HOMOGENIZATION OF SPHERICAL INCLUSIONS

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Abstract—The homogenization of cubically arranged, homogeneous spherical inclusions in a background material is addressed. This is accomplished by the solution of a local problem in the unit cell. An exact series representation of the effective relative permittivity of the heterogeneous material is derived, and the functional behavior for small radii of the spheres is given. The solution is utilizing the translation properties of the solutions to the Laplace equation in spherical coordinates. A comparison with the classical mixture formulas, e.g., the Maxwell Garnett formula, the Bruggeman formula, and the Rayleigh formula, shows that all classical mixture formulas are correct to the first (dipole) order, and, moreover, that the Maxwell Garnett formula predicts several higher order terms correctly. The solution is in agreement with the Hashin-Shtrikman limits.

Appendix A. The Spherical Coordinate Solutions

Appendix B. Evaluation of Some Integrals

Appendix C. The Translation Matrices $P_{nn'}$

Appendix D. The Sum $S_{nn'}$

References

1. INTRODUCTION

The electromagnetic homogenization problem of materials is to find the macroscopic electromagnetic response of a material with a microscopic structure. If the microscopic structure is periodic with periodicity ϵ, the homogenization is to find the effective material parameters as the periodicity $\epsilon \to 0$, see Figure 1. The unit cell of the problem with periodicity ϵ is denoted the Y^ϵ-cell, see Figure 1.
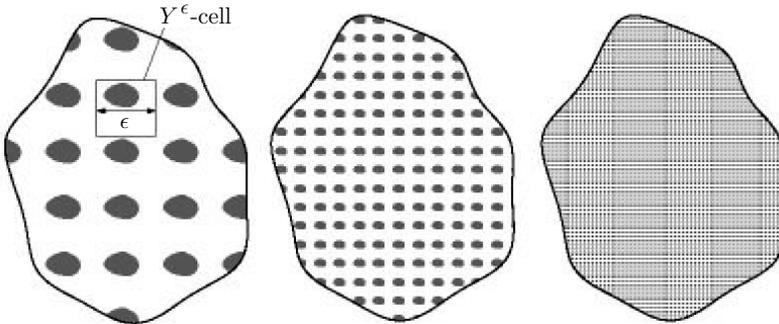
Figure 1. A material with a microstructure with periodicity ϵ and the Y^ϵ-cell. The size of the periodicity ϵ is decreasing from left to right.

This homogenization problems is well studied in the mathematical literature, see e.g., [1, 2, 4, 7, 15, 18] for excellent reviews on the subject. Recent advances in the field of two-scaled convergence [12] have proven valuable in this context. The generic problem is an electrostatic problem (local problem) in the reference unit cell Y with sides of unit length. The unit cell Y^ϵ-cell is the Y-cell scaled by ϵ. Usually the problem is formulated in the weak sense and a solution of the problem is sought by the means of the finite element method (FEM).

In this paper, we are concerned with the homogenization problem of a material consisting of two different isotropic materials (two-phase material). More precisely, our problem is to find the effective relative

permittivity for material consisting of homogeneous, isotropic spherical inclusions in a homogeneous, isotropic background material. We limit ourselves to a cubic lattice, but the method developed in this paper has potential also in other lattice configurations. The series solution is obtained by the use of the translation properties of the solutions to the Laplace equation in spherical coordinates. These translation matrices provide an excellent tool in finding the solution of the local problem, and they are reviewed in an appendix.

The solution of the local problem seems not to be have solved with the technique presented in this paper before, but the problem has been addressed in the literature, see e.g., [7, p. 45]. Also, the early results by Lord Rayleigh [13] are relevant. However, Lord Rayleigh does not solve the local problem in the form of the present paper, but uses a more physical point of attack.

Although simple in its geometry, the type of material addressed in this paper is important in many applications, e.g., glass micro balloon material in radome constructions. An excellent expose of other important applications with many illustrations is given by Sihvola [16].

2. THE LOCAL PROBLEM

In this section, we give a short review of the homogenization of a material with microstructure. For simplicity, we only treat the case of isotropic permittivity. The more general problem of homogenizing an anisotropic material is presented in e.g., [4, 19] or in the references cited in the Section 1.

The relative permittivity $\varepsilon(\boldsymbol{y})$ is Y-periodic and belongs to $L^\infty(Y)$, where $Y = (0,1)^3$, i.e., $\varepsilon(\boldsymbol{y})$ is measurable and bounded a.e. on Y, and $\varepsilon(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) = \varepsilon(\boldsymbol{y})$ for every $\boldsymbol{y} \in \mathbb{R}^3$, $i = 1,2,3$, where $\hat{\boldsymbol{e}}_i$, $i = 1,2,3$, are the Cartesian basis vectors in $\mathbb{R}^3$. This assumption assumes that the inhomogeneities are arranged in a cubic lattice[†]. Moreover, the material is assumed non-magnetic, i.e., the relative permeability $\mu = 1$, in this paper.

The homogenized permittivity of a periodic structure with a periodicity that approaches zero relies on the solution of a local boundary value problem in the unit cell Y. This local problem is to find weak solutions $\chi_j(\boldsymbol{y}) \in H^1_\#(Y)$ (Sobolev space with one weak

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[†] The more general case $Y = (0, a_1) \times (0, a_2) \times (0, a_3)$, where $a_i > 0$, and $\varepsilon(\boldsymbol{y} + a_i \hat{\boldsymbol{e}}_i) = \varepsilon(\boldsymbol{y})$ for every $\boldsymbol{y} \in \mathbb{R}^3$, $i = 1,2,3$, can be solved with a similar technique.

derivative and Y-periodic[‡]) for $j = 1, 2, 3$, satisfying [4, 19]

$$\iiint\limits_{Y} \nabla w(\boldsymbol{y}) \cdot (\varepsilon(\boldsymbol{y})\hat{\boldsymbol{e}}_j - \varepsilon(\boldsymbol{y})\nabla\chi_j(\boldsymbol{y})) \, \mathrm{d}v = 0, \quad \forall w \in H^1_{\#}(Y) \quad (1)$$

The volume measure of $\mathbb{R}^3$ is denoted $\mathrm{d}v$. The homogenized relative permittivity, ε^h_{ij}, is then fund as an appropriate average of the solution of (1) and the permittivity $\varepsilon(\boldsymbol{y})$. The result is [4, 19]

$$\varepsilon^h_{ij} = <\varepsilon(\boldsymbol{y})>\delta_{ij} - <\varepsilon(\boldsymbol{y})\frac{\partial}{\partial y_i}\chi_j(\boldsymbol{y})> \quad (2)$$

where the average over the Y-cell is defined as (the volume of the unit cell, $|Y|$, is in our case chosen so that $|Y| = 1$)

$$<f> = \frac{1}{|Y|}\iiint\limits_{Y} f(\boldsymbol{y})\mathrm{d}v$$

This homogenization procedure applies to the homogenization of a general relative permittivity $\varepsilon(\boldsymbol{y})$ in the unit cell. Notice that, in general, the homogenized material is anisotropic. Below, in Section 4, we specialize to a permittivity that takes two constant values in the unit cell Y.

