) in  $\chi$ .

Comparing now formula (33) to the widely used approximation

$$\chi^0 \simeq \chi_2^0 = \sum_{\alpha} \chi_{\alpha} \langle E^2 \rangle_{\alpha}^2 / E^{04} = y^2 \langle \mu^4 \chi \rangle, \quad (42)$$

we see that the latter is *not* exact to second order in the contrast, save in  $d = 1$ : its expansion indeed reads

$$\chi_2^0 = \langle \chi \rangle \left\{ 1 + \left[ \frac{2(d+2)}{d^2} \frac{\langle \delta \varepsilon^2 \rangle}{\langle \varepsilon \rangle^2} - \frac{4}{d} \frac{\langle \delta \varepsilon \delta \chi \rangle}{\langle \varepsilon \rangle \langle \chi \rangle} \right] t^2 + O(t^3) \right\}. \quad (43)$$

Hence, the gaussian decoupling which accounts for the vector character of the electric field, of importance in nonlinear problems, is superior to the simple approximation  $\langle E^4 \rangle \simeq \langle E^2 \rangle^2$ .

### C. Dilute limit

For a binary medium with two components of constitutive parameters  $(\varepsilon_1, \chi_1)$  and  $(\varepsilon_2, \chi_2)$ , the dilute limit is the limiting situation where the volume fraction  $p$  of (e.g.) component 2 is small. Setting

$$T = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + (d-1)\varepsilon_1}, \quad (44)$$

an expansion, to first order in  $p$  of (33) yields

$$\chi^0 = \chi_1 + \left\{ (\chi_2 - \chi_1)(T-1)^4 + \chi_1 T^2 \left[ 2 \frac{d(d+8)}{d+2} - 4T + T^2 \right] \right\} p + O(p^2). \quad (45)$$

Bergman computed the exact effective nonlinear conductivity of a binary medium in the dilute limit, for spherical inclusions<sup>4,33</sup>. His result takes the form

$$\begin{aligned} \chi_{\text{exact}}^0 = \chi_1 + & \left\{ (\chi_2 - \chi_1)(T-1)^4 \right. \\ & \left. + \chi_1 T^2 \left[ 2 \frac{d(d+8)}{d+2} + 4 \frac{d(d-4)}{d+2} T + \frac{d(3d^2 - 10d + 16)}{3(d+2)} T^2 \right] \right\} p + O(p^2). \end{aligned} \quad (46)$$

Compared to (46), expression (45) becomes exact when  $\chi_1 = 0$  (nonlinear inclusion in a linear host). It also becomes exact for  $d = 1$ , and in the limit  $d \rightarrow \infty$  where the field is  $E = E_0$  in each phase so that  $\chi^0 = \langle \chi \rangle$ . Moreover, it is exact up to order  $T^2$ , which is consistent with its correct limiting weak-contrast behavior. For  $d = 2$ , it is exact up to order  $T^3$ . However, the term of order  $T^4$  is wrong for  $2 \leq d < \infty$ . The reason for this misbehaviour is that the dilute limit requires an exact computation of  $\langle E^4 \rangle_\alpha$  (to linear order)<sup>4</sup>, whereas we approximate it through a gaussian average.

On the other hand, expression (42) yields

$$\chi_2^0 = \chi_1 + \{(\chi_2 - \chi_1)(T - 1)^4 + \chi_1 T^2 [2(d + 2) - 4T + T^2]\} p + O(p^2), \quad (47)$$

once again a result less precise than (45).

At the present time, since it requires an exact solution for the one-body problem, the test of the dilute limit appears to be the most challenging one for nonlinear effective-medium theories.

This test is illustrated in Fig. 4 which displays  $\chi^0$ , as calculated from (33) and (42), against the concentration  $p$  of medium 2, for moderate dielectric contrast and  $d = 2$ . The thick line segments at  $p = 0, 1$  represent exact tangents obtained from (46). The tangent at  $p = 1$  follows from substituting  $(1 - p)$  for  $p$  and interchanging the indices 1 and 2 in (46). Though not exact in the dilute limit, formula (33) yields tangents quite close to the exact ones. The marked inaccuracy of formula (42) near  $p = 0$  and  $p = 1$  results in a lower value for  $\chi^0$  in the whole concentration range.

