

# Optimal bounds on electric-field fluctuations for random composites

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The electric field inside a two-phase composite is studied when the composite sample is subjected to a constant applied electric field. Upper and lower bounds on the covariance tensor of the electric field are found in terms of the effective dielectric properties of the composite. The lower bounds are shown to be optimal for two well-known families of microgeometries. Lower bounds on the covariance tensor are found when only the phase area fractions and the two-point correlation function are available. For statistically isotropic composites optimal lower bounds are derived when only the phase area fractions are known. © 2000 American Institute of Physics. [S0021-8979(00)02920-0]

## I. INTRODUCTION

It is important to understand the behavior of the local electric field in composite materials as regions containing high fields are most often the first to suffer damage during service. The higher moments of the electric field provide information on the variation of the local electric field that is not revealed by the effective properties of the composite. Unfortunately, higher moments of the local electric field cannot be obtained directly through simple boundary measurements. However, effective properties can be easily measured by subjecting a composite sample to a uniform electric field and measuring the current passing through the boundary of the sample. In this work, we develop bounds on the covariance tensor of the electric field in terms of the effective dielectric constant. We exhibit microstructures for which these bounds are optimal. These bounds are used to recover bounds on the covariance tensor when only the phase area fractions and the two-point correlation function are available. For isotropic composites, we obtain a lower bound that is the most restrictive one in terms the area fraction occupied by each phase. The method introduced here relies on the explicit computation of the convex hull of a curve.

To fix ideas, we consider the two-dimensional problem for which the composite is made from long parallel cylinders with dielectric constants  $\epsilon_1$  and  $\epsilon_2$ . We suppose that the cylinders are parallel to the  $\mathbf{e}^3$  axis and that the composite is periodic in the plane transverse to the cylinders with period cell  $\Omega$ . Aside from periodicity, we make no assumption on the configuration of the two dielectrics inside the period. The dielectric constant for the composite is written  $\epsilon(\mathbf{x})$  taking values  $\epsilon_1$  in phase one and  $\epsilon_2$  in phase two. The composite is subjected to a constant electric field  $\bar{\mathbf{E}}$  with  $\bar{E}_3=0$  and non-zero components in the transverse plane. The local electric field  $\mathbf{E}$  is decomposed into a periodic fluctuation  $\nabla\varphi$  and the constant field  $\bar{\mathbf{E}}$  and is written  $\mathbf{E}=\nabla\varphi+\bar{\mathbf{E}}$  or  $\nabla\varphi=\mathbf{E}-\bar{\mathbf{E}}$ . The electric displacement  $\mathbf{D}$  is related to the electric field by  $\mathbf{D}=\epsilon(\mathbf{x})\mathbf{E}$  and

$$\text{div } \mathbf{D}=0. \tag{1}$$

We denote the local field induced by a uniform applied field  $\mathbf{e}^i$  along the  $i$ th coordinate direction by  $\mathbf{E}^i$ , where  $|\mathbf{e}^i|=1$ ,  $i=1,2$ , and the covariance tensor  $\sigma$  is defined by

$$\sigma_{ij}=\frac{1}{V}\int_{\Omega}(\mathbf{E}^i-\mathbf{e}^i)\cdot(\mathbf{E}^j-\mathbf{e}^j)dx, \tag{2}$$

where  $V$  is the area of the period cell  $\Omega$ . We write  $\bar{\mathbf{E}}=\mathbf{e}^1\bar{E}_1+\mathbf{e}^2\bar{E}_2$  and the mean-square fluctuation in the local field due to the applied field  $\bar{\mathbf{E}}$  is given by

$$\frac{1}{V}\int_{\Omega}(\mathbf{E}-\bar{\mathbf{E}})\cdot(\mathbf{E}-\bar{\mathbf{E}})dx=\sigma_{ij}\bar{E}_i\bar{E}_j, \tag{3}$$

where repeated indices indicate summation. The effective dielectric tensor is given by

$$\epsilon_{ij}^e=\frac{1}{V}\int_{\Omega}\epsilon(\mathbf{x})\mathbf{E}^i\cdot\mathbf{e}^jdx. \tag{4}$$

The area fraction of each phase is denoted by  $\theta_1$  and  $\theta_2$ , where  $\theta_1+\theta_2=1$ . Without loss of generality, we suppose that  $\epsilon_2>\epsilon_1$  and define the contrast  $\lambda$  to be the aspect ratio  $\epsilon_2/\epsilon_1$ . We set  $h=1/(\lambda-1)$ . In earlier work<sup>1</sup> optimal bounds on the mean-square fluctuation were obtained in terms of the area fractions of the phases. For every vector  $\bar{\mathbf{E}}$  the bounds are given by

$$0\leq\sigma_{ij}\bar{E}_i\bar{E}_j\leq U(\theta_2,\bar{\mathbf{E}}), \tag{5}$$

where  $U(\theta_2,\bar{\mathbf{E}})$  depends upon the contrast  $\lambda$  and is given by

$$U(\theta_2,\bar{\mathbf{E}})=(\theta_2f(1-\theta_2))|\bar{\mathbf{E}}|^2, \text{ for } h\geq 1, \tag{6}$$

and for  $h\leq 1$ ,

$$U(\theta_2,\bar{\mathbf{E}})=\begin{cases} (\theta_2f(h))|\bar{\mathbf{E}}|^2, & \text{if } \theta_2\leq 1-h, \\ (\theta_2f(1-\theta_2))|\bar{\mathbf{E}}|^2 & \text{if } \theta_2\geq 1-h. \end{cases} \tag{7}$$

Here, the function  $f(z)$  is defined by

$$f(z)=\frac{z}{(h+z)^2}. \tag{8}$$

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Extremal sequences of configurations that attain the bounds are shown to be given by the well-known finite-rank laminar microstructures.<sup>1</sup> The lower bound is attained by laminates of the first rank with layers oriented parallel to the applied field. On the other hand, the upper bound is saturated by laminates of first or second rank depending on the magnitude of the contrast. We mention that these bounds are established for three-dimensional problems as well in Ref. 1.