3. CLASSICAL MIXTURE FORMULA — SPHERICAL INCLUSIONS

Before we proceed with the solution of the local problem for a two-phased material, it is instructive to review some of the results on classical mixture formulae, which are obtained by the use of physical arguments and approximations. Some of these classical mixture formulae are derived for a random distribution of spheres, but they are nevertheless often applied to a regular lattice problem. The formulae apply only to the case of inclusions of simple shapes, e.g., spheres or, more generally, ellipsoids, see [16].

The classical mixture formulae apply to spherical inclusions in a background material. Therefore, let the material consist of two phases; a background material with relative permittivity ε_b and periodically arranged spherical inclusions (cubic lattice) with relative permittivity ε_i. We denote the radius of the sphere by a, and without loss of

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[‡] More precisely, $H^1_{\#}(Y)$ is the closure of $C^\infty_{\#}(Y)$ in the H^1-norm, where $C^\infty_{\#}(Y)$ is the subset of $C^\infty(\mathbb{R}^3)$ of Y-periodic functions [4].

generality we let the periodicity be 1. The volume fraction of the inclusions is then $f = 4\pi a^3/3$, $a < 1/2$.

Several of the effective relative permittivity expressions of a mixture of homogeneous spherical inclusions are represented in the formula [16]

$$\frac{\varepsilon^h - \varepsilon_b}{\varepsilon^h + 2\varepsilon_b + \nu(\varepsilon^h - \varepsilon_b)} = f\frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b + \nu(\varepsilon^h - \varepsilon_b)}$$

where ε^h is the effective permittivity of the mixture. The integer ν represents different mixture formulas, e.g., $\nu = 0$ the Maxwell Garnett formula, $\nu = 2$ the Böttcher mixture rule or Bruggeman formula, and $\nu = 3$ the coherent potential (CP) formula. The Maxwell Garnett formula is explicitly

$$\varepsilon^h = \varepsilon_b + 3f\varepsilon_b\frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)} \tag{3}$$

Another mixture formula was derived by Lord Rayleigh and is given by [5, 8–11, 13, 14, 16]

$$\varepsilon^h = \varepsilon_b + \frac{3f\varepsilon_b}{(\varepsilon_i + 2\varepsilon_b)/(\varepsilon_i - \varepsilon_b) - f - 1.305f^{10/3}(\varepsilon_i - \varepsilon_b)/(\varepsilon_i + 4\varepsilon_b/3)}$$

This formulae is identical to the Maxwell Garnett formula for small values of the radius a. In fact, we have

$$\begin{aligned}
\varepsilon^h &= \varepsilon_b + 3f\varepsilon_b\frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b) - 1.305f^{10/3}(\varepsilon_i - \varepsilon_b)^2/(\varepsilon_i + 4\varepsilon_b/3)} \\
&= \varepsilon_b + 3f\varepsilon_b\frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)} \\
&\quad \cdot \left\{ 1 + \frac{1.305f^{10/3}(\varepsilon_i - \varepsilon_b)^2/(\varepsilon_i + 4\varepsilon_b/3)}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)} \right\} + \cdots \\
&\approx \varepsilon_b + 3f\varepsilon_b\frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)} \\
&\quad + 9f\varepsilon_b\frac{155a^{10}(\varepsilon_i - \varepsilon_b)^3}{(3\varepsilon_i + 4\varepsilon_b)[\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)]^2} + \cdots
\end{aligned}$$

All classical mixture formulae have their domain of validity for small volume fractions f. The differences between the different formulae are best seen from the power series expansion in f, i.e.,

$$\varepsilon^h = \varepsilon_b + \alpha f + \beta f^2 + O(f^3)$$

Table 1. The different coefficients in an expansion of $\varepsilon^h = \varepsilon_b + \alpha f + \beta f^2$ for small volume fractions f and different mixture formulae. The constant $\nu = 0$ for the Maxwell Garnett formula, $\nu = 2$ for the Böttcher mixture rule or Bruggeman formula, and $\nu = 3$ for the coherent potential (CP) formula. The coefficient $\alpha = 3\varepsilon_b(\varepsilon_i - \varepsilon_b)/(\varepsilon_i + 2\varepsilon_b)$.

ν	β
0	$\alpha^2/(3\varepsilon_b)$
2	$\alpha^2\varepsilon_i/(\varepsilon_b(\varepsilon_i + 2\varepsilon_b))$
3	$\alpha^2(4\varepsilon_i - \varepsilon_b)/(3\varepsilon_b(\varepsilon_i + 2\varepsilon_b))$

The coefficient $\alpha = 3\varepsilon_b(\varepsilon_i - \varepsilon_b)/(\varepsilon_i + 2\varepsilon_b)$ is the same in all these formulae, and this contribution represents the dipole contribution. The β coefficient for the different formulae is given in Table 1 [16, p. 164]. Note that all mixture formulae agree up to first order in f; then the formulae corresponding to different ν-values differ (the Maxwell Garnett and the Rayleigh formulae agree). Below, we show that the homogenization procedure verifies the correctness of the first term α and also that the Maxwell Garnett formula is correct in a power series expansion in a up to a^{12}. In fact, no other mixture formula (Rayleigh formula is similar to the Maxwell Garnett formula) gives the correct result beyond the dipole term (power a^3). Moreover, the Rayleigh formula is correct up to order 13 when compared to the result obtained by the solution of the local problem treated in this paper. Lord Rayleigh's result is verified with the technique used in this paper in Section 6.

4. SPHERICAL INCLUSIONS — SOLUTION OF LOCAL PROBLEM

The local problem, (1), for a permittivity that takes two constant values in the unit cell Y is now addressed. To this end, let V be an open subset of the Y-cell with a Lipschitz continuous[§] boundary S. The relative permittivity assumes the value ε_i inside V and ε_b elsewhere in Y, i.e., the permittivity is of the form
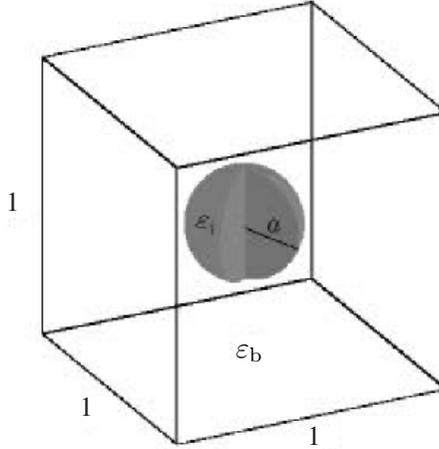
$$\varepsilon(\boldsymbol{y}) = \varepsilon_b + (\varepsilon_i - \varepsilon_b)\chi_V \qquad (4)$$

[§] This assumption makes the traces on the surface S below well-defined.

where χ_V is the characteristic function of the inclusion V. Below, we apply only to the case of a single spherical inclusion, i.e.,

$$\chi(\boldsymbol{y}) = H(a - y), \qquad \boldsymbol{y} \in Y \tag{5}$$

where $y = |\boldsymbol{y}|$ and $a < 1/2$ is the radius of the sphere, and H is the Heaviside step function. This geometry is depicted in Figure 2.