#### D. Percolative behavior

Before examining the predictions of Equ. (33) near the percolation transition, I first briefly review the critical behaviour of WNL composites, and discuss a flaw of the Bruggeman formula in this context.

An insulator/perfect conductor binary mixture undergoes a percolation transition for a critical metal fraction  $p = p_c$ <sup>32</sup>. For WNL phases, the critical behaviour of the field fluctuations is now well understood<sup>35</sup>. In the limiting situation where  $\varepsilon_1 \ll \varepsilon_2$ , and for  $p \leq p_c$ , one observes a behaviour

$$\varepsilon^0 \propto \varepsilon_1 (p - p_c)^{-s}, \quad \chi^0 \propto \chi_1 (p - p_c)^{-(2s + \kappa')}; \quad (48)$$

on the contrary, for  $p > p_c$ , one has

$$\varepsilon^0 \propto \varepsilon_2 (p_c - p)^t, \quad \chi^0 \propto \chi_2 (p_c - p)^{(2t - \kappa)}, \quad (49)$$

where  $s$  and  $t$  are the superconductivity and conductivity exponents, and where  $\kappa$  and  $\kappa'$  are the noise exponents which characterize an anomalous nonlinear susceptibility enhancement near the threshold<sup>34,35</sup>. As a consequence of the exact inequality  $\langle E^4 \rangle \geq \langle E^2 \rangle^2$ , the exponents  $\kappa$  and  $\kappa'$  are necessarily positive.

In the Bruggeman formula,  $p_c = 1/d$ , and  $\varepsilon_0 \simeq \varepsilon_1/(1 - dp)$  for  $p < 1/d$ ,  $\varepsilon_0 \simeq \varepsilon_2(1 - dp)/(1 - d)$  for  $p > 1/d$  ( $\varepsilon_1 \ll \varepsilon_2$ ), whence  $s = t = 1$ . Reporting these expressions into (33) yields  $\kappa = \kappa' = 0$ , a result shared by all effective-medium formulae based on (15).

These values can be compared to exact bounds obtained for  $\kappa$  and  $\kappa'$  on the basis of the links-nodes-blob (LNB) model for electric percolation, now commonly accepted as a model for e.g. random resistor networks (RRNs). These are<sup>35</sup>

$$(3d - 4)\nu - 2t + 1 \leq \kappa \leq 2(d - 1)\nu - t, \quad (50a)$$

$$(4 - d)\nu - 2s + 1 \leq \kappa' \leq 2\nu - s, \quad (50b)$$

(for  $d \leq 6$ ; the values for  $d > 6$  are that of  $d = 6$ . Moreover  $t$  is not defined for  $d = 1$ ), where  $\nu$  is the correlation length exponent:  $\xi \propto |p - p_c|^{-\nu}$ . Though the latter information is absent from the Bruggeman theory, an inequality  $\nu > 0$  must hold in any situation relevant to percolating systems. In (50), the lower bounds must not be greater than the upper ones, which leads to lower bounds for the exponents  $s$  and  $t$  themselves, namely

$$s \geq (2 - d)\nu + 1, \quad (51)$$

$$t \geq (d - 2)\nu + 1. \quad (52)$$

In the Bruggeman theory,  $s = t = 1$  so that only  $\nu = 0$  – a problematic value by itself – is allowed, save for  $d = 2$ . Reporting  $\nu = 0$  into (50) leads to  $\kappa = \kappa' = -1$ , which is unacceptable. For  $d = 2$ , one obtains  $\kappa = \kappa' = 2\nu - 1$ . Compared to the values  $\kappa = \kappa' = 0$  of the above effective-medium theory, this gives  $\nu = 1/2$ , a non-conflicting value, though lower than the exact result  $4/3$ .