In this work, we extend the analysis to obtain bounds on the covariance tensor when in addition to knowing the area fractions of the phases we know two-point correlation information on the microgeometry and we can measure the effective dielectric constant. We introduce a method for obtaining bounds that is based upon the computation of the convex hull of a suitable curve. For completeness, we apply this method to recover the bounds (5), (6), and (7). The indicator function of phase two is denoted by  $\chi_2$ , taking the value one in phase two and zero outside. Following Willis<sup>2</sup> we introduce the tensor  $T$  that measures the local anisotropy of the composite defined by

$$T_{il} = \sum_{k \neq 0} \frac{k_i k_l}{|\mathbf{k}|^2} \frac{1}{V} \int_{\Omega} e^{2\pi i \mathbf{k} \cdot \mathbf{t}} c_{bb}(\mathbf{t}) dt. \tag{9}$$

Here,  $\mathbf{k}$  is a wave vector in Fourier space and  $c_{bb}(\mathbf{t})$  is the two-point correlation,

$$c_{bb}(\mathbf{t}) = \frac{1}{V} \int_{\Omega} \chi_2(\mathbf{x} + \mathbf{t}) \chi_2(\mathbf{x}) dx.$$

This function gives the probability that the ends of a rod of length and orientation described by the vector  $\mathbf{t}$  lies in phase two. The average of the two dielectric constants is written as  $\langle \epsilon \rangle = \theta_1 \epsilon_1 + \theta_2 \epsilon_2$ . We choose our coordinate system so that  $\mathbf{e}^1$  and  $\mathbf{e}^2$  are unit eigenvectors for the covariance tensor and the lower bound is given by

$$\begin{aligned} \sigma \geq & \frac{(\langle \epsilon \rangle - (\epsilon^e \mathbf{e}^1 \cdot \mathbf{e}^1))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{e}^1 \cdot \mathbf{e}^1)} \mathbf{e}^1 \otimes \mathbf{e}^1 \\ & + \frac{(\langle \epsilon \rangle - (\epsilon^e \mathbf{e}^2 \cdot \mathbf{e}^2))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{e}^2 \cdot \mathbf{e}^2)} \mathbf{e}^2 \otimes \mathbf{e}^2, \end{aligned} \tag{10}$$

where the inequality holds in the sense of quadratic forms, and  $\mathbf{e}^i \otimes \mathbf{e}^j$  is the matrix given by  $\mathbf{e}_k^i \mathbf{e}_l^j$ . In Sec. IV, we exhibit two well-known classes of microstructures for which this lower bound holds with equality. In order to write the upper bound we set  $\lambda_k^e = \epsilon^e \mathbf{e}^k \cdot \mathbf{e}^k$ , for  $k=1,2$ , and  $t_k = T \mathbf{e}^k \cdot \mathbf{e}^k$ , for  $k=1,2$  and the upper bound is given by

$$\sigma \leq a^1 \mathbf{e}^1 \otimes \mathbf{e}^1 + a^2 \mathbf{e}^2 \otimes \mathbf{e}^2, \tag{11}$$

where

$$\begin{aligned} a^k = & (\lambda_k^e - \epsilon_1) \left( \frac{1}{\epsilon_1} - \frac{1}{\epsilon_2} \right) + \frac{\theta_2^2 \epsilon_1 \epsilon_2}{(\theta_2 - t_k)(\epsilon_2 - \epsilon_1)^2} \\ & \times \left( \frac{\lambda_k^e - \epsilon_1}{\theta_2 \epsilon_1} - \frac{\epsilon_2 - \epsilon_1}{\epsilon_2} \right)^2, \quad k=1,2. \end{aligned} \tag{12}$$

Lower bounds on the covariance tensor can be obtained in terms of the two-point correlation function and area fractions

of the phases. To do this, we employ the anisotropic upper bounds on the effective dielectric constant<sup>3</sup> given by

$$\epsilon^e \leq \bar{\epsilon}, \tag{13}$$

where

$$\bar{\epsilon} = \epsilon_2 I - \theta_1 (\epsilon_2 - \epsilon_1) \left( I - \frac{\epsilon_2 - \epsilon_1}{\epsilon_2 \theta_1} T \right)^{-1}. \tag{14}$$

Here,  $I$  is the  $2 \times 2$  identity matrix. As before, the inequality given in Eq. (13) holds in the sense of quadratic forms and substitution into Eq. (10) gives the lower bound

$$\begin{aligned} \sigma \geq & \frac{(\langle \epsilon \rangle - (\bar{\epsilon} \mathbf{e}^1 \cdot \mathbf{e}^1))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{e}^1 \cdot \mathbf{e}^1)} \mathbf{e}^1 \otimes \mathbf{e}^1 \\ & + \frac{(\langle \epsilon \rangle - (\bar{\epsilon} \mathbf{e}^2 \cdot \mathbf{e}^2))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{e}^2 \cdot \mathbf{e}^2)} \mathbf{e}^2 \otimes \mathbf{e}^2. \end{aligned} \tag{15}$$

When the composite is statistically isotropic  $\bar{\epsilon}$  reduces to the Hashin and Shtrikman (HS) upper bound on effective properties given by  $\bar{\epsilon} = I \epsilon_{HS}$ , where

$$\epsilon_{HS} = \epsilon_2 - (1 - \theta_2) \left( \frac{1}{\epsilon_2 - \epsilon_1} - \frac{\theta_2}{2 \epsilon_2} \right)^{-1}, \tag{16}$$

$T = I[\theta_2(1 - \theta_2)/2]$ , and the lower bound on the covariance tensor becomes

$$\sigma \geq I \frac{2(\langle \epsilon \rangle - \epsilon_{HS})^2}{(\epsilon_2 - \epsilon_1)^2 \theta_2 (1 - \theta_2)}. \tag{17}$$

This bound is shown to be the most restrictive one that can be obtained in terms of area fractions and the dielectric constants of the two materials. This is shown in Sec. IV.

