# Intraphase fluctuations in heterogeneous magnetic materials

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The main purpose of homogenization is the determination of the effective behavior (or macroscopic behavior) of heterogeneous materials. Mean fields per phase are generally used in homogenization and represent sufficient information in most cases. However, more information about the field distribution can be necessary, particularly in nonlinear cases. Then, intraphase fluctuations have to be determined. This paper presents a method, based on homogenization tools, for the determination of both estimates and bounds for the intraphase fluctuations. The presented applications deal with magnetic materials and the results obtained with homogenization are compared to those obtained using a finite element modeling. © 2009 American Institute of Physics. [DOI: 10.1063/1.3152789]

### **I. INTRODUCTION**

The determination of the effective properties of heterogeneous materials is a long-standing problem in physics. Unless the microstructure is fully described, the effective properties cannot be exactly determined. Analytical solutions exist for very simple microstructures. For more complex microstructures, finite element modeling is sometimes used to estimate the effective properties.

Homogenization is an alternative modeling approach that enables the determination of bounds or estimates on the effective properties from few pieces of information about the microstructure. Some optimal bounds for the effective properties have been derived.<sup>1,2</sup> Additional information on the effective property function have been studied by Bergman and Milton<sup>3,4</sup> to define more restrictive bounds. For example, from the knowledge of the effective properties for a given set of parameters, improved bounds have been derived by Milton and by Avellaneda.<sup>4,5</sup>

In the case of magnetic materials, homogenization enables the determination of the effective permeability  $\tilde{\mu}$  (permeability of the equivalent homogeneous medium), linking the macroscopic magnetic field  $\overline{\mathbf{H}}$  and the macroscopic magnetic induction  $\overline{\mathbf{B}}$  in the real medium (RM) (see Fig. 1),

$$\overline{\mathbf{B}} = \langle \mathbf{B}(\mathbf{x}) \rangle = \langle \mu(\mathbf{x}) \cdot \mathbf{H}(\mathbf{x}) \rangle = \widetilde{\mu} \cdot \langle \mathbf{H}(\mathbf{x}) \rangle = \widetilde{\mu} \cdot \overline{\mathbf{H}}, \quad (1)$$

where  $\mu(\mathbf{x})$  is the local permeability and  $\mathbf{x}$  the spatial position. The operator  $\langle \cdot \rangle$  represents an averaging operation over the whole volume of the RM [ $\langle \mathbf{H}(\mathbf{x}) \rangle = 1/V \cdot \int_{V} \mathbf{H}(\mathbf{x}) \cdot dV$ , with *V* the volume of the RM].

These techniques generally rely on a mean field approach. Indeed a first homogenization description of the local behavior can be obtained with the mean field per phase. In many cases, this piece of information can be sufficient. For example, in a magnetostriction problem, the local magnetic field has to be determined in order to define the local strain. If the constitutive law is linear (piezomagnetic behavior), the knowledge of the mean magnetic field is sufficient to determine the mean strain. But when the constitutive law is nonlinear, then the mean strain is not directly linked to the mean magnetic field through the constitutive law (see Fig. 2). This is the reason why the determination of intraphase fluctuations is necessary.

The study of the second order moments  $\langle \mathbf{H}(\mathbf{x})^2 \rangle$  is a first approach for the description of field fluctuations. Although particularly important to study some nonlinear effects, these fluctuations have been less intensively studied. Axell<sup>6</sup> derived bounds for second order moments in two-phase materials for isotropic composites. Lipton<sup>7</sup> derived a lower bound for *n*th order moments. Cheng and Torquato<sup>8</sup> studied the field fluctuations in random composites through a finite element model. We propose in that paper a method to derive bounds and estimates for second order moments in the general case and in the particular case of isotropic composites.

In the first part, homogenization techniques are presented. The second part is dedicated to the use of homogenization tools to determine second order moments. The optimality of the derived bounds is discussed. In the last part, an application on biphasic composites is studied and the results obtained with the presented method are compared to the ones obtained from a finite element modeling. An example about the use of second order moments in a magnetostriction problem is finally presented.

## **II. HOMOGENIZATION**

### A. Theory

The distribution of the magnetic field in a heterogeneous material can be obtained by defining a localization operator  $A(\mathbf{x})$  linking the local magnetic field  $\mathbf{H}(\mathbf{x})$  to the macroscopic one  $\mathbf{H}$ ,

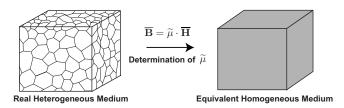


FIG. 1. Homogenization scheme.