Of course, the fact that Bruggeman's formula is a poor approximation near the percolation threshold is well-known. However, this discussion enlightens a fundamental inconsistency in the Bruggeman exponent values which goes beyond mere numerical inadequacy, as long as one wishes to estimate the properties of systems obeying the LNB scheme. It is to be noted that for  $d = 2$ , where no inconsistency appears, the exact equality  $s = t$  holds (a consequence of self-duality)<sup>36</sup> and is obeyed by the Bruggeman exponents.

The above argument is to be brought together with another one by Bergman. Discussing the failure of a “non ambiguous” non-linear Bruggeman-type model, he concluded that bounds for fluctuations always refer to some particular type of microstructure, and that no such bounds exist which apply to all materials<sup>4</sup>.

Because of these problems, comparisons between effective-medium theories built on the Bruggeman formula, and simulations on RRNs are expected to be more significant in dimension  $d = 2$  (another reason for preferring the two-dimensional case as a test-bench is that the bond percolation threshold  $p_c = 1/2$  on a square lattice is exactly reproduced by Bruggeman's formula). In Fig. 5 are displayed formulas (33) and (42) against  $p$ , in a high contrast situation ( $\varepsilon_2/\varepsilon_1 = 10000$ ). The same trend as in Fig. 4 is observed: formula (33) yields a higher estimate. The values for  $\varepsilon_{1,2}$  and  $\chi_{1,2}$  are the same as that used in Fig. 3 of a paper by Levy and Bergman<sup>37</sup>, where simulations results on RRNs are reported, and compared to (42). The authors remarked that the height of the peak in  $\chi^0$  at  $p_c$  was badly underestimated by (42), which gives  $\chi_{\text{peak}}^0 \simeq 0.5$  only. In their numerical simulations, for a  $30 \times 30$  network, the peak height is found to be  $\chi_{\text{peak}}^0 \simeq 0.86$ . In the infinite-size limit, this value is expected to increase somewhat. In Fig. 5, the height of the peak given by (33) is  $\chi_{\text{peak}}^0 \simeq 1.0$ , which thus compares fairly well to simulations.

### E. Other power-law nonlinearities

In this section, potentials of the type (5) where<sup>1</sup>

$$w_x^{\text{nl}}(E) = \frac{\chi(x)}{\gamma + 1} E^{\gamma+1} \quad (53)$$

are considered. For simplicity,  $\gamma$  is assumed to be constant in the material. The gaussian averages over the field in each phase are carried out numerically, with the help of a bi-variate integration routine. One integration variable is the field modulus  $E$ , and the other is the cosine  $\mathbf{E} \cdot \mathbf{E}^0 / (E E^0)$ . In fig. 6 are shown plots for

$$\chi^0 = \langle \chi(E/E^0)^{\gamma+1} \rangle, \quad (54)$$

computed from gaussian averages, for a moderate dielectric contrast and various powers  $\gamma > 1$  ranging from  $\gamma = 1.5$  to  $\gamma = 5$ , against the volume fraction  $p$  of medium 2 (solid lines). An exponential enhancement of  $\chi^0$  is observed with increasing  $\gamma$ . Its origin lies in the existence of a non-zero probability for  $E > E^0$  in the composite, since  $\mathbf{E}^0 = \langle \mathbf{E} \rangle$  by definition. These fluctuations are amplified by the nonlinearity, while that for which  $E < E^0$  are reduced. The larger the width of the probability distribution, the larger this enhancement, so that the peak culminates in the region  $p \simeq p_c = 1/2$ . Once again, the estimates of the present theory are much higher than that predicted by the decoupling assumption

$$\chi_2^0 = \sum_{\alpha} p_{\alpha} \chi_{\alpha} \langle (E/E^0)^2 \rangle_{\alpha}^{(\gamma+1)/2}, \quad (55)$$

(dashed lines), especially for large values of  $\gamma$ . This is understandable, since the functions to be averaged can be written  $w_x^{\text{nl}}(E) = \chi(x) f(E^2)$ , where  $f(z) = z^{(1+\gamma)/2} / (1 + \gamma)$ . For  $\gamma > 1$ , these functions are convex, so that  $\langle f(z) \rangle \geq f(\langle z \rangle)$ . Therefore, the decoupling assumption always underestimates the fluctuations when applied to such potentials. An overestimation would instead take place with concave potentials.