Related earlier work includes bounds for the mean-square fluctuation of the electric field given in the work of Beran.<sup>4</sup> More recently, bounds on the mean-square fluctuation inside each phase have been developed in Ref. 5. We point out that our results apply to the mathematically analogous cases of field fluctuations in steady-state heat transport, dc electric conductivity, and diffusion problems. The methods developed here apply to higher-dimensional problems. Moreover, these methods can be used to generate bounds in terms of higher-order correlation functions that partially describe the composite geometry. These topics are pursued elsewhere.

## II. INTEGRAL REPRESENTATION FORMULA FOR THE COVARIANCE TENSOR

The bounds follow from an integral representation formula for the covariance tensor. Such a formula was introduced for the effective dielectric constant in the work of Bergman.<sup>6</sup> Here, we write the formula for the effective dielectric constant in the form subsequently developed by Golden and Papanicolaou<sup>7</sup> given by

$$(\epsilon^e - \epsilon_1 I) / \epsilon_1 = \int_0^1 f^e(z) d\mu_{ij}(z), \tag{18}$$

where  $f^e(z) = 1/(h+z)$  and the matrix valued measure  $\mu_{ij}$  on  $[0,1]$  is the spectral measure of the self-adjoint operator  $\Gamma = P \chi_2$  where  $P = \nabla(\Delta)^{-1} \nabla$ . Here,  $\mu_{ij} \bar{E}_i \bar{E}_j \geq 0$  for every

vector  $\bar{\mathbf{E}}$ . In this section, we follow the scattering theory approach of Refs. 6, 7, and 8 to obtain the representation formula for the covariance given by

$$\begin{aligned} \sigma &= \int_0^1 f(z) d\mu_{ij}(z) \\ &= (\epsilon^e - \epsilon_1 I) / \epsilon_1 - h \int_0^1 \frac{1}{(h+z)^2} d\mu_{ij}(z). \end{aligned} \quad (19)$$

This formula has been reported earlier in Ref. 1.

To expedite the presentation, we rewrite the local dielectric constant  $\epsilon(\mathbf{x})$  as a positive perturbation from the uniform state  $\epsilon_1$ , i.e.,  $\epsilon(\mathbf{x}) = \epsilon_1 + (\epsilon_2 - \epsilon_1)\chi_2$  and  $\mathbf{E} = \nabla\varphi + \bar{\mathbf{E}}$ . Expanding  $\epsilon(\mathbf{x})$  in Eq. (1) gives

$$-\epsilon_1 \Delta\varphi = \text{div}((\epsilon_2 - \epsilon_1)\chi_2(\nabla\varphi + \bar{\mathbf{E}})). \quad (20)$$

Dividing both sides by  $\epsilon_1$ , applying  $(\Delta)^{-1}$  to both sides, and manipulation gives

$$\nabla\varphi + \bar{\mathbf{E}} + P[(\lambda - 1)\chi_2(\nabla\varphi + \bar{\mathbf{E}})] = \bar{\mathbf{E}}, \quad (21)$$

or

$$[\mathbf{I} + (\lambda - 1)\Gamma]\mathbf{E} = \bar{\mathbf{E}}, \quad (22)$$

from which we obtain the desired expression

$$\mathbf{E} = [\mathbf{I} + (\lambda - 1)\Gamma]^{-1}\bar{\mathbf{E}}. \quad (23)$$

Next, we write  $\epsilon^e$  in the equivalent from

$$\epsilon^e \bar{\mathbf{E}} \cdot \bar{\mathbf{E}} = \frac{1}{V} \int_{\Omega} \epsilon(\mathbf{x}) \mathbf{E} \cdot \mathbf{E} dx. \quad (24)$$

Expanding  $\epsilon(\mathbf{x})$  as  $\epsilon(\mathbf{x}) = \epsilon_1 + \chi_2(\epsilon_2 - \epsilon_1)$  and substitution into Eq. (24) gives

$$\epsilon^e \bar{\mathbf{E}} \cdot \bar{\mathbf{E}} = \frac{1}{V} \int_{\Omega} \epsilon_1 |\mathbf{E}|^2 dx + \frac{(\epsilon_2 - \epsilon_1)}{V} \int_{\Omega} \chi_2 |\mathbf{E}|^2 dx. \quad (25)$$

Rearranging terms gives the formula

$$\frac{1}{V} \int_{\Omega} |\mathbf{E}|^2 dx = \frac{\epsilon^e \bar{\mathbf{E}} \cdot \bar{\mathbf{E}}}{\epsilon_1} - \frac{(\lambda - 1)}{V} \int_{\Omega} \chi_2 |\mathbf{E}|^2 dx, \quad (26)$$

and

$$\begin{aligned} \frac{1}{V} \int_{\Omega} |\mathbf{E}|^2 dx &= \frac{1}{V} \int_{\Omega} |\nabla\varphi + \bar{\mathbf{E}}|^2 dx \\ &= \frac{1}{V} \int_{\Omega} |\mathbf{E} - \bar{\mathbf{E}}|^2 dx + |\bar{\mathbf{E}}|^2. \end{aligned} \quad (27)$$

Combining Eqs. (26) and (27) gives

$$\sigma_{ij} \bar{E}_i \bar{E}_j = \frac{(\epsilon^e - \epsilon_1 I) \bar{\mathbf{E}} \cdot \bar{\mathbf{E}}}{\epsilon_1} - \frac{(\lambda - 1)}{V} \int_{\Omega} \chi_2 |\mathbf{E}|^2 dx. \quad (28)$$

To get the representation we appeal to spectral theory. For any two periodic square integrable vector fields we write as in Ref. 8,

$$\langle \eta, \psi \rangle = \frac{1}{V} \int_{\Omega} \chi_2(\eta \cdot \psi) dx.$$

Here  $\langle \eta, \psi \rangle$  is an inner product for the Hilbert space of periodic square integrable vector fields. Substitution of Eq. (23) into Eq. (28) gives

$$\begin{aligned} \sigma_{ij} \bar{E}_i \bar{E}_j &= \frac{(\epsilon^e - \epsilon_1 I) \bar{\mathbf{E}} \cdot \bar{\mathbf{E}}}{\epsilon_1} - (\lambda - 1) \\ &\quad \times \langle [\mathbf{I} + (\lambda - 1)\Gamma]^{-1} \bar{\mathbf{E}}, [\mathbf{I} + (\lambda - 1)\Gamma]^{-1} \bar{\mathbf{E}} \rangle. \end{aligned} \quad (29)$$