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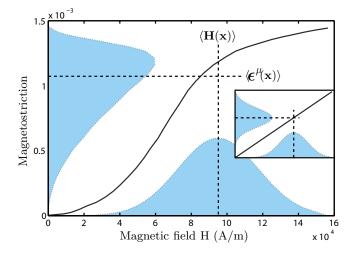


FIG. 2. (Color online) Field distributions and mean values. In the nonlinear case, the relationship between  $\langle \epsilon^{\mu}(\mathbf{x}) \rangle$  and  $\langle \mathbf{H}(\mathbf{x}) \rangle$  is not directly given by the constitutive law.

$$\mathbf{H}(\mathbf{x}) = \mathbf{A}(\mathbf{x}) \cdot \mathbf{H}.$$
 (2)

This localization operator  $A(\mathbf{x})$  depends on the position  $\mathbf{x}$ , on the local permeability  $\mu(\mathbf{x})$ , and on the microstructure of the RM. Moreover, since  $\langle \mathbf{H}(\mathbf{x}) \rangle = \overline{\mathbf{H}}$ , it must verify Eq. (3) (with I the 3 × 3 identity tensor),

$$\langle \mathbf{A}(\mathbf{x}) \rangle = \mathbb{I}. \tag{3}$$

Then, combining Eqs. (1)–(3), the effective permeability  $\tilde{\mu}$  can be written as

$$\widetilde{\boldsymbol{\mu}} = \langle \boldsymbol{\mu}(\mathbf{x}) \cdot \mathbf{A}(\mathbf{x}) \rangle. \tag{4}$$

The homogenization approach classically relies on the determination of the mean local field per phase  $\mathbf{H}_i$ . This mean local field per phase  $\mathbf{H}_i$  can also be linked to the macroscopic one  $\overline{\mathbf{H}}$  [similarly to Eq. (2)],

$$\mathbf{H}_{i} = \langle \mathbf{H}(\mathbf{x}) \rangle_{i} = \langle \mathbf{A}(\mathbf{x}) \cdot \mathbf{H} \rangle_{i} = \langle \mathbf{A}(\mathbf{x}) \rangle_{i} \cdot \mathbf{H} = \mathbf{A}_{i} \cdot \mathbf{H}, \qquad (5)$$

where  $\langle \cdot \rangle_i$  represents an averaging operation over the only phase *i* and  $A_i$  the mean localization operator on phase *i*.

The effective permeability  $\tilde{\mu}$  can be determined (for a *n*-phasic material),

$$\widetilde{\boldsymbol{\mu}} = \langle \boldsymbol{\mu}(\mathbf{x}) \cdot \mathbf{A}(\mathbf{x}) \rangle = \sum_{i=1}^{n} f_i \cdot \boldsymbol{\mu}_i \cdot \mathbf{A}_i \tag{6}$$

since  $\mu(\mathbf{x})$  is constant per phase (considering linear behavior).  $f_i$  stands for the volumetric fraction of phase *i*.

In the particular case of a biphasic material, the localization tensors  $A_i$  can be conversely retrieved from the effective permeability  $\tilde{\mu}$  [using Eqs. (3) and (6)],

$$A_{1} = \frac{1}{f_{1}} (\mu_{1} - \mu_{2})^{-1} \cdot (\tilde{\mu} - \mu_{2}),$$

$$A_{2} = \frac{1}{f_{2}} (\mu_{2} - \mu_{1})^{-1} \cdot (\tilde{\mu} - \mu_{1}).$$
(7)

Therefore, in the biphasic case, the mean field per phase can be estimated (or bounded) from the estimates (or bounds) on the effective permeability  $\tilde{\mu}$ . Many methods can be used to

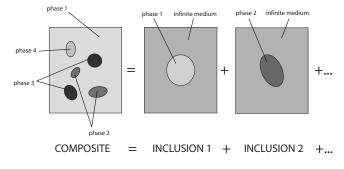


FIG. 3. Principle of homogenization based on inclusion problems.

build these estimates (or bounds). We made the choice to use the homogenization techniques based on inclusion problems.

### **B. Inclusion problems**

Homogenization models based on inclusion problems rely on the hypothesis that the mean field in each phase *i* is similar to the corresponding field in a inclusion (with the same properties  $\mu_i$  than in the RM) embedded in a infinite homogeneous medium with permeability  $\mu_m$ .<sup>9</sup> Then, a *n*-phasic problem can be studied such as *n* uncorrelated inclusion problems (see Fig. 3).

The basic inclusion problem is defined by (i) an inclusion embedded in an infinite medium and (ii) an uniform magnetic field  $\mathbf{H}^{\infty}$  applied at the infinity.

The inclusion shapes are linked to the distribution of the corresponding phases in the RM. For example, an isotropic distribution of one phase in the RM would be modeled by a spherical inclusion in the inclusion problem. It has to be noticed that, in the case of an ellipsoidal inclusion, the field in this inclusion is uniform and can be deduced analytically.<sup>10</sup>

For the sake of simplicity, we will focus on that paper on two dimensional (2D) composites with isotropic constituents (the permeability tensors can be reduced to scalars). In that case, the definition of the effective permeability  $\tilde{\mu}$  is given by

$$\widetilde{\mu} = \frac{\left\langle \frac{\mu_i}{\mu_i + \mu_m} \right\rangle}{\left\langle \frac{1}{\mu_i + \mu_m} \right\rangle}.$$
(8)

The choice of the infinite medium permeability  $\mu_m$  is a degree of freedom in order to describe the microstructure. Some particular choices can lead to classical estimates in homogenization such as the Wiener (W) bounds, the Hashin and Shtrikman (HS) ones, or the self-consistent (SC) estimate.<sup>9</sup>

But homogenization techniques can also provide more information than estimates or bounds on the effective permeability  $\tilde{\mu}$ . Section III presents a method to determine second order moments of the magnetic field per phase.