## IV. CONCLUSION

In this article, a theory for the nonlinear susceptibility of weakly nonlinear composites is proposed. This theory is based on a multimodal gaussian approximation for the overall probability distribution of the electric field, to linear order. The parameters which define this distribution (means and second moments, in each phase of the disordered medium), are obtained from Bruggeman's theory, whose limitations are discussed in this context. The present model provides for the first time an analytical estimate for the probability distribution of the electric field, for arbitrarily high dielectric contrast, in percolating media. The resulting effective nonlinear susceptibility is exact to second order



in the contrast, and close to the exact result in the dilute limit. Significant quantitative improvement is obtained on previous nonlinear effective-medium theories, even in the percolating regime, at least in the two-dimensional case. This study emphasizes the importance of accounting for the vector character of the electric field when averaging the local potentials. Improvements of the present theory could consist in finding a better approximation than the gaussian one for the components of the probability distribution of the field. More realistic distributions could be obtained by, e.g., extending the perturbative theory of Barthélémy and Orland<sup>22</sup> to high dielectric contrasts via self-consistent effective-medium approximations.

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## APPENDIX A: SELF-CONSISTENT ANISOTROPIC LINEAR THEORY WITH PERMANENT POLARIZATION

With local potentials of the form (12), the homogeneized potential reads

$$\tilde{W}_{\text{lin}}(E^0) = \frac{1}{2} E_i^0 \varepsilon_{ij}^0 E_j^0 + P_i^0 E_i^0 + \frac{1}{2} \langle \Delta P_i \mu_{ij} g_{jk} \Delta P_j \rangle, \quad (\text{A1})$$

where

$$g_{ij} = - \int \frac{d\Omega_{\mathbf{k}}}{S_d} \frac{k_i k_j}{k_l \varepsilon_{lm}^0 k_m}, \quad (\text{A2})$$

$$\mu_{ij}^\alpha = [1 - g(\varepsilon^\alpha - \varepsilon^0)]_{ij}^{-1}, \quad (\text{A3})$$

$$P_i^0 = \langle P_j \mu_{ji} \rangle, \quad (\text{A4})$$

$$\Delta P_i^\alpha = P_i^\alpha - P_i^0, \quad (\text{A5})$$

and the Bruggeman condition for  $\varepsilon_{ij}^0$  is  $\langle \mu_{ij} \rangle = \delta_{ij}$ .

Formula (A1) has been derived by means of a functional formalism<sup>38</sup>, used with a trial potential of the form<sup>39</sup>  $W^0(E) = (1/2) E_i \varepsilon_{ij}^0 E_j + P_i^0 E_i + c^0$ . Rather than to give a demonstration which would complicate the present article, the validity of (A1) is established by looking at its consequences. First, one can easily check that, for a binary medium, (A1) exactly reduces to formula (2.16) in the work by Ponte Castañeda and Kailasam<sup>15</sup>. Next, the average field per phase,  $\langle E_i \rangle_\alpha = M_i^\alpha$ , is (using the symmetry of the tensors  $g$  and  $\mu g$ ;  $\mu$  itself is not necessarily symmetric)

$$M_i^\alpha = \frac{1}{p_\alpha} \frac{\partial \tilde{W}_{\text{lin}}(E^0)}{\partial P_i^\alpha} = \mu_{ij}^\alpha (E_j^0 + g_{jk} \Delta P_k^\alpha). \quad (\text{A6})$$

From the definition of  $P_i^0$ , one deduces that  $\langle M \rangle = E^0$ , in agreement with the boundary conditions. Finally, the macroscopic constitutive relation derived from the effective potential, namely

$$D_i^0 = \frac{\partial \tilde{W}_{\text{lin}}(E^0)}{\partial E_i^0} = \varepsilon_{ij}^0 E_j^0 + P_i^0, \quad (\text{A7})$$

is consistent with the equation

$$D_i^0 = \langle \varepsilon_{ij} E_j + P_i \rangle = \langle \varepsilon_{ij} M_j^\alpha + P_i^\alpha \rangle, \quad (\text{A8})$$

with  $M_j^\alpha$  computed with (A6). This directly follows from the equivalent form of the Bruggeman equation,  $\langle \varepsilon_{ij} \mu_{jk} \rangle = \varepsilon_{ij}^0 \langle \mu_{jk} \rangle = \varepsilon_{ij}^0$ , and from the identity  $\mu_{ij}^\alpha = \delta_{ij} + \mu_{ik}^\alpha g_{kl} (\varepsilon_{li}^\alpha - \varepsilon_{li}^0)$ .