It is easily seen that  $\Gamma$  is a positive symmetric operator with norm less than or equal to 1. Spectral theory delivers the spectral family  $R(z)$  associated with  $\Gamma$  for which  $\langle R(z)\mathbf{e}^i, \mathbf{e}^j \rangle$  is a function of bounded variation on  $[0, 1]$ . The spectral measure  $\mu_{ij}$  is given by  $\mu_{ij}(0, z] \equiv \mu_{ij}(z) = \langle R(z)\mathbf{e}^i, \mathbf{e}^j \rangle$  and

$$\begin{aligned} &\langle [\mathbf{I} + (\lambda - 1)\Gamma]^{-1} \bar{\mathbf{E}}, [\mathbf{I} + (\lambda - 1)\Gamma]^{-1} \bar{\mathbf{E}} \rangle \\ &= \int_0^1 \frac{1}{(1 + z(\lambda - 1))^2} d\mu_{ij}(z) \bar{E}_i \bar{E}_j. \end{aligned} \quad (30)$$

Collecting these results gives the integral representation formula (19).

### III. BOUNDS ON THE COVARIANCE TENSOR

Choosing the coordinate system such that  $\mathbf{e}^1, \mathbf{e}^2$  are unit eigenvectors for the covariance tensor, the covariance tensor is given by

$$\sigma = \sum_{i=1}^2 \left( \frac{(\epsilon^e - \epsilon_1 I)}{\epsilon_1} \mathbf{e}^i \cdot \mathbf{e}^i - h \int_0^1 \frac{1}{(h+z)^2} d\nu_i(z) \right) \mathbf{e}^i \otimes \mathbf{e}^i, \quad (31)$$

where  $\nu_i, i=1, 2$  are the positive measures given by  $\nu_i(z) = \mu(z)\mathbf{e}^i \cdot \mathbf{e}^i$ . From perturbation theory we obtain, as in Ref. 7 the constraints on the moments of  $\nu_i$  given by

$$\begin{aligned} \theta_2 &= \int_0^1 1 d\nu_i(z), \quad i=1, 2, \\ \langle \Gamma \chi_2 \mathbf{e}^i, \mathbf{e}^i \rangle &= \int_0^1 z d\nu_i(z), \quad i=1, 2. \end{aligned} \quad (32)$$

The last constraint that we consider is the one given by the representation formula for the effective dielectric constant, i.e.,

$$\frac{(\epsilon^e - \epsilon_1 I)}{\epsilon_1} \mathbf{e}^i \cdot \mathbf{e}^i = \int_0^1 \frac{1}{h+z} d\nu_i(z), \quad i=1, 2. \quad (33)$$

It is well known that Fourier expansion gives  $\langle \Gamma \chi_2 \mathbf{e}^i, \mathbf{e}^i \rangle = T \mathbf{e}^i \cdot \mathbf{e}^i$ , where  $T$  is given by formula (9). It is easily checked that  $T$  is positive semidefinite with eigenvalues in the interval  $[0, \theta_2(1 - \theta_2)]$  and  $\text{trace}\{T\} = \theta_2(1 - \theta_2)$ . Introducing the probability measures defined by  $\theta_2 p^i = \nu_i$ , the lower bound on the covariance tensor is written as

$$\begin{aligned} \sigma &\geq \sum_{i=1}^2 \left( \frac{(\epsilon^e - \epsilon_1 I)}{\epsilon_1} \mathbf{e}^i \cdot \mathbf{e}^i \right. \\ &\quad \left. - \theta_2 h \sup_{(p^i \in \mathcal{A}^i)} \int_0^1 \frac{1}{(h+z)^2} dp^i(z) \right) \mathbf{e}^i \otimes \mathbf{e}^i, \end{aligned} \quad (34)$$

where the admissible set of probability measures are those with moments given by

$$\mathcal{A}^i = \begin{cases} m_1^i \equiv \frac{T\mathbf{e}^i \cdot \mathbf{e}^i}{\theta_2} = \int_0^1 z dp^i(z), & i=1,2, \\ m_2^i \equiv \frac{(\epsilon^e - \epsilon_1 I)}{\theta_2 \epsilon_1} \mathbf{e}^i \cdot \mathbf{e}^i = \int_0^1 \frac{1}{h+z} dp^i(z), & i=1,2. \end{cases} \quad (35)$$

It is clear that an upper bound is obtained in a similar way by taking the infimum over all probability measures in  $\mathcal{A}^i$ . To compute bounds, we consider the curve  $\mathcal{C}$  given by  $(z, 1/(h+z), 1/(h+z)^2)$  with curve parameter  $0 \leq z \leq 1$ . The convex hull of this curve is given by the set of points  $(c_1, c_2, c_3)$  generated by the moments

$$\begin{aligned} c_1 &= \int_0^1 z dp(z), \\ c_2 &= \int_0^1 \frac{1}{h+z} dp(z), \\ c_3 &= \int_0^1 \frac{1}{(h+z)^2} dp(z), \end{aligned} \quad (36)$$

as one traverses the set of all probability measures  $p$  defined on  $[0,1]$ . We write  $\underline{c} = (c_1, c_2, c_3)$  and denote this set of points by  $co\{\mathcal{C}\}$  and the lower bound becomes

$$\sigma \geq \theta_2 \sum_{i=1}^2 \left( \frac{(\epsilon^e - \epsilon_1 I)}{\epsilon_1} \mathbf{e}^i \cdot \mathbf{e}^i - \theta_2 h \sup_{\substack{c \in co\{\mathcal{C}\} \\ c_1 = m_1^i, c_2 = m_2^i}} \{c_3\} \right) \mathbf{e}^i \otimes \mathbf{e}^i. \quad (37)$$