#### **III. SECOND ORDER MOMENTS**

In a first part, the relation giving the second order moment of the magnetic field per phase  $\langle \mathbf{H}(\mathbf{x})^2 \rangle_i$  is derived. In a

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second part, some results are presented on particular microstructures for which analytical solutions are known. A method that enables to determine bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_i$ , when bounds on the effective permeability  $\tilde{\mu}$  are given, is then presented. The optimality of these bounds is discussed.

#### A. Theory

Homogenization can also be applied to energetic quantities.<sup>4</sup> Equation (9) is verified when uniform boundary conditions are applied (Tellegen's theorem),

$$\langle \mathbf{B}(\mathbf{x}) \cdot \mathbf{H}(\mathbf{x}) \rangle = \langle \mathbf{B}(\mathbf{x}) \rangle \cdot \langle \mathbf{H}(\mathbf{x}) \rangle = \mathbf{B} \cdot \mathbf{H}.$$
(9)

The first member is equal to

$$\langle \mathbf{B}(\mathbf{x}) \cdot \mathbf{H}(\mathbf{x}) \rangle = \frac{1}{V} \int_{V} \mu(\mathbf{x}) \cdot \mathbf{H}(\mathbf{x})^{2} \cdot dV.$$
 (10)

Since the permeability is constant per phase, this equation can be rewritten as

$$\langle \mathbf{B}(\mathbf{x}) \cdot \mathbf{H}(\mathbf{x}) \rangle = \sum_{i=1}^{n} f_i \cdot \mu_i \cdot \langle \mathbf{H}(\mathbf{x})^2 \rangle_i.$$
 (11)

The last member in Eq. (9) can be rewritten with the introduction of the (isotropic) effective permeability  $\tilde{\mu}$ ,

$$\mathbf{B} \cdot \mathbf{H} = (\tilde{\boldsymbol{\mu}} \cdot \mathbf{H}) \cdot \mathbf{H} = \tilde{\boldsymbol{\mu}} \cdot \mathbf{H}^2.$$
(12)

Finally, Eq. (9) leads to

$$\widetilde{\boldsymbol{\mu}} \cdot \overline{\mathbf{H}}^2 = \sum_{i=1}^n f_i \cdot \boldsymbol{\mu}_i \cdot \langle \mathbf{H}(\mathbf{x})^2 \rangle_i.$$
(13)

Considering that the effective applied magnetic field **H** is a constant field, the last equation can be differentiated as

$$(\tilde{\boldsymbol{\mu}} + \delta \tilde{\boldsymbol{\mu}}) \cdot \overline{\mathbf{H}}^2 = \sum_{i=1}^n f_i \cdot (\boldsymbol{\mu}_i + \delta \boldsymbol{\mu}_i) \cdot \langle (\mathbf{H}(\mathbf{x}) + \delta \mathbf{H}(\mathbf{x}))^2 \rangle_i.$$
(14)

Moreover, the use of a variational principle indicates that the second term in the second member of the following equation is equal to zero:

$$\sum_{i=1}^{n} f_{i} \cdot \mu_{i} \cdot \langle (\mathbf{H}(\mathbf{x}) + \delta \mathbf{H}(\mathbf{x}))^{2} \rangle_{i} = \sum_{i=1}^{n} f_{i} \cdot \mu_{i} \cdot \langle \mathbf{H}(\mathbf{x})^{2} \rangle_{i}$$
$$+ 2\sum_{i=1}^{n} f_{i} \cdot \mu_{i} \cdot \langle \mathbf{H}(\mathbf{x}) \cdot \delta \mathbf{H}(\mathbf{x}) \rangle_{i} + \sum_{i=1}^{n} f_{i} \cdot \mu_{i} \cdot \langle \delta \mathbf{H}(\mathbf{x})^{2} \rangle_{i}$$
(15)

since the magnetic field minimizes the energy for a given configuration (configuration made of permeabilities  $\mu_i$ ). Moreover, the last term in the second member can be neglected (second order term).

Using Eq. (14) with Eq. (15) gives

$$(\widetilde{\boldsymbol{\mu}} + \delta\widetilde{\boldsymbol{\mu}}) \cdot \overline{\mathbf{H}}^2 = \sum_{i=1}^n f_i \cdot \boldsymbol{\mu}_i \cdot \langle \mathbf{H}(\mathbf{x})^2 \rangle_i + \sum_{i=1}^n f_i \cdot \delta\boldsymbol{\mu}_i \cdot \langle (\mathbf{H}(\mathbf{x}) + \delta\mathbf{H}(\mathbf{x}))^2 \rangle_i.$$
 (16)

The second term in the second member can be simplified keeping only the first order term and neglecting second and third order terms,

$$\sum_{i=1}^{n} f_{i} \cdot \delta \mu_{i} \cdot \langle (\mathbf{H}(\mathbf{x}) + \delta \mathbf{H}(\mathbf{x}))^{2} \rangle_{i} \approx \sum_{i=1}^{n} f \cdot \delta \mu_{i} \cdot \langle \mathbf{H}(\mathbf{x})^{2} \rangle_{i}.$$
(17)