Figure 2. The geometry of the problem with a spherical inclusion of radius a in the unit cell Y.

It is appropriate to reformulate the local problem as a partial differential equation problem. We start by finding the equivalent differential problem. The volume integral is split in two parts — one containing the integral over V, and one over $Y \backslash V$.

$$\varepsilon_b \iiint\limits_{Y \backslash V} \nabla w(\boldsymbol{y}) \cdot (\hat{\boldsymbol{e}}_j - \nabla \chi_j(\boldsymbol{y})) \mathrm{d}v + \varepsilon_i \iiint\limits_{V} \nabla w(\boldsymbol{y}) \cdot (\hat{\boldsymbol{e}}_j - \nabla \chi_j(\boldsymbol{y})) \mathrm{d}v = 0$$

Apply the Green's theorem and use the periodic boundary conditions of the test function w and the solution χ_j. We get, assuming two times differential solutions χ_j in V and $Y \backslash V$, respectively

$$\iint\limits_{S} w(\boldsymbol{y}) \left\{ (\varepsilon_i - \varepsilon_b)\hat{\boldsymbol{e}}_j \cdot \hat{\boldsymbol{\nu}}(\boldsymbol{y}) + \varepsilon_b \frac{\partial}{\partial y} \chi_j(\boldsymbol{y}^+) - \varepsilon_i \frac{\partial}{\partial y} \chi_j(\boldsymbol{y}^-) \right\} \mathrm{d}S$$

$$+ \varepsilon_b \iiint\limits_{Y \backslash V} w(\boldsymbol{y}) \nabla^2 \chi_j(\boldsymbol{y})) \mathrm{d}v + \varepsilon_i \iiint\limits_{V} w(\boldsymbol{y}) \nabla^2 \chi_j(\boldsymbol{y})) \mathrm{d}v = 0$$

where dS is the surface measure of the boundary S, and $\hat{\boldsymbol{\nu}}(\boldsymbol{y})$ is outward directed unit normal vector. We also denote the traces of the position vector from the outside and the inside of S by $\boldsymbol{y}^{\pm}$, respectively. Since the test function is arbitrary, the local problem is rewritten as an equivalent boundary value problem for $j = 1, 2, 3$, i.e.,

$$
\begin{cases}
\nabla^2 \chi_j(\boldsymbol{y}) = 0, & \boldsymbol{y} \in Y, \ \boldsymbol{y} \notin S \\
\dfrac{\partial}{\partial y} \chi_j(\boldsymbol{y}^+) - \varepsilon \dfrac{\partial}{\partial y} \chi_j(\boldsymbol{y}^-) = \hat{\boldsymbol{e}}_j \cdot \hat{\boldsymbol{\nu}}(\boldsymbol{y})(1 - \varepsilon), & \boldsymbol{y} \in S \\
\chi_j(\boldsymbol{y}^+) = \chi_j(\boldsymbol{y}^-), & \boldsymbol{y} \in S \\
\chi_j(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) = \chi_j(\boldsymbol{y}), & \boldsymbol{y} \in \partial Y, \ i = 1, 2, 3
\end{cases}
\tag{6}
$$

where

$$
\varepsilon = \varepsilon_i / \varepsilon_b
$$

The solution to equation (6) is uniquely defined up to a constant [4].

In the next section, we specialize to the spherical inclusion case, (5), and we make an Ansatz in spherical solutions of the Laplace equation to solve this problem. From the solution of this problem, we calculate the effective relative permittivity for a spherical inclusion, see Section 5, and in Section 6 the effective relative permittivity for small values of the radius a is extracted.

4.1. Ansatz

The solution of (6) for the spherical inclusion, (5), is now sought. The unit normal vector is in this case $\hat{\boldsymbol{\nu}}(\boldsymbol{y}) = \hat{\boldsymbol{y}} = \boldsymbol{y}/|\boldsymbol{y}|$. We make an Ansatz in solutions to the Laplace equation in spherical coordinates to determine the solution. The solutions to the Laplace equation in spherical coordinates are denoted u_n (singular at the origin) and v_n (regular at the origin), see Appendix A for more details about these definitions, and we adopt the notion of multi-index $n = \{\sigma, m, l\}$, where $\sigma = $ e (even), o (odd), $m = 0, 1, 2, \ldots, l - 1, l$ and $l = 0, 1, 2, 3, \ldots$.

Due to the symmetries of the problem it suffices to solve for one value of j in (6), i.e., $j = 3$, and below we denote this solution $\psi = \chi_j$. Moreover, the symmetries of the problem (even in $y_1 \to -y_1$ and $y_2 \to -y_2$, and odd in $y_3 \to -y_3$) imply that the only functions u_n and v_n that contribute in these expansions must have l odd integer, $\sigma = $ e, and m even integer. The pertinent index set is therefore $I = \{(\sigma, m, l) = (\mathrm{e}, m, l): m \text{ even}, l \text{ odd}\}$.

Outside the sphere we make an Ansatz as linear combination in the singular functions u_n periodically extended by the cubic lattice, and inside the sphere the solution must be regular and we expand in

the regular functions v_n, i.e.,

$$
\begin{cases}
\psi(\boldsymbol{y}) = \sum_{n \in I} a_n \sum_{i \in \mathbb{Z}^3} (u_n(\boldsymbol{y} + \boldsymbol{t}_i) - (\boldsymbol{y} \cdot \boldsymbol{\Phi}_i + \phi_i)\delta_{n,\mathrm{e}01}) + C v_{\mathrm{e}01}(\boldsymbol{y}), \\
\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \boldsymbol{y} \in Y \backslash V \\
\psi(\boldsymbol{y}) = \sum_{n \in I} b_n v_n(\boldsymbol{y}), \qquad\qquad\qquad\qquad \boldsymbol{y} \in V
\end{cases}
\tag{7}
$$

where the translation vector, $\boldsymbol{t}_i = i_1 \hat{\boldsymbol{e}}_1 + i_2 \hat{\boldsymbol{e}}_2 + i_3 \hat{\boldsymbol{e}}_3$, $i = (i_1, i_2, i_3) \in \mathbb{Z}^3$, and where

$$
\begin{cases}
\phi_i = (1 - \delta_{i,(000)})u_{\mathrm{e}01}(\boldsymbol{t}_i) = (1 - \delta_{i,(000)})\dfrac{a^2}{y^2}Y_{\mathrm{e}01}(\hat{\boldsymbol{y}})\Big|_{\boldsymbol{y}=\boldsymbol{t}_i} \\[2mm]
\boldsymbol{\Phi}_i = (1 - \delta_{i,(000)})\nabla u_{\mathrm{e}01}(\boldsymbol{y})\Big|_{\boldsymbol{y}=\boldsymbol{t}_i} = (1 - \delta_{i,(000)})\nabla \left(\dfrac{a^2}{y^2}Y_{\mathrm{e}01}(\hat{\boldsymbol{y}})\right)\Big|_{\boldsymbol{y}=\boldsymbol{t}_i}
\end{cases}
$$