Thus, to compute the lower bound (and the upper bound) we obtain an explicit representation of the convex hull  $co\{\mathcal{C}\}$  and maximize (minimize)  $c_3$  over it with  $c_1 = m_1^i$  and  $c_2 = m_2^i$ ,  $i=1,2$ . Computation of the convex hull follows from standard methods<sup>9</sup> and is given by all points  $\underline{c}$  satisfying the inequalities

$$\frac{c_2}{h+1} + \frac{\left(c_2 - \frac{1}{h+1}\right)^2}{1 - \frac{c_1+h}{h+1}} \leq c_3 \leq \frac{c_2}{h} - \frac{\left(\frac{1}{h} - c_2\right)^2}{\frac{c_1+h}{h} - 1},$$

and

$$0 \leq c_1 \leq 1. \quad (38)$$

Application of the elementary bounds  $\langle \epsilon^{-1} \rangle^{-1} \leq \epsilon^e \mathbf{e}^i \cdot \mathbf{e}^i \leq \langle \epsilon \rangle$  gives

$$\frac{1}{h+1} \leq \frac{1}{h+1-\theta_2} \leq c_2 \leq \frac{1}{h}. \quad (39)$$

The supremum in Eq. (37) is obtained by setting  $c_1 = m_1^i$  and  $c_2 = m_2^i$ ,  $i=1,2$  in the right-most inequality of Eq. (38) and the lower bound given by Eq. (10) is obtained. Similar arguments using the left-most inequality in Eq. (38) deliver the upper bound (11). For completeness, the computation of  $co\{\mathcal{C}\}$  is given in the Appendix.

Next, we apply the method developed in this article to recover the lower bounds (5), (6), and (7). We observe from Eq. (19) that the representation formula for the mean-square fluctuation can be written as

$$\sigma_{ij} \bar{E}_i \bar{E}_j = (\epsilon_{ij}^e - \epsilon_1 I_{ij}) / \epsilon_1 \bar{E}_i \bar{E}_j - \left( h \int_0^1 \frac{1}{(h+z)^2} d\nu(z) \right) |\bar{\mathbf{E}}|^2. \quad (40)$$

Here,  $\nu$  is the measure given by  $\nu(z) = \mu \mathbf{e} \cdot \mathbf{e}$  where  $\mathbf{e} = \bar{\mathbf{E}} / |\bar{\mathbf{E}}|$ . We substitute for  $(\epsilon_{ij}^e - \epsilon_1 I_{ij}) / \epsilon_1 \bar{E}_i \bar{E}_j$  using the identity

$$(\epsilon_{ij}^e - \epsilon_1 I_{ij}) / \epsilon_1 \bar{E}_i \bar{E}_j = \left( \int_0^1 \frac{1}{h+z} d\nu(z) \right) |\bar{\mathbf{E}}|^2, \quad (41)$$

to obtain

$$\sigma_{ij} \bar{E}_i \bar{E}_j = \left( \int_0^1 \frac{z}{(h+z)^2} d\nu(z) \right) |\bar{\mathbf{E}}|^2. \quad (42)$$

The constraints on  $\nu$  follow from Eq. (32) and are given by

$$\begin{aligned} \theta_2 &= \int_0^1 1 d\nu(z), \\ \bar{r} &= \int_0^1 z d\nu(z), \end{aligned} \quad (43)$$

where  $0 \leq \bar{r} \leq \theta_2(1 - \theta_2)$ . We introduce the probability measure  $p(z)$  defined by  $\theta_2 p(z) = \nu(z)$  and the bounds on the mean-square fluctuation are given by

$$\begin{aligned} \inf_{(p \in \mathcal{A})} \theta_2 \left( \int_0^1 \frac{z}{(h+z)^2} dp(z) \right) |\bar{\mathbf{E}}|^2 \\ \leq \sigma_{ij} \bar{E}_i \bar{E}_j \leq \sup_{(p \in \mathcal{A})} \theta_2 \left( \int_0^1 \frac{z}{(h+z)^2} dp(z) \right) |\bar{\mathbf{E}}|^2, \end{aligned} \quad (44)$$

where the admissible class  $\mathcal{A}$  of probability measures are those with first moment  $\bar{z} = \int_0^1 z dp(z)$  that satisfy  $0 \leq \bar{z} \leq (1 - \theta_2)$ . To compute these bounds we are lead as before to consider the convex hull a curve. Here, the curve is given by  $(z, z/(h+z)^2)$  for  $0 \leq z \leq 1$ . Its convex hull is given by all points  $(b_1, b_2)$  described by

$$\begin{aligned} b_1 &= \int_0^1 z dp(z), \\ b_2 &= \int_0^1 \frac{z}{(h+z)^2} dp(z), \end{aligned} \quad (45)$$

as  $p$  traverses over all probability measures. This is seen to be the convex hull of the graph of the function  $f(z) = z/(h+z)^2$  over the  $z$  axis on  $0 \leq z \leq 1$ . The function  $f(z)$  is strictly positive on  $0 < z < \infty$  with  $f(0) = 0$  and  $\lim_{z \rightarrow \infty} f(z) = 0$ . Moreover,  $f(z)$  has a global maximum over  $[0, \infty)$  at  $z = h$ , with  $f'(z) \geq 0$  for  $z \leq h$  and  $f'(z) \leq 0$  for  $z \geq h$ . Since  $f$  is strictly positive except for  $z = 0$ , we choose  $p \in \mathcal{A}$  of the form  $p(z) = \delta(z)$  and we obtain the desired lower bound given by 0. For  $h \geq (1 - \theta_2)$ , one readily checks that  $b_2 \leq b_1/(h+b_1)^2$  for all points  $(b_1, b_2)$  in the convex hull for which  $b_1 \leq (1 - \theta_2)$ . Thus,  $\int_0^1 f(z) dp(z) \leq f(\bar{z}) \leq f(1 - \theta_2)$ .

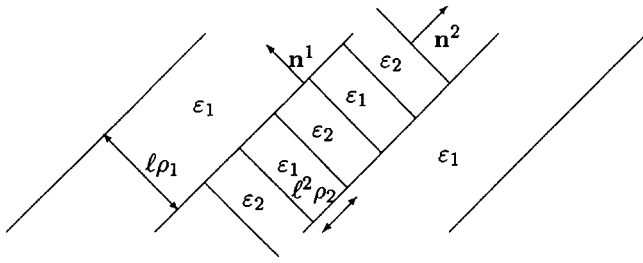


FIG. 1. Laminate of second rank.