Then, using Eq. (16) with Eqs. (11) and (17) gives the following relation:

$$\delta \widetilde{\boldsymbol{\mu}} \cdot \overline{\mathbf{H}}^2 = \sum_{i=1}^n f_i \cdot \delta \boldsymbol{\mu}_i \cdot \langle \mathbf{H}(\mathbf{x})^2 \rangle_i, \tag{18}$$

which provides us the following equation to determine the second order moment of the magnetic field per phase:

$$\langle \mathbf{H}(\mathbf{x})^2 \rangle_i = \frac{1}{f_i} \frac{\partial \tilde{\mu}}{\partial \mu_i} \overline{\mathbf{H}}^2.$$
 (19)

This relation was derived earlier by Bergman.<sup>3</sup>

#### **B. Exact solutions**

For some particular microstructures, analytical solutions for the field distribution exist, as well as for the effective permeability.

The most simple example is the study of laminated composites. The magnetic field  $\mathbf{H}(\mathbf{x})$  is uniform per phase. The Wiener bounds are exact estimates for the effective permeability  $\tilde{\mu}$ . The first case is obtained when the laminate direction is parallel to the field, then the magnetic field is uniform in the whole composite ( $\mathbf{H}(\mathbf{x})=\overline{\mathbf{H}}$ ). The effective permeability is equal to

$$\widetilde{\mu} = \sum_{i=1}^{n} f_i \cdot \mu_i.$$
<sup>(20)</sup>

Applying Eq. (19) leads to

$$\langle \mathbf{H}(\mathbf{x})^2 \rangle_i = \overline{\mathbf{H}}^2 = \langle \mathbf{H}(\mathbf{x}) \rangle_i^2,$$
 (21)

which is consistent with the fact that the magnetic field is uniform in the composite.

The second case is obtained when the laminate direction is perpendicular to the magnetic field, the magnetic induction is uniform in the whole composite  $(\mathbf{B}(\mathbf{x})=\overline{\mathbf{B}})$ . The effective permeability is given by

$$\frac{1}{\tilde{\mu}} = \sum_{i=1}^{n} \frac{f_i}{\mu_i}.$$
(22)

Applying Eq. (19) leads to

12

$$\langle \mathbf{B}(\mathbf{x})^2 \rangle_i = \overline{\mathbf{B}}^2 = \langle \mathbf{B}(\mathbf{x}) \rangle_i^2,$$
 (23)

which is consistent again.

Another example of microstructure with analytical solution is the Hashin cylinders (or spheres) assemblage.<sup>2,11</sup> The

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composite is made of composite cylinders consisting of an inner cylinder (phase 2, volumetric fraction  $f_2$ ) embedded in a concentric outer cylinder (phase 1, volumetric fraction  $f_1 = 1 - f_2$ ). The whole space is filled with such composite cylinders sized down to the infinitesimally small. In that case, the magnetic field in phase 2 is uniform. The effective permeability is exactly equal to 1 of the Hashin and Shtrikman bound,

$$\widetilde{\mu} = \mu_1 \cdot \frac{(1 - f_2)\mu_1 + (1 + f_2)\mu_2}{(1 + f_2)\mu_1 + (1 - f_2)\mu_2}.$$
(24)

Applying Eq. (19) leads to

$$\langle \mathbf{H}(\mathbf{x})^2 \rangle_1 = \frac{(\mu_2 + \mu_1)^2 + f_2(\mu_2 - \mu_1)^2}{((1 + f_2)\mu_1 + (1 - f_2)\mu_2)^2} \cdot \overline{\mathbf{H}}^2,$$
  
$$\langle \mathbf{H}(\mathbf{x})^2 \rangle_2 = \frac{4\mu_1^2}{((1 + f_2)\mu_1 + (1 - f_2)\mu_2)^2} \cdot \overline{\mathbf{H}}^2 = \langle \mathbf{H}(\mathbf{x}) \rangle_2^2.$$
(25)

The same relations can be extracted from the analytical field distribution given in Ref. 11.

### C. Bounds on $\partial \tilde{\mu} / \partial \mu_i$

In the general case, the microstructure of materials is not exactly described, so that exact effective permeability cannot be given. Estimates can be built but bounds on the effective permeability can be preferred. Nevertheless, bounds on the effective permeability  $\tilde{\mu}$  do not provide any information about the derivative  $\partial \tilde{\mu} / \partial \mu_i$ . However, more refined bounds on  $\tilde{\mu}$  can be obtained from an additional piece of information and will help us to bound the derivative  $\partial \tilde{\mu} / \partial \mu_i$ . This point is the object of this section.

Let us suppose that the effective permeability is known for a given configuration (obtained from experimentation, for example). Then, for the same fixed microstructure and changing the phase properties (permeabilities), the effective permeability may vary. From the information on the effective permeability for this particular configuration, more restrictive bounds on the effective permeability  $\tilde{\mu}$  can be obtained for the same microstructure<sup>4</sup> (see Fig. 4).