We observe that the translated terms in (7) for $n \in I$ behave for large translations as

$$
u_n(\boldsymbol{y} + \boldsymbol{t}_i) - (\boldsymbol{y} \cdot \boldsymbol{\Phi}_i + \phi_i)\delta_{n,\mathrm{e}01} = O(|i|^{-4}) \text{ as } |i| \to \infty
$$

where $|i| = \sqrt{i_1^2 + i_2^2 + i_3^2}$. The series in (7) is therefore absolutely convergent for every $\boldsymbol{y} \in Y$ w.r.t. the summation i.

The terms in the Ansatz are by construction Y-periodic for all $I \ni n \neq \{\mathrm{e}01\}$. Notice that the term corresponding to $n = \{\mathrm{e}01\}$ has to be compensated with a term $C v_{\mathrm{e}01} \sim y_3$ so that the entire solution becomes periodic by an appropriate adjustment of the constant C. To see this, define

$$
w(\boldsymbol{y}) = \sum_{i \in \mathbb{Z}^3} [u_{\mathrm{e}01}(\boldsymbol{y} + \boldsymbol{t}_i) - (\boldsymbol{y} \cdot \boldsymbol{\Phi}_i + \phi_i)]
$$

This sum is absolutely convergent and any rearrangement of the terms gives the same value of the sum [17]. We get

$$
\begin{aligned}
w(\boldsymbol{y} + \boldsymbol{t}_k) - w(\boldsymbol{y}) &= \sum_{i \in \mathbb{Z}^3} [u_{\mathrm{e}01}(\boldsymbol{y} + \boldsymbol{t}_k + \boldsymbol{t}_i) - ((\boldsymbol{y} + \boldsymbol{t}_k) \cdot \boldsymbol{\Phi}_i + \phi_i)] \\
&\quad - \sum_{i \in \mathbb{Z}^3} [u_{\mathrm{e}01}(\boldsymbol{y} + \boldsymbol{t}_i) - (\boldsymbol{y} \cdot \boldsymbol{\Phi}_i + \phi_i)] \\
&= \boldsymbol{y} \cdot \sum_{i \in \mathbb{Z}^3} (\boldsymbol{\Phi}_i - \boldsymbol{\Phi}_{i-k}) + \sum_{i \in \mathbb{Z}^3} (\phi_i - \phi_{i-k} - \boldsymbol{t}_k \cdot \boldsymbol{\Phi}_{i-k})
\end{aligned}
$$

where $i - k = (i_1 - k_1, i_2 - k_2, i_3 - k_3)$. The last two sums are absolutely convergent. Instead of computing these two sums, we compensate by adding the last linear term, Cv_{e01}, in the Ansatz, (7), and adjust the constant C by an integral computation that is easier to perform. The constant term is immaterial in the computations below, since the solution of the local problem is undetermined by a constant.

Notice that the sums (see Appendix A for an explicit expression of u_{e01}, and also Appendix D for similar summations)

$$
\lim_{R \to \infty} \sum_{\substack{i \in \mathbb{Z}^3 \\ |\boldsymbol{t}_i| \leq R}} \phi_i = a^2 \sqrt{\frac{3}{4\pi}} \lim_{R \to \infty} \sum_{\substack{\mathbb{Z}^3 \ni i \neq (000) \\ |\boldsymbol{t}_i| \leq R}} \frac{i_3}{\left(i_1^2 + i_2^2 + i_3^2\right)^{3/2}} = 0
$$

and

$$
\lim_{R \to \infty} \sum_{\substack{i \in \mathbb{Z}^3 \\ |\boldsymbol{t}_i| \leq R}} \boldsymbol{\Phi}_i
$$
$$
= a^2 \sqrt{\frac{3}{4\pi}} \lim_{R \to \infty} \sum_{\substack{\mathbb{Z}^3 \ni i \neq (000) \\ |\boldsymbol{t}_i| \leq R}} \frac{\hat{\boldsymbol{e}}_3 \left(i_1^2 + i_2^2 + i_3^2\right) - 3i_3 \left(i_1 \hat{\boldsymbol{e}}_1 + i_2 \hat{\boldsymbol{e}}_2 + i_3 \hat{\boldsymbol{e}}_3\right)}{\left(i_1^2 + i_2^2 + i_3^2\right)^{5/2}}
$$
$$
= \boldsymbol{0}
$$

by symmetry, and therefore, all sums in (7) can be interpreted as

$$
\begin{cases}
\psi(\boldsymbol{y}) = \sum_{n \in I} a_n \lim_{R \to \infty} \sum_{\substack{i \in \mathbb{Z}^3 \\ |\boldsymbol{t}_i| \leq R}} u_n(\boldsymbol{y} + \boldsymbol{t}_i) + Cv_{e01}(\boldsymbol{y}), & \boldsymbol{y} \in Y \backslash V \\
\psi(\boldsymbol{y}) = \sum_{n \in I} b_n v_n(\boldsymbol{y}), & \boldsymbol{y} \in V
\end{cases} \tag{8}
$$

4.2. The Solution

We now determine the expansion coefficients a_n and b_n as well as the constant C in (8) so that the boundary conditions on the sphere S are satisfied and the periodicity of the solution is guaranteed.

The translation properties of u_n, see (A1) and Appendix C, is very useful here.

$$
u_n(\boldsymbol{y} + \boldsymbol{d}) = \sum_{n'} P_{nn'}(\boldsymbol{d}) v_{n'}(\boldsymbol{y}), \qquad y < d
$$

This sum is absolutely convergent for all arguments $\boldsymbol{y}$ and $\boldsymbol{d}$ such that $y < d$. The definition of the $S_{nn'}$, see Appendix D and (D1), readily

implies that the solution exterior to the sphere can be written as[||]

$$\psi(\boldsymbol{y}) = \sum_{n \in I, n'} a_n \{\delta_{nn'} u_{n'}(\boldsymbol{y}) + S_{nn'} v_{n'}(\boldsymbol{y})\} + C v_{\mathrm{e}01}(\boldsymbol{y}), \quad \boldsymbol{y} \in Y \backslash V$$

where

$$S_{nn'} = \lim_{R \to \infty} \sum_{\substack{\mathbb{Z}^3 \ni i \neq (000) \\ |\boldsymbol{t}_i| \leq R}} P_{nn'}(\boldsymbol{t}_i)$$