For this case, one chooses  $p(z) = \delta(z - (1 - \theta_2))$ , and the upper bounds given by Eq. (6) and the second equation in Eq. (7) are obtained. When  $h \leq (1 - \theta_2)$  it is evident that  $b_2 \leq f(h)$  for all  $b_1$  in  $[0, 1 - \theta_2]$ . Thus, the optimal measure  $p = \delta(z - h)$  is chosen and the upper bound given by the first equation in Eqs. (7) is obtained.

**IV. LAYERED MICROSTRUCTURES AND OPTIMAL LOWER BOUNDS**

In this section, we establish optimality for the lower bounds presented in the Introduction. To do this, we provide explicit formulas for the limit of the covariance tensors for a class of layered materials. These layered materials exhibit fine structure on two scales and are commonly known as “finite-rank laminar microstructures,” see Fig. 1. We introduce the length scale “ $l$ ,” characterizing the scale of the microstructure and examine the limiting behavior as  $l$  tends to zero. The layered material is described in a hierarchical way. We start by layering  $\epsilon_2$  with  $\epsilon_1$  in the proportions  $l^2(1 - \rho_2)$  and  $l^2\rho_2$ , respectively. The layer normals are taken parallel to the direction specified by the unit vector  $\mathbf{n}^2$ . We then take the finely layered material and layer it with material  $\epsilon_1$  at the coarser length scale  $l$ . Here, the layer width of the finely layered material is  $l(1 - \rho_1)$  and the layers of material  $\epsilon_1$  are of width  $l\rho_1$ . The layer direction of the coarse layers is given by the unit vector  $\mathbf{n}^1$ . For future reference, we call this structure a rank 2 laminate with core of material 2. For fixed  $l$  we denote the electric field by  $\mathbf{E}^l$  and the associated covariance tensor by  $\sigma^l$ . The first goal is to provide an explicit formula for  $\sigma = \lim_{l \rightarrow 0} \sigma^l$ . From homogenization theory,<sup>10</sup> the electric field  $\mathbf{E}^l$  admits the decomposition

$$\mathbf{E}^l = \mathbf{P}^l \bar{\mathbf{E}} + \mathbf{z}^l,$$

where the matrix  $\mathbf{P}^l$  is called the corrector matrix and  $\mathbf{z}^l \rightarrow 0$  strongly in mean square.

To expedite the presentation, we introduce characteristic functions to describe the layer geometry. We let  $\chi^1(t)$  be the periodic function on  $[0, 1]$  such that  $\chi^1 = 1$  for  $0 \leq t \leq \rho_1$  and  $\chi^1 = 0$  elsewhere. Similarly, we let  $\chi^2(t)$  be the periodic function on  $[0, 1]$  such that  $\chi^2 = 1$  for  $0 \leq t \leq \rho_2$  and  $\chi^2 = 0$  elsewhere. Then, the layers at the scale  $l$  are described by  $\chi^{1,l} = \chi^1(\mathbf{n}^1 \cdot \mathbf{x}/l)$  and the layers at scale  $l^2$  are described by  $\chi^{2,l} = \chi^2(\mathbf{n}^2 \cdot \mathbf{x}/l^2)$ . We focus now only on laminates with layering directions  $\mathbf{n}^1$  and  $\mathbf{n}^2$  orthogonal to each other. We apply theorem 2.1 of Briane<sup>11</sup> and a straightforward calculation shows that the correctors are of the form

$$\mathbf{P}^l = \chi^{1,l} \mathbf{P}^1 + (1 - \chi^{1,l}) [\chi^{2,l} \mathbf{P}^2 + (1 - \chi^{2,l}) \mathbf{P}^3], \tag{46}$$

where the constant matrices  $\mathbf{P}^1$ ,  $\mathbf{P}^2$ , and  $\mathbf{P}^3$  are given by

$$\mathbf{P}^1 = \mathbf{I} + (1 - \rho_1) \left( \frac{(1 - \rho_2)(\lambda - 1)}{(1 - \rho_1(1 - \rho_2) + \rho_1(1 - \rho_2)\lambda)} \right) \mathbf{n}^1 \otimes \mathbf{n}^1, \tag{47}$$

$$\begin{aligned} \mathbf{P}^2 = & \mathbf{I} - \rho_1 \left( \frac{(1 - \rho_2)(\lambda - 1)}{(1 - \rho_1(1 - \rho_2) + \rho_1(1 - \rho_2)\lambda)} \right) \mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + (1 - \rho_2) \left( \frac{\lambda - 1}{(1 - \rho_2) + \rho_2\lambda} \right) \mathbf{n}^2 \otimes \mathbf{n}^2, \end{aligned} \tag{48}$$

and

$$\begin{aligned} \mathbf{P}^3 = & \mathbf{I} - \rho_1 \left( \frac{(1 - \rho_2)(\lambda - 1)}{(1 - \rho_1(1 - \rho_2) + \rho_1(1 - \rho_2)\lambda)} \right) \mathbf{n}^1 \otimes \mathbf{n}^1 \\ & - \rho_2 \left( \frac{\lambda - 1}{(1 - \rho_2) + \rho_2\lambda} \right) \mathbf{n}^2 \otimes \mathbf{n}^2, \end{aligned} \tag{49}$$

where  $\mathbf{I}$  is the  $2 \times 2$  identity and  $\mathbf{n}^1 \otimes \mathbf{n}^1$  and  $\mathbf{n}^2 \otimes \mathbf{n}^2$  are the rank one matrices  $\mathbf{n}_i^1 \mathbf{n}_j^1$  and  $\mathbf{n}_i^2 \mathbf{n}_j^2$ , respectively.