For example, some bounds (Wiener ones, Hashin and Shtrikman ones, etc.) can be determined for a composite if the phase permeabilities are known as well as the volumetric fractions  $f_i$  (an additional assumption could be isotropy). But, from an additional information, more restrictive bounds derived from the previous bounds can be written as the ratio of two polynomials. The following example is derived from Wiener bounds with a known value of the function  $\tilde{\mu}(\mu_i^*, \mu_2^*) = \tilde{\mu}^*$ :

$$\widetilde{\mu}_{1}(\mu_{1},\mu_{2}) = \frac{a_{0}\mu_{2}^{2} + a_{1}\mu_{2}\mu_{1}}{\mu_{2} + b_{1}\mu_{1}},$$

$$\widetilde{\mu}_{2}(\mu_{1},\mu_{2}) = \frac{a_{1}'\mu_{2}\mu_{1} + a_{2}'\mu_{1}^{2}}{\mu_{2} + b_{1}'\mu_{1}},$$
(26)

where the  $a_i$  and  $b_i$  coefficients depend on the previous bounds and on the additional information.<sup>4</sup> For example, in



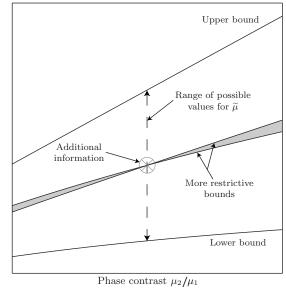


FIG. 4. More restrictive bounds from an additional information about  $\tilde{\mu}$ .

this particular case, the coefficients are determined so that the effective permeability verifies:  $\tilde{\mu}(1,1)=1$ ,  $\partial \tilde{\mu} / \partial \mu_1|_{\mu_1=\mu_2=1}=f_1$ , and  $\tilde{\mu}(\mu_1^*,\mu_2^*)=\tilde{\mu}^*$ .

Until that point, more restrictive bounds have been obtained for the effective permeability  $\tilde{\mu}$  but no information about the derivative  $\partial \tilde{\mu} / \partial \mu_i$  is given. The noticeable point in Fig. 4 is that the new bounds (logically) cross. Then, the derivative  $\partial \tilde{\mu} / \partial \mu_i$  is necessarily bounded by the derivatives of the new bounds at that point. In that way, when bounds are provided on the effective permeability, it is possible to determine some bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_i$  by scanning the full scale of possible values for  $\tilde{\mu}$  and take these values for the additional information. Then, bounds for the derivative  $\partial \tilde{\mu} / \partial \mu_i$  are obtained for each possible value of  $\tilde{\mu}$ . The optimality of such bounds will be discussed in the next paragraph.

### **D.** Optimality

The optimality of the bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_i$ obtained from the proposed method is proven by the definition of a microstructure attaining these bounds. The bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_i$  obtained from the Wiener bounds on the effective permeability  $\tilde{\mu}$  are attained for the confocalellipsoid assemblage defined in Ref. 7.

In the case of 2D isotropic composites, an additional property of the effective permeability can be used, known as the Keller's relation,<sup>12</sup>

$$\widetilde{\mu}(\mu_1,\mu_2)\cdot\widetilde{\mu}(\mu_2,\mu_1)=\mu_1\cdot\mu_2. \tag{27}$$

Then, using this additional information for isotropic 2D composites, bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_i$  obtained from the Hashin and Shtrikman bounds on the effective permeability  $\tilde{\mu}$  are attained for the composite consisting of densely packed, doubly coated circular cylinders defined in Ref. 13.

Unfortunately, for isotropic three dimensional (3D) composites, the Keller relation does not apply since some 3D

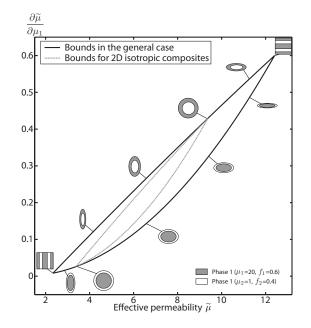


FIG. 5. Bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_1$  as a function of the effective permeability  $\tilde{\mu}$  in the general case and in the case of 2D isotropic composites. The bounds are optimal since they are attained for some particular microstructures.

isotropic composites have been shown not to respect Keller's relation.<sup>14</sup> The relation becomes an inequality,

$$\widetilde{\mu}(\mu_1,\mu_2)\cdot\widetilde{\mu}(\mu_2,\mu_1) \ge \mu_1\cdot\mu_2.$$
(28)

Figures 5 and 6 present the derivative  $\partial \tilde{\mu} / \partial \mu_i$  as the function of the effective permeability  $\tilde{\mu}$ . The phase properties are  $\mu_1=20$ ,  $\mu_2=1$ , and  $f_1=0.6$ .

The corresponding microstructures attaining the bounds are also presented in the figures. These microstructures show a continuous way to transform, for a given composition, a "lower Wiener" microstructure into an "upper Wiener" one

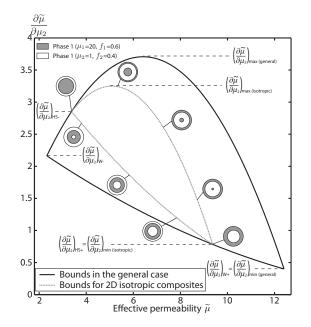


FIG. 6. Bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_2$  as a function of the effective permeability  $\tilde{\mu}$  in the general case and in the case of 2D isotropic composites. The bounds are optimal since they are attained for particular microstructures.