To satisfy the continuity conditions on the surface of the sphere, we must, by orthogonality of the spherical harmonics, have

$$C \delta_{n,\mathrm{e}01} + \sum_{n' \in I} a_{n'} \{\delta_{n'n} + S_{n'n}\} = b_n, \qquad n \in I \tag{9}$$

Since $u_n(a\hat{\boldsymbol{y}}) = v_n(a\hat{\boldsymbol{y}}) = Y_n(a\hat{\boldsymbol{y}})$. Moreover, the discontinuity of the normal derivative on the surface of the sphere implies, due to orthogonality of the spherical harmonics on the unit sphere, that $(n \in I)$

$$\sum_{n' \in I} a_{n'} \{-(l+1)\delta_{n'n} + lS_{n'n}\} + C\delta_{n,\mathrm{e}01} - \varepsilon l b_n + (\varepsilon - 1)a\sqrt{\frac{4\pi}{3}} \delta_{n,\mathrm{e}01} = 0$$

$$\tag{10}$$

where we used

$$\frac{\partial}{\partial y} u_n(a\hat{\boldsymbol{y}}) = -\frac{l+1}{a} Y_n(\hat{\boldsymbol{y}}), \qquad \frac{\partial}{\partial y} v_n(a\hat{\boldsymbol{y}}) = \frac{l}{a} Y_n(\hat{\boldsymbol{y}})$$

and $\hat{\boldsymbol{e}}_3 \cdot \hat{\boldsymbol{y}} = \cos\theta = \sqrt{4\pi/3} Y_{\mathrm{e}01}(\hat{\boldsymbol{y}})$, see definition of the spherical harmonics $Y_n(\hat{\boldsymbol{y}})$ in Appendix A.

From the continuity of ψ on S and the periodicity of ψ on ∂Y, we get

$$\iiint_Y \nabla\psi(\boldsymbol{y}) \mathrm{d}v = \iint_{\partial Y} \hat{\boldsymbol{\nu}}(\boldsymbol{y})\psi(\boldsymbol{y}) \mathrm{d}S = \boldsymbol{0}$$

which implies

$$\lim_{R \to \infty} \iint_{\partial Y} \hat{\boldsymbol{\nu}}(\boldsymbol{y}) \left\{ \sum_{n \in I} a_n \sum_{\substack{i \in \mathbb{Z}^3 \\ |\boldsymbol{t}_i| \leq R}} u_n(\boldsymbol{y} + \boldsymbol{t}_i) + C v_{\mathrm{e}01}(\boldsymbol{y}) \right\} \mathrm{d}S = \boldsymbol{0}$$

[||] The index n' here runs over all σ, m, l-values, but is below, due to symmetry, shown to run only over the index set I.

The integral can be computed exactly, see Appendix B, (B1) and (B2), and this relation also restores the periodicity of the solution. The result is

$$a^2\sqrt{\frac{4\pi}{3}}\hat{e}_3 a_{\text{e}01} + C\sqrt{\frac{3}{4\pi}}\hat{e}_3\frac{1}{a} = \mathbf{0}$$

or ($f = 4\pi a^3/3$)

$$C = -fa_{\text{e}01} \tag{11}$$

We now eliminated the coefficients b_n and C in (10) by the use of (9) and (11). The result is ($n \in I$)

$$\sum_{n'\in I} a_{n'}\{-(l+1+l\varepsilon)\delta_{n'n} + l(1-\varepsilon)S_{n'n}\} + (\varepsilon-1)fa_{\text{e}01}\delta_{n,\text{e}01}$$

$$= -(\varepsilon-1)a\sqrt{\frac{4\pi}{3}}\delta_{n,\text{e}01}$$

or

$$\sum_{n'\in I} a_{n'}M_{n'n} = (\varepsilon-1)a\sqrt{\frac{4\pi}{3}}\delta_{n,\text{e}01}, \qquad n \in I \tag{12}$$

where

$$M_{n'n} = [l(\varepsilon+1)+1-(\varepsilon-1)f\delta_{n,\text{e}01}]\delta_{n'n} + l(\varepsilon-1)S_{n'n}, \quad n,n' \in I \tag{13}$$

The solution to (12) is then

$$a_n = (\varepsilon-1)a\sqrt{\frac{4\pi}{3}}(M^{-1})_{\text{e}01n}, \qquad n \in I$$

and the final solution of (7) is

$$\begin{cases} \psi(\boldsymbol{y}) = (\varepsilon-1)a\sqrt{\dfrac{4\pi}{3}}\displaystyle\sum_{n\in I}(M^{-1})_{\text{e}01n}\{u_n(\boldsymbol{y}) + w_n(\boldsymbol{y})\}, & \boldsymbol{y} \in Y\backslash V \\[2ex] \psi(\boldsymbol{y}) = (\varepsilon-1)a\sqrt{\dfrac{4\pi}{3}}\displaystyle\sum_{n\in I}(M^{-1})_{\text{e}01n}\{v_n(\boldsymbol{y}) + w_n(\boldsymbol{y})\}, & \boldsymbol{y} \in V \end{cases}$$

$$\tag{14}$$

where

$$w_n(\boldsymbol{y}) = \sum_{n'\in I} S_{nn'}v_{n'}(\boldsymbol{y}) - f\delta_{n,\text{e}01}v_n(\boldsymbol{y}), \qquad n \in I$$

Specifically, the solution (14) evaluated on the sphere is

$$\psi(a\hat{\boldsymbol{y}}) = (\varepsilon-1)a\sqrt{\frac{4\pi}{3}}\sum_{nn'\in I}(M^{-1})_{\text{e}01n}\{\delta_{nn'}(1-f\delta_{n,\text{e}01}) + S_{nn'}\}Y_{n'}(\hat{\boldsymbol{y}})$$

$$\tag{15}$$

This is the explicit solution of the problem on the sphere provided the sum converges.

5. THE HOMOGENIZED RELATIVE PERMITTIVITY

In this section, we find a closed form expression of the effective relative permittivity of the heterogeneous material with spherical inclusions.

From the solution ψ in (14), we compute the effective relative permittivity of the problem. Due to the symmetry of the problem, the homogenized relative permittivity is isotropic, and its value is, see (2) ($\varepsilon_{ij}^h = \varepsilon^h \delta_{ij}$)

$$\varepsilon^h = <\varepsilon(\boldsymbol{y})> - <\varepsilon(\boldsymbol{y})\frac{\partial}{\partial y_3}\psi(\boldsymbol{y})>$$

where the last term is transformed to a surface integral over the sphere S, viz.,

$$<\varepsilon(\boldsymbol{y})\frac{\partial}{\partial y_3}\psi(\boldsymbol{y})> = (\varepsilon_i - \varepsilon_b)\hat{\boldsymbol{e}}_3 \cdot \iint\limits_S \psi(\boldsymbol{y})\hat{\boldsymbol{y}}\mathrm{d}S$$

since the contribution from the boundary ∂Y vanish.