For  $l$  fixed, the covariance matrix is written

$$\sigma_{ij}^l = \frac{1}{V} \int_{\Omega} (\mathbf{E}^{l,i} - \mathbf{e}^i) \cdot (\mathbf{E}^{l,j} - \mathbf{e}^j) dx, \tag{50}$$

where  $\mathbf{E}^{l,i}$  is the local electric field induced by the constant applied field  $\mathbf{e}^i$ . And  $\mathbf{E}^{l,i} = \mathbf{P}^l \mathbf{e}^i + \mathbf{z}^{l,i}$  where  $\mathbf{z}^{l,i}$  goes to zero in mean square. Next, we observe that because of the separation of scales, the sequence  $(1/V) \int_{\Omega} \chi^{1,l} \chi^{2,l} dx$  converges to the product  $\rho_1 \rho_2$  and that the total area fraction of the  $\epsilon_2$  phase is given by  $\theta_2 = (1 - \rho_1)(1 - \rho_2)$ . Collecting results and taking limits gives

$$\lim_{l \rightarrow 0} \sigma_{ij}^l = \sigma_{ij}, \tag{51}$$

where the matrix  $\sigma_{ij}$  is given by

$$\sigma_{ij} = \int_0^1 f(z) \mu_{ij}(dz), \tag{52}$$

where

$$\begin{aligned} \mu(dz) = & [\theta_2 \delta(z - \rho_1(1 - \rho_2)) \mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + \theta_2 \delta(z - \rho_2) \mathbf{n}^2 \otimes \mathbf{n}^2] dz, \end{aligned} \tag{53}$$

and  $f(z)$  is given by Eq. (8). We also consider the phase-interchanged second rank laminate. Here, material 1 is layered with material 2 at the smallest scale and this structure is layered at the next largest scale with material 2. For this construction, the relative width of the  $\epsilon_1$  layers is  $1 - \rho_2$  and the relative width of the layer containing the layers of  $\epsilon_1$  material is  $1 - \rho_1$ . Here, the total area fraction of material 2 in the composite is  $\theta_2 = \rho_1 + (1 - \rho_1)\rho_2$ . For future reference, we call this structure a rank 2 laminate with a core of material 1. The associated covariance tensor at fixed  $l$  is written  $\bar{\sigma}_{ij}^l$ . A calculation identical to the previous one shows that  $\lim_{l \rightarrow 0} \bar{\sigma}_{ij}^l = \bar{\sigma}_{ij}$ , where

$$\bar{\sigma}_{ij} = \int_0^1 \tilde{f}(z) \bar{\mu}_{ij}(dz), \tag{54}$$



where

$$\begin{aligned} \bar{\mu}(dz) = & [(1 - \theta_2)\delta(z - \rho_1(1 - \rho_2))\mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + (1 - \theta_2)\delta(z - \rho_2)\mathbf{n}^2 \otimes \mathbf{n}^2]dz, \end{aligned} \tag{55}$$

and

$$\tilde{f}(z) = \frac{z}{\left(z + \frac{1}{(1/\lambda) - 1}\right)^2}. \tag{56}$$

Next, we show that  $\sigma_{ij}$  and  $\bar{\sigma}_{ij}$  attain the lower bound (10). This requires that  $\sigma_{ij}$  and  $\bar{\sigma}_{ij}$  be restated in terms of  $\langle \epsilon \rangle$ ,  $T$ , and  $\epsilon^e$ . In what follows, we do this to find that the lower bound (10) matches the formulas for  $\sigma_{ij}$  and  $\bar{\sigma}_{ij}$ . To start, we write the formulas for the well-known effective tensors of rank 2 laminates. The effective property for a rank 2 laminate with a core of material 2<sup>12</sup> is given by

$$\epsilon = \epsilon_1 I + \epsilon_1 \int_0^1 \frac{1}{h+z} d\mu = \epsilon_1 I + \epsilon_1 \theta_2 \left( hI + \frac{1}{\theta_2} T \right)^{-1}, \tag{57}$$

with

$$\int_0^1 z d\mu = T = \theta_2 \rho_1 (1 - \rho_2) \mathbf{n}^1 \otimes \mathbf{n}^1 + \theta_2 \rho_2 \mathbf{n}^2 \otimes \mathbf{n}^2. \tag{58}$$

Similarly, the effective property of a rank 2 laminate with a core of material 1 is given by

$$\begin{aligned} \bar{\epsilon} = & \epsilon_1 I + \epsilon_1 \int_0^1 \frac{1}{h+z} d\bar{\mu} \\ = & \epsilon_2 I - \theta_1 \epsilon_2 \left( \frac{1}{(1-\lambda)^{-1}} I - \frac{1}{\theta_1} \bar{T} \right)^{-1} \end{aligned} \tag{59}$$

and

$$\begin{aligned} \int_0^1 z d\bar{\mu} = & \bar{T} = (1 - \theta_2)\rho_1(1 - \rho_2)\mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + (1 - \theta_2)\rho_2\mathbf{n}^2 \otimes \mathbf{n}^2. \end{aligned} \tag{60}$$

A straightforward substitution of these formulas into the formulas for  $\sigma$  and  $\bar{\sigma}$  gives the required identities

$$\begin{aligned} \sigma = & \frac{(\langle \epsilon \rangle - (\underline{\epsilon} \mathbf{n}^1 \cdot \mathbf{n}^1))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{n}^1 \cdot \mathbf{n}^1)} \mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + \frac{(\langle \epsilon \rangle - (\underline{\epsilon} \mathbf{n}^2 \cdot \mathbf{n}^2))^2}{(\epsilon_2 - \epsilon_1)^2 (T \mathbf{n}^2 \cdot \mathbf{n}^2)} \mathbf{n}^2 \otimes \mathbf{n}^2 \end{aligned} \tag{61}$$

and

$$\begin{aligned} \bar{\sigma} = & \frac{(\langle \epsilon \rangle - (\bar{\epsilon} \mathbf{n}^1 \cdot \mathbf{n}^1))^2}{(\epsilon_2 - \epsilon_1)^2 (\bar{T} \mathbf{n}^1 \cdot \mathbf{n}^1)} \mathbf{n}^1 \otimes \mathbf{n}^1 \\ & + \frac{(\langle \epsilon \rangle - (\bar{\epsilon} \mathbf{n}^2 \cdot \mathbf{n}^2))^2}{(\epsilon_2 - \epsilon_1)^2 (\bar{T} \mathbf{n}^2 \cdot \mathbf{n}^2)} \mathbf{n}^2 \otimes \mathbf{n}^2. \end{aligned} \tag{62}$$