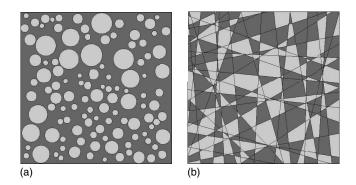


FIG. 7. Microstructures modeled with the finite element method.

passing through Hashin and Shtrikman microstructures, using confocal-ellipsoid assemblage (Fig. 5). Such a transformation can also be performed for isotropic composites using doubly coated circular cylinders (Fig. 6). These continuous transformations could also be applied, for a given effective permeability, with a variation of volumetric fraction  $f_1$ . A similar proposal, using an assemblage of two types of Hashin spheres, has been made by Gilormini<sup>15</sup> for the effective bulk modulus and conductivity of isotropic two-phase composite.

### **IV. RESULTS**

In order to validate the presented method, results are compared to the ones obtained from a finite element modeling. The modeled materials are 2D biphasic magnetic materials with a phase contrast  $\mu_2/\mu_1$  equal to 5. The finite element modeling models different types of microstructures (phase 1/phase 2): (i) type I: inclusions/matrix and matrix/ inclusions [Fig. 7(a)]; (ii) type II: mosaic microstructure [Fig. 7(b)].

For these two types of microstructure, the finite element modeling has been realized for a large number of random microstructures (500 per volumetric fraction and per microstructure type, random position and radius for the disks in the first type, and random allocation of the phases in the second type).

#### A. Effective permeability

Figure 8 shows the results obtained from homogenization and finite element modeling about the effective permeability  $\tilde{\mu}$  as a function of the volumetric fraction  $f_1$  of phase 1.

Results for the mosaic microstructure are presented with error bars because the variability of the effective permeability  $\tilde{\mu}$  is quite important. On the opposite, results for the matrix/inclusions (and inclusions/matrix) are presented with points representing the mean value of the different computations because the variability of the effective permeability  $\tilde{\mu}$ is very low for this type of microstructure.

Classical estimates in homogenization are presented too. The Wiener bounds are obtained by choosing  $\mu_m = 0$  and  $\mu_m \rightarrow \infty$  in the inclusion problems. The more restrictive HS bounds for isotropic composites are obtained by choosing  $\mu_m = \mu_1$  and  $\mu_m = \mu_2$  in the inclusion problems. The SC estimate can be a relevant estimate for microstructures where

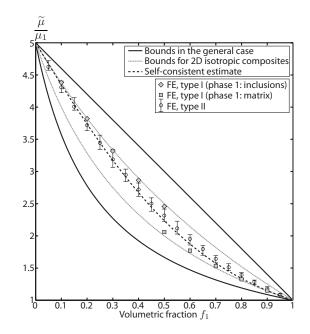


FIG. 8. Effective permeability  $\tilde{\mu}$  as a function of the volumetric fraction of phase 1.

none of the phase plays a particular role. They are obtained by choosing  $\mu_m = \tilde{\mu}$  in the inclusion problems, leading to an iterative computation.

As expected, the finite element results are bounded by the Wiener bounds. Since the studied finite element microstructures can be considered isotropic, HS estimates should be bounds for the finite element results. A very few number of computations do not remain between these bounds, the isotropy of the corresponding microstructures may not be verified. In the following, the HS estimates will be considered as bounds. The first comment about Fig. 8 is that the effective permeability for the matrix/inclusions (and also inclusions/matrix) microstructures is quite close to the HS bounds. The HS bounds seem to be appropriate to give a quite accurate estimate for matrix/inclusion problems. The second comment is that the variability for the mosaic microstructure is quite high but the mean value for the effective permeability seems to be correctly estimated by the selfconsistent estimate. This estimate seems well suited to this type of microstructure.

#### B. Derivatives of the effective permeability

Figure 9 shows the derivative of the effective permeability  $\partial \tilde{\mu} / \partial \mu_1$  [determined in the finite element modeling by computing the value  $f_1 \langle \mathbf{H}(\mathbf{x})^2 \rangle_1 / \overline{\mathbf{H}}^2$  according to Eq. (19)] as a function of the volumetric fraction  $f_1$  of phase 1 for a given phase contrast  $\mu_2 / \mu_1$  equal to 5. Figure 10 presents the same results for a contrast equal to 100. The homogenization results are given with two types of estimates: (i) estimates of  $\partial \tilde{\mu} / \partial \mu_1$  obtained directly by deriving the estimates of  $\tilde{\mu}$  (W and HS bounds, SC estimate), (ii) and bounds on  $\partial \tilde{\mu} / \partial \mu_1$ obtained by scanning the range of possible values of  $\tilde{\mu}$  (according to the bounds on the effective permeability  $\tilde{\mu}$ , see Fig. 8) and taking the minimum and maximum values of the derivative computed from Eq. (26) according to the method described in Sec. III C.

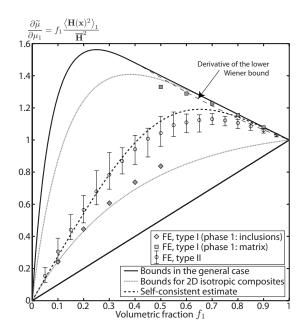


FIG. 9. Derivative of the effective permeability  $\partial \tilde{\mu}/\partial \mu_1$  as a function of the volumetric fraction  $f_1$  of phase 1, contrast  $\mu_2/\mu_1=5$  (same legend for finite element results than Fig. 8).