The homogenized relative permittivity then is ($f = 4\pi a^3/3$)

$$\frac{\varepsilon^h}{\varepsilon_b} = 1 - f + \varepsilon f - (\varepsilon - 1)\sqrt{\frac{4\pi}{3}}a^2 \iint\limits_\Omega \psi(a\hat{\boldsymbol{y}})Y_{\mathrm{e}01}(\hat{\boldsymbol{y}})\mathrm{d}\Omega \qquad (16)$$

Here the surface measure of the unit sphere Ω in $\mathbb{R}^3$ is denoted $\mathrm{d}\Omega$. We notice that only the projection of the solution ψ on the spherical harmonics $Y_{\mathrm{e}01}$ is important in the computation of the homogenized relative permittivity.

Due to the orthogonality of the spherical harmonics on the unit sphere, the relative permittivity ε^h can be written in a series as, see (15)

$$\frac{\varepsilon^h}{\varepsilon_b} = 1 + f + (\varepsilon-1)f - (\varepsilon-1)^2 f \sum_{n\in I}(M^{-1})_{\mathrm{e}01n}\{(1-f)\delta_{n,\mathrm{e}01} + S_{n\mathrm{e}01}\} \quad (17)$$

This solution of the homogenized relative permittivity is one of the main results of this paper. This expression gives the relative permittivity of the spherical inclusions correct to all orders of a, provided the series converges. Below, we extract the first non-vanishing contributions in powers of the radius a.

The summation in (17) is restricted to the index set I. All other terms do not contribute to the sum since the symmetry of the problem (even in $y_1 \to -y_1$ and $y_2 \to -y_2$, and odd in $y_3 \to -y_3$) implies that $S_{n\mathrm{e}01} = 0$, if l even integer, $\sigma = \mathrm{o}$, or m odd integer.

6. SMALL VOLUME FRACTION ANALYSIS

In this section, we extract the leading contribution in a power series expansion in the radius a of ε^h.

The dependence of a in the quantities $S_{nn'}$ is, see Appendix D

$$S_{nn'} = O(a^{l+l'+1})$$

Also, from the fact $S_{\mathrm{e01e01}} = S_{\mathrm{e01e23}} = S_{\mathrm{e23e01}} = 0$, we get the lowest order contribution only from terms S_{e03e01}, and S_{e01e03} which each contribute as $O(a^5)$. The absence of the terms S_{e01e01}, S_{e01e23}, and S_{e23e01} is an effect of the symmetry of the lattice (cubic lattice) and for an other type of lattice these terms can contribute.

The symmetries of the problem simplify the evaluation of the inverse $(M^{-1})_{\mathrm{e01}n}$. To leading order in a, we have, see (13) $(nn' \in I)$

$$
\begin{aligned}
M_{nn'} &= [l(\varepsilon+1) + 1 - f(\varepsilon-1)\delta_{n,\mathrm{e01}}]\delta_{nn'} + (\varepsilon-1)S_{\mathrm{e03e01}}\delta_{n,\mathrm{e03}}\delta_{n'\mathrm{e01}} \\
&\quad + 3(\varepsilon-1)S_{\mathrm{e01e03}}\delta_{n,\mathrm{e01}}\delta_{n',\mathrm{e03}} + O(a^7)
\end{aligned}
\tag{18}
$$

where the elements in $O(a^7)$ contain only matrix entries with at least one index $l \geq 5$. The inverse has to the same leading order in a the form[¶] $(n \in I)$

$$(M^{-1})_{\mathrm{e01}n} = \frac{\delta_{n,\mathrm{e01}}}{\varepsilon+2-f(\varepsilon-1)} - \frac{3(\varepsilon-1)S_{\mathrm{e01e03}}\delta_{n,\mathrm{e03}}}{(3\varepsilon+4)[\varepsilon+2-f(\varepsilon-1)]} + O(a^7) \tag{19}$$

For the diagonal term $(M^{-1})_{\mathrm{e01e01}}$ we also need higher order terms. The identity

$$
\begin{aligned}
1 &= \sum_{n \in I}(M^{-1})_{\mathrm{e01}n}M_{n\mathrm{e01}} \\
&= (M^{-1})_{\mathrm{e01e01}}M_{\mathrm{e01e01}} + (M^{-1})_{\mathrm{e01e03}}M_{\mathrm{e03e01}} + O(a^{14})
\end{aligned}
$$

implies by the use of (18) and (19)

$$1 = (M^{-1})_{\mathrm{e01e01}}(\varepsilon+2-f(\varepsilon-1)) - \frac{3(\varepsilon-1)^2 S_{\mathrm{e01e03}}S_{\mathrm{e03e01}}}{(3\varepsilon+4)[\varepsilon+2-f(\varepsilon-1)]} + O(a^{12})$$

[¶] Assume $\mathbf{M}$ has the form

$$\mathbf{M} = \mathbf{D} + a^5\mathbf{A} + O(a^7)$$

where $\mathbf{D}$ is a diagonal matrix, $\mathbf{D}$ and $\mathbf{A}$ are independent of a. The inverse is then

$$\mathbf{M}^{-1} = \mathbf{D}^{-1} - a^5\mathbf{D}^{-1} \cdot \mathbf{A} \cdot \mathbf{D}^{-1} + O(a^7)$$

The diagonal term $(M^{-1})_{\mathrm{e01e01}}$ is therefore

$$(M^{-1})_{\mathrm{e01e01}} = \frac{1}{\varepsilon + 2 - f(\varepsilon - 1)} + \frac{3(\varepsilon - 1)^2 S_{\mathrm{e01e03}} S_{\mathrm{e03e01}}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2} + O(a^{12})$$

We are now ready to insert all these results and limits into (17). The result is (remember $S_{\mathrm{e01e01}} = 0$)

$$
\begin{aligned}
\frac{\varepsilon^h}{\varepsilon_b} &= 1 + (\varepsilon - 1)f - \frac{(\varepsilon - 1)^2 f(1 - f)}{\varepsilon + 2 - f(\varepsilon - 1)} \\
&\quad - \frac{3f(1 - f)(\varepsilon - 1)^4 S_{\mathrm{e01e03}} S_{\mathrm{e03e01}}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2} \\
&\quad + \frac{3f(\varepsilon - 1)^3 S_{\mathrm{e01e03}} S_{\mathrm{e03e01}}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]} + O(a^{15})
\end{aligned}
$$

which simplifies to

$$\frac{\varepsilon^h}{\varepsilon_b} = 1 + \frac{3f(\varepsilon - 1)}{\varepsilon + 2 - f(\varepsilon - 1)} + \frac{9f(\varepsilon - 1)^3 S_{\mathrm{e01e03}} S_{\mathrm{e03e01}}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2} + O(a^{15}) \quad (20)$$