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FIG. 1. Field probability density function  $\mathcal{P}_{\parallel}(E_{\parallel}/E^0)$  ( $E_{\parallel} = \mathbf{E} \cdot \hat{\mathbf{E}}^0$ ) in the Bruggeman approximation, for various volume fractions  $p_2$  of component 2. The dielectric constant ratio is  $\varepsilon_2/\varepsilon_1 = 5$ . Dots:  $p_2 = 0.05$  (violet, highest curve);  $p_2 = 0.25$  (blue, lowest curve). Solid line:  $p_2 = 0.5$  (green). Dashes:  $p_2 = 0.75$  (orange, lowest curve);  $p_2 = 0.95$  (red, highest curve).

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FIG. 2. Field probability density function  $\mathcal{P}_{\perp}(E_{\perp}/E^0)$  ( $E_{\perp} = \|\mathbf{E} - E_{\parallel}\mathbf{E}^0\|$ ) in the Bruggeman approximation, for various volume fractions  $p_2$  of component 2. The dielectric constant ratio is  $\varepsilon_2/\varepsilon_1 = 5$ . Dots:  $p_2 = 0.05$  (violet, highest curve);  $p_2 = 0.25$  (blue, lowest curve). Solid line:  $p_2 = 0.5$  (green). Dashes:  $p_2 = 0.75$  (orange, lowest curve);  $p_2 = 0.95$  (red, highest curve).

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FIG. 3. Field probability density function  $\mathcal{P}_{\parallel}(E_{\parallel}/E^0)$  ( $E_{\parallel} = \mathbf{E} \cdot \hat{\mathbf{E}}^0$ ) in the Bruggeman approximation, for various volume fractions  $p_2$  of component 2. The dielectric constant ratio is  $\varepsilon_2/\varepsilon_1 = 1000$ . Solid:  $p_2 = 0.12$ ; dashes:  $p_2 = 0.24$ ; dots:  $p_2 = 0.36$ .

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FIG. 4. Comparison between Eqs. (33) (solid line) and (42) (dashes). Effective nonlinear susceptibility vs. volume fraction  $p$  of component 2. The space dimension is  $d = 2$ . The parameters are:  $\varepsilon_1 = 1$ ,  $\varepsilon_2 = 10$ ,  $\chi_1 = 0.01$ ,  $\chi_2 = 0.1$ . The segments at  $p = 0$  and  $p = 1$  represent exact slopes computed with expression (46).

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FIG. 5. Comparison between Eqs. (33) (solid line) and (42) (dashes). Effective nonlinear susceptibility vs. volume fraction  $p$  of component 2. The space dimension is  $d = 2$ . The parameters are:  $\varepsilon_1 = 1$ ,  $\varepsilon_2 = 10^4$ ,  $\chi_1 = 10^{-4}$ ,  $\chi_2 = 0.1$ . The segment at  $p = 1$  represents the exact slope computed with expression (46).

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FIG. 6. Comparison between Eqs. (54) (solid line) and (55) (dashes) for power-law nonlinearities  $w_x^{\text{nl}} = \chi(x)E^{\gamma+1}/(\gamma+1)$ . Effective nonlinear susceptibility vs. volume fraction  $p$  of component 2. The space dimension is  $d = 2$ . The parameters are:  $\varepsilon_1 = 1$ ,  $\varepsilon_2 = 10$ ,  $\chi_1 = 0.01$ ,  $\chi_2 = 0.1$ . From bottom to top:  $\gamma = 1.5$  (blue), 2 (cyan), 3 (turquoise), 4 (green), 5 (black).