In one hand, these results illustrate that the derivatives of the previous effective permeability bounds cannot be considered as bounds on the derivative  $\partial \tilde{\mu} / \partial \mu_1$ . Some finite element results bypass these estimates (for example, some finite element results for the type I microstructures bypass the derivative  $\partial \tilde{\mu} / \partial \mu_1$  of the W bound).

On the other hand, the finite element results for the type II microstructure seem to fit with the SC estimate (derivative of the SC estimate of  $\tilde{\mu}$ ). For type I microstructures, the results are close to the bounds for isotropic composites for the contrast 5, whereas it is not really the case with the contrast 100 (at least, for the upper bound). This is due to the

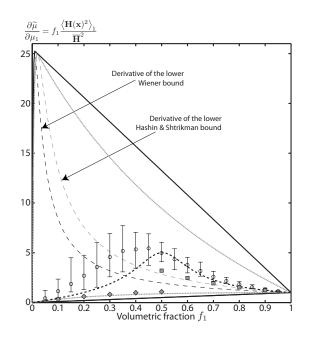


FIG. 10. Derivative of the effective permeability  $\partial \tilde{\mu}/\partial \mu_1$  as a function of the volumetric fraction  $f_1$  of phase 1, contrast  $\mu_2/\mu_1=100$  (same legend than Fig. 9).

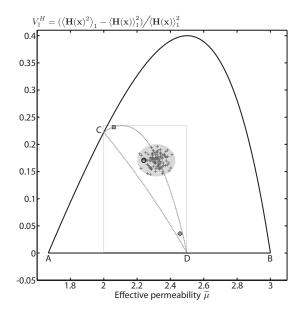


FIG. 11. Variance of the magnetic field  $\mathbf{H}(\mathbf{x})$  in phase 1 as a function of the effective permeability  $\tilde{\mu}$ , contrast  $\mu_2/\mu_1=5$  (see legend in Fig. 12).

fact that the maximum value for  $\partial \tilde{\mu} / \partial \mu_1$  (depending on  $\tilde{\mu}$ ) is quite high compared to its value when taking  $\tilde{\mu}$  equal to the HS limit. This point appears clearly in Fig. 6.

It also appears in these figures, that in the case of a type I microstructure, the differentiation of HS estimate provides a satisfying approximation of the second order moment.

It must be noticed that the lower bounds for  $\partial \tilde{\mu} / \partial \mu_1$ (both in the general case and isotropic composites) are exactly equal to the derivatives of the Wiener and Hashin and Shtrikman for  $\tilde{\mu}$  (in that example with contrast  $\mu_2 / \mu_1$  higher than 1, it is equal to the derivatives of the upper bounds).

The bounds for the derivative  $\partial \tilde{\mu} / \partial \mu_1$ , for a given volumetric fraction  $f_1$ , are large due to the loss of the estimation of  $\tilde{\mu}$ .

### C. Variance

It is convenient to present the evolution of the variance as a function of  $\tilde{\mu}$ . The variance of the magnetic field in phase *i* is defined as

$$V_i^H = \frac{\langle \mathbf{H}(\mathbf{x})^2 \rangle_i - \langle \mathbf{H}(\mathbf{x}) \rangle_i^2}{\langle \mathbf{H}(\mathbf{x}) \rangle_i^2}.$$
(29)

Figure 11 shows the variance of the magnetic field in phase 1 as a function of the effective permeability  $\tilde{\mu}$  in the particular case of a volumetric fraction of phase 1 equal to  $f_1=0.5$  and contrast  $\mu_2/\mu_1=5$ . Figure 12 is the variance of the magnetic field in phase 2 with contrast  $\mu_2/\mu_1=100$  (volumetric fraction is still  $f_1=0.5$ ). The end points A and B, respectively, correspond to the lower and upper Wiener bounds on the effective permeability, and the variance of the magnetic field (in both phases) is equal to zero in these cases. The end points C and D, respectively, correspond to the effective permeability; only one of the phases presents a zero variance for the magnetic field in these cases.

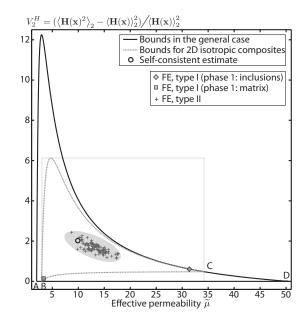


FIG. 12. Variance of the magnetic field  $\mathbf{H}(\mathbf{x})$  in phase 2 as a function of the effective permeability  $\tilde{\mu}$ , contrast  $\mu_2/\mu_1=100$ .

The value  $\langle \mathbf{H}(\mathbf{x})^2 \rangle_i$  is computed from Eqs. (19) and (26) and the value  $\langle \mathbf{H}(\mathbf{x}) \rangle_i^2$  is computed from Eq. (7).