We observe that the first term is the Maxwell Garnett term, see (3). This expansion is in agreement with the result reported in e.g., [7]. The lowest order correction term is

$$\varepsilon_{\mathrm{corr}} = \frac{9f(\varepsilon - 1)^3 S_{\mathrm{e01e03}} S_{\mathrm{e03e01}}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2}$$

and since, see (D2)

$$S_{\mathrm{e01e03}} S_{\mathrm{e03e01}} = 196 a^{10} (4S_1 + 2S_2 - S_3)^2 \approx 155 a^{10}$$

where the sums are

$$S_1 = \sum_{ijk=1}^{\infty} \frac{3i^2 j^2 - i^4}{(i^2 + j^2 + k^2)^{9/2}}, \quad S_2 = \sum_{ij=1}^{\infty} \frac{3i^2 j^2 - 2i^4}{(i^2 + j^2)^{9/2}}, \quad S_3 = \sum_{i=1}^{\infty} \frac{1}{i^5}$$

we have the final result for the first correction term to the Maxwell Garnett formula

$$
\begin{aligned}
\varepsilon_{\mathrm{corr}} &= \frac{1764 f(\varepsilon - 1)^3 a^{10} (4S_1 + 2S_2 - S_3)^2}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2} \\
&\approx 9f \frac{155(\varepsilon - 1)^3 a^{10}}{(3\varepsilon + 4)[\varepsilon + 2 - f(\varepsilon - 1)]^2}
\end{aligned}
$$

This result is in agreement with the result obtained by Lord Rayleigh [13], who used a different technique to obtain the result. Moreover, it is consistent with the Hashin-Shtrikman's bounds in (21) since $\varepsilon_{\mathrm{corr}}$ is positive for $\varepsilon > 1$.

6.1. Hashin-Shtrikman's Bounds

The Hashin-Shtrikman's bounds constitute the limit values of the effective permittivity ε^h. For the case $\varepsilon > 1$, the homogenized value ε^h for spherical inclusions is bounded by [4, 16]

$$1 + \frac{3f(\varepsilon - 1)}{\varepsilon + 2 - f(\varepsilon - 1)} \leq \frac{\varepsilon^h}{\varepsilon_b} \leq \varepsilon + \frac{3(1 - f)\varepsilon(1 - \varepsilon)}{1 + 2\varepsilon - (1 - f)(1 - \varepsilon)} \qquad (21)$$

If $\varepsilon < 1$ the inequalities are reversed.

7. NUMERICAL TREATMENT

The original problem given in (6)

$$\begin{cases} \nabla^2 \psi(\boldsymbol{y}) = 0, & \boldsymbol{y} \in Y, \ \boldsymbol{y} \notin S \\ \dfrac{\partial}{\partial y}\psi(\boldsymbol{y}^+) - \varepsilon \dfrac{\partial}{\partial y}\psi(\boldsymbol{y}^-) = \hat{\boldsymbol{e}}_3 \cdot \hat{\boldsymbol{y}}(1 - \varepsilon), & \boldsymbol{y} \in S \\ \psi(\boldsymbol{y}^+) = \psi(\boldsymbol{y}^-), & \boldsymbol{y} \in S \\ \psi(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) = \psi(\boldsymbol{y}), & \boldsymbol{y} \in \partial Y, \ i = 1, 2, 3 \end{cases}$$

for the permittivity profile in (5) is easily transformed to a homogeneous boundary value problem.

Let ψ_0 denote the function

$$\psi_0(\boldsymbol{y}) = \frac{\varepsilon - 1}{\varepsilon + 2}a\sqrt{\frac{4\pi}{3}} \left\{ \begin{array}{c} v_{\mathrm{e}01}(\boldsymbol{y}) \\ u_{\mathrm{e}01}(\boldsymbol{y}) \end{array} \right\} = \frac{\varepsilon - 1}{\varepsilon + 2}a\cos\theta \left\{ \begin{array}{c} y/a \\ (a/y)^2 \end{array} \right\} \left\{ \begin{array}{c} y < a \\ y > a \end{array} \right. \tag{22}$$

The function $\Psi = \psi - \psi_0$ then satisfies

$$\begin{cases} \nabla^2 \Psi(\boldsymbol{y}) = 0, & \boldsymbol{y} \in Y, \ \boldsymbol{y} \notin S \\ \dfrac{\partial}{\partial y}\Psi(\boldsymbol{y}^+) - \varepsilon \dfrac{\partial}{\partial y}\Psi(\boldsymbol{y}^-) = 0, & \boldsymbol{y} \in S \\ \Psi(\boldsymbol{y}^+) = \Psi(\boldsymbol{y}^-), & \boldsymbol{y} \in S \\ \Psi(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) = \Psi(\boldsymbol{y}) - \psi_0(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) + \psi_0(\boldsymbol{y}), & \boldsymbol{y} \in \partial Y, \ i = 1, 2, 3 \end{cases}$$

or formally

$$\begin{cases} \nabla \cdot [\varepsilon(\boldsymbol{y})\nabla\Psi(\boldsymbol{y})] = 0, & \boldsymbol{y} \in Y \\ \Psi(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) = \Psi(\boldsymbol{y}) - \psi_0(\boldsymbol{y} + \hat{\boldsymbol{e}}_i) + \psi_0(\boldsymbol{y}), & \boldsymbol{y} \in \partial Y, \ i = 1, 2, 3 \end{cases}$$

which is a problem without source term. However, the problem is now no longer periodic at the boundary, since ψ_0 is on odd function in y_3.

Due to the symmetry and the periodicity of the original problem (even in $y_1 \to -y_1$ and $y_2 \to -y_2$, and odd in $y_3 \to -y_3$), we have

$$\frac{\partial \psi(\pm \hat{e}_1/2)}{\partial y_1} = 0, \qquad \frac{\partial \psi(\pm \hat{e}_2/2)}{\partial y_2} = 0, \qquad \psi(\pm \hat{e}_3/2) = 0$$

Thus, we obtain the following mixed boundary value problem:

$$\begin{cases} \nabla \cdot [\varepsilon(\boldsymbol{y}) \nabla \Psi(\boldsymbol{y})] = 0, & \boldsymbol{y} \in Y \\ \dfrac{\partial \Psi(\pm \hat{e}_1/2)}{\partial y_1} = \dfrac{\partial \Psi(\pm \hat{e}_2/2)}{\partial y_2} = 0 \\ \Psi(\pm \hat{e}_3/2) = \mp \dfrac{\varepsilon - 1}{\varepsilon + 2} \dfrac{1}{2} \left(\dfrac{a}{\sqrt{y_1^2 + y_2^2 + 1/4}} \right)^3 \end{cases} \tag{23}$$

Finally, from (16) and (22), we get

$$\frac{\varepsilon^h}{\varepsilon_b} = 1 + 3f \frac{\varepsilon - 1}{\varepsilon + 2} - (\varepsilon - 1)a^2 \iint\limits_{\Omega} \Psi(a\hat{\boldsymbol{y}}) \cos\theta \mathrm{d}\Omega \tag{24}$$

which we easily evaluate numerically from the numerical solution of (23). The last integral term is the correction term to the dipole contribution. A numerical illustration of these calculations is depicted in Figure 3.
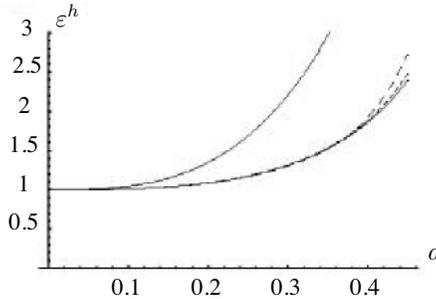
Figure 3. The homogenized relative permittivity ε^h of a spherical inclusion as a function of the radius a. In this figure $\varepsilon_b = 1$ and $\varepsilon_i = 16$. The solid line depicts the Hashin-Shtrikman limits, see (21). The small radius approximation of the homogenized relative permittivity ε^h, see (20), is shown in the dotted line, and the broken line shows the result of the numerical computations, see (24). Notice that the lower Hashin-Shtrikman's limit is identical to the Maxwell Garnett formula (3).