We conclude by showing that the lower bound given by Eq. (17) is the most restrictive for statistically isotropic composites when only the phase area fractions are known. We consider a rank 2 laminate with material 1 as the core. Selecting  $\rho_1$  and  $\rho_2$  such that  $\rho_1(1 - \rho_2) = \theta_2/2$  and  $\rho_2 = \theta_2/2$ , we ob-

tain a composite with an isotropic effective dielectric constant given by  $\bar{\epsilon} = \epsilon_{HS} I$ ,  $\bar{T} = [\theta_2(1 - \theta_2)/2]I$ , and the covariance tensor is

$$\bar{\sigma} = I \frac{2(\langle \epsilon \rangle - \epsilon_{HS})^2}{(\epsilon_2 - \epsilon_1)^2 \theta_2 (1 - \theta_2)}. \tag{63}$$

It now follows that the bound (17) is the best possible given the phase area fractions and the dielectric constants of each phase.

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### APPENDIX

We outline the set of calculations for the derivation of the convex hull  $co\{C\}$  of the curve  $C$  given by  $[z, 1/(h+z), 1/(h+z)^2]$  with curve parameter  $0 \leq z \leq 1$ . In order to bring out the underlying geometry of the problem, we reparametrize the curve setting  $z = t^{-1} - h$  and the curve is given by  $(t^{-1}, t, t^2) - (h, 0, 0)$  with  $a \leq t \leq b$ , where  $a = 1/(h+1)$  and  $b = 1/h$ . The convex hull given by Eq. (36) is written

$$\begin{aligned} c_1 = & \int_a^b t^{-1} d\tilde{p}(t) - h, \\ c_2 = & \int_a^b t d\tilde{p}(t), \\ c_3 = & \int_0^1 t^2 d\tilde{p}(t), \end{aligned} \tag{A1}$$

where  $\tilde{p}$  traverses all probability measures defined on the interval  $[a, b]$ . Writing  $b_1 = \int_a^b t^{-1} d\tilde{p}(t)$ ,  $b_2 = \int_a^b t d\tilde{p}(t)$ , and  $b_3 = \int_a^b t^2 d\tilde{p}(t)$ , we have  $c_1 = b_1 - h$ ,  $c_2 = b_2$ , and  $c_3 = b_3$ . The formula for the convex hull  $co\{C\}$  follows from an explicit formula for the convex hull of the curve  $(t^{-1}, t, t^2)$ ,  $a \leq t \leq b$ . We follow standard procedure and first compute the conic hull of the curve  $(1, t^{-1}, t, t^2)$  for  $a \leq t \leq b$  given by the set of points  $(b_0, b_1, b_2, b_3)$  generated by the moments

$$\begin{aligned} b_0 = & \int_a^b 1 dm(t), \quad b_1 = \int_a^b t^{-1} dm(t), \\ b_2 = & \int_a^b t dm(t), \quad b_3 = \int_a^b t^2 dm(t), \end{aligned} \tag{A2}$$

where  $m$  traverses the set of all positive measures defined on  $[a, b]$ . The convex hull is recovered by taking the intersec-

tion of the conic hull with the hyperplane (1,0,0,0). Following Ref. 9 the dual cone to the curve  $(1, t^{-1}, t, t^2)$ ,  $a \leq t \leq b$  is given by the set of all non-negative polynomials  $q(t) = a_0 + a_{-1}t^{-1} + a_1t + a_2t^2$  on  $[a, b]$ . Noting that  $(1, t^{-1}, t, t^2) = t^{-1}(t, 1, t^2, t^3)$ , we observe that  $q(t) = t^{-1}r(t)$  where  $r(t) = a_0t + a_{-1} + a_1t^2 + a_2t^3$ , thus the set of vectors of coefficients  $(a_0, a_{-1}, a_1, a_2)$  corresponding to all non-negative polynomials  $q(t)$  is precisely the one corresponding to all non-negative polynomials  $r(t)$ . Thus, from the result of Markov and Lukács, given in Ref. 9, all non-negative polynomials of the form  $q(t)$  on  $[a, b]$  are given by

$$q(t) = x_0^2(1 - at^{-1}) + 2x_0x_1(t - a) + x_1^2(t^2 - at) + y_0^2(bt^{-1} - 1) + 2y_0y_1(b - t) + y_1^2(bt - t^2), \tag{A3}$$

where  $(x_0, x_1)$  and  $(y_0, y_1)$  are any two pairs of real numbers. The theory for the moment problem (see Ref. 9, Chap. 3) together with Eq. (A3) shows that the conic hull of the curve  $(1, t^{-1}, t, t^2)$ ,  $a \leq t \leq b$  is given by all  $(b_0, b_1, b_2, b_3)$  for which the quadratic forms

$$x_0^2(b_0 - ab_1) + 2x_0x_1(b_2 - ab_0) + x_1^2(b_3 - ab_2) \geq 0, \tag{A4}$$

$$y_0^2(bb_1 - b_0) + 2y_0y_1(bb_0 - b_2) + y_1^2(bb_2 - b_3) \geq 0,$$

are non-negative. This is equivalent to the constraints on the matrices of the quadratic forms given by

$$\det \begin{pmatrix} b_3 - ab_2 & b_2 - ab_0 \\ b_2 - ab_0 & b_0 - ab_1 \end{pmatrix} \geq 0, \tag{A5}$$

$$\det \begin{pmatrix} bb_2 - b_3 & bb_0 - b_2 \\ bb_0 - b_2 & bb_1 - b_0 \end{pmatrix} \geq 0, \tag{A6}$$

and the inequalities

$$b_0 \geq ab_1, \quad b_3 \geq ab_2, \tag{A7}$$

$$bb_1 \geq b_0, \quad bb_2 \geq b_3.$$

We recover the formulas for the convex hull given in Sec. III [Eq. (38)], upon setting  $b_0 = 1$  and setting  $c_1 + h = b_1$ ,  $c_2 = b_2$ , and  $c_3 = b_3$  in Eqs. (A5), (A6), and (A7).

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