These figures (particularly Fig. 12) show that taking minimum and maximum values for the derivative  $\partial \tilde{\mu} / \partial \mu_i$  when scanning the range of possible values of  $\tilde{\mu}$  is not judicious if the information about  $\tilde{\mu}$  is known, since this information is lost. The corresponding bounds would be rectangles on these figures. It would be preferable to present bounds as a function of the volumetric fraction *f* and of the effective permeability  $\tilde{\mu}$ ; but this kind of 3D curves cannot be presented easily.

The finite element results for type I microstructures show a low variability in variance and effective permeability. Moreover, the results are very close to the C and D end points, corresponding to HS estimates for the effective permeability and the use of the derivative  $\partial \tilde{\mu} / \partial \mu_1$  of the HS bounds for the determination of the second order moment  $\langle \mathbf{H}(\mathbf{x})^2 \rangle_1$ . The SC estimate also seems to be a good estimate for type II microstructure.

### D. Application

An example of application is presented in this paragraph. It deals with magnetostriction, including the nonlinear relationship between the magnetic field and the magnetostrictive strain. A classical model<sup>16</sup> gives the magnetostrictive strain  $\epsilon^{\mu}(\mathbf{x})$  as a function of the magnetic induction  $\mathbf{B}(\mathbf{x})$ ,

$$\boldsymbol{\epsilon}^{\mu}(\mathbf{x}) = \boldsymbol{\alpha} \cdot \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1/2 & 0 \\ 0 & 0 & -1/2 \end{pmatrix} \cdot \mathbf{B}(\mathbf{x})^2 = \underline{\boldsymbol{\alpha}} \cdot \mathbf{B}(\mathbf{x})^2.$$
(30)

Let us study a composite made of two magnetostrictive phases. The two phases have different permeabilities  $\mu_i$  and different magnetostriction coefficients  $\alpha_i$ . Then, the macroscopic magnetostrictive strain  $\overline{\epsilon^{\mu}}$  can be deduced from the magnetic state in the composite (the composite is supposed to be elastically homogeneous),

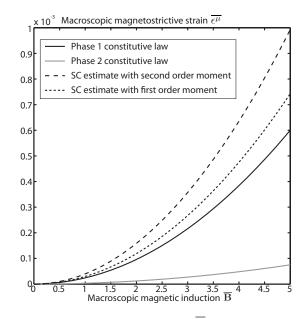


FIG. 13. Macroscopic magnetostrictive strain  $\overline{\epsilon^{\mu}}$  as a function of the macroscopic magnetic induction  $\overline{\mathbf{B}}$ .

$$\boldsymbol{\epsilon}^{\mu} = \langle \boldsymbol{\epsilon}^{\mu}(\mathbf{x}) \rangle = f_1 \cdot \alpha_1 \cdot \langle \mathbf{B}(\mathbf{x})^2 \rangle_1 + f_2 \cdot \alpha_2 \cdot \langle \mathbf{B}(\mathbf{x})^2 \rangle_2. \quad (31)$$

A classical approximation is the mean field approach, which assimilates the second order moments as the square of the first order moments  $\langle \mathbf{H}(\mathbf{x})^2 \rangle_i \rightarrow \langle \mathbf{H}(\mathbf{x}) \rangle_i^2$ , which is generally wrong (underestimation of the second order moment).

A better estimate can be obtained with the determination of second order moments. These second order moments can be computed from Eq. (19) and  $\langle \mathbf{B}(\mathbf{x})^2 \rangle_i = \mu_i^2 \cdot \langle \mathbf{H}(\mathbf{x})^2 \rangle_i$ .

Figure 13 presents the macroscopic magnetostrictive strain  $\epsilon^{\mu}$  as a function of the macroscopic magnetic induction  $\mathbf{B}$  for the two phases and an estimate for the composite. The phase properties are  $\mu_1/\mu_2=15$ ,  $\alpha_1=24\times10^{-4}$ ,  $\alpha_2=3\times10^{-4}$ , and  $f_1=0.7$ . We can see on this figure that approximating the second order moments as squares of first order moments underestimates the macroscopic strain for about 25% in this example. By the way, it is interesting to notice that the composite presents a higher magnetostriction than each of the two constituents (because of the concavity of the constitutive law [Eq. (30)].

### **V. CONCLUSION**

Homogenization techniques can provide estimates or bounds for the effective behavior of composite materials with very low computational cost compared to the finite element method. In addition to mean field per phase, it can also provide estimates or bounds on the intraphase fluctuations. Such information can be decisive in the case of nonlinear behavior. In the example dealing with a magnetostriction model presented in this paper, the magnetostrictive strain can be about 25% underestimated when using only mean field approach, compared to the case introducing field fluctuations.

We presented in this paper a method to determine the intraphase fluctuations in heterogeneous materials using homogenization tools. The results have been compared to those obtained from a finite element modeling. It has been shown that the self-consistent estimate provides a satisfying approximation of both mean field and second order moment, for mosaic microstructure, where none of the phase plays a specific role. In the case of matrix/inclusion microstructure, the Hashin and Shtrikman estimate is well suited for both mean field and second order moment. If bounds rather than estimates are needed, we defined optimal bounds for the second order moments in the general case and more restrictive ones in the particular case of isotropic 2D composites.

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