8. CONCLUSIONS AND DISCUSSIONS

An exact expression of the homogenized relative permittivity for homogeneous, isotropic spherical inclusions in a homogeneous, isotropic background material is computed by means of the translation matrices of the solutions to the Lapalace equation in spherical coordinates. This result is compared with the classical mixture formulae in physics. It is found that the first (dipole) term, which all formulae predict, is retrieved. The correct higher order terms are only predicted by the Maxwell Garnett (and the Rayleigh) formula. In fact, the Maxwell Garnett formula is correct up to order a^{12}. The correction term is small, of the order a^{13}, which could explain the success of this classical mixture formula.

The method presented in this paper can be generalized in various ways, and it has potential for materials with anisotropic and more complex lattice configurations than the cubic one analyzed here. Also, higher order correction terms in the power series expansion in the radius a can be extracted.

ACKNOWLEDGMENT

APPENDIX A. THE SPHERICAL COORDINATE SOLUTIONS

The appropriate solutions to the Laplace equation in spherical coordinates are[+]

$$
\begin{cases}
v_n(\boldsymbol{y}) = \left(\dfrac{y}{a}\right)^{l} Y_{\sigma m l}(\hat{\boldsymbol{y}}) \\[4mm]
u_n(\boldsymbol{y}) = \left(\dfrac{y}{a}\right)^{l+1} Y_{\sigma m l}(\hat{\boldsymbol{y}})
\end{cases}
$$

[+] We adopt the notion of multi-index $n = \sigma m l$.

where the spherical harmonics, $Y_{\sigma ml}(\hat{\boldsymbol{y}})$, are orthogonal over the unit sphere Ω. The multi-index, $n = \sigma ml$, takes the values $\sigma = $ e (even), o (odd), $m = 0, 1, 2, \ldots, l-1, l$, and $l = 0, 1, 2, 3, \ldots$. The length of the vector $\boldsymbol{y}$ is denoted by $y = |\boldsymbol{y}|$ and $\hat{\boldsymbol{y}} = \boldsymbol{y}/|\boldsymbol{y}|$. The explicit expression of $Y_{\sigma ml}(\hat{\boldsymbol{y}})$ is [3]

$$Y_{\sigma ml}(\theta, \phi) = \sqrt{\frac{2 - \delta_{m,0}}{2\pi}} \sqrt{\frac{2l+1}{2} \frac{(l-m)!}{(l+m)!}} P_l^m(\cos\theta) \left\{ \begin{matrix} \cos m\phi \\ \sin m\phi \end{matrix} \right\}$$

and $P_l^m(\cos\theta)$ denotes the Associated Legendre functions. The spherical angles of $\hat{\boldsymbol{y}}$ are denoted θ and ϕ, respectively.

Of special importance in this paper is the solutions corresponding to $n = $ e01. They are

$$\left\{ \begin{aligned} v_{\text{e}01}(\boldsymbol{y}) &= \sqrt{\frac{3}{4\pi}} \frac{y_3}{a} \\ u_{\text{e}01}(\boldsymbol{y}) &= \sqrt{\frac{3}{4\pi}} \frac{y_3}{a} \left(\frac{a}{y}\right)^3 \end{aligned} \right.$$

since $\hat{\boldsymbol{e}}_3 \cdot \hat{\boldsymbol{y}} = \cos\theta = \sqrt{4\pi/3}\, Y_{\text{e}01}(\hat{\boldsymbol{y}})$.

Translation of the argument in the singular spherical solutions, u_n, see Figure A1, can be expressed in the regular solutions, v_n, as [3]

$$u_n(\boldsymbol{y} + \boldsymbol{d}) = \sum_{n'} P_{nn'}(\boldsymbol{d}) v_{n'}(\boldsymbol{y}), \qquad y < d \tag{A1}$$

where the matrix $P_{nn'}$ is explicitly given in [3] and in Appendix C. This translation relation is of paramount importance for the analysis presented in this paper.

APPENDIX B. EVALUATION OF SOME INTEGRALS

In this section, we evaluate two integrals of importance in the analysis of this paper. The first integral is

$$I_n = \lim_{R \to \infty} \sum_{\substack{i \in \mathbb{Z}^3 \\ |\boldsymbol{t}_i| \leq R}} \iint_{\partial Y} \hat{\boldsymbol{\nu}}(\boldsymbol{y}) u_n(\boldsymbol{y} + \boldsymbol{t}_i) \mathrm{d}S$$

where $\mathrm{d}S$ is the surface measure of the boundary surface ∂Y. This integral is readily transformed into an integral over a "ragged sphere" of radius R, which we denote by S_R, since all contributions at all surfaces cancel except at the end points of the summation in i. We get

$$I_n = \lim_{R \to \infty} \iint_{S_R} \hat{\boldsymbol{\nu}}(\boldsymbol{y}) u_n(\boldsymbol{y}) \mathrm{d}S$$
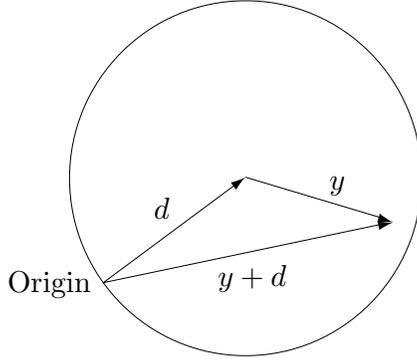
Figure A1. Translation of the origin.

This surface integral is in the limit $R \to \infty$ identical to an integral over the sphere $y = R + 1$, i.e.,

$$I_n = \lim_{R \to \infty} \iint_{y=R+1} \hat{\boldsymbol{y}} u_n(\boldsymbol{y}) \mathrm{d}S$$

since the difference between the surface integral is

$$\iiint_{V_R} \nabla u_n(\boldsymbol{y}) \mathrm{d}v$$

where V_R is the volume between S_R and $y = R + 1$. This integral approaches zero in the limit $R \to \infty$, since the volume V_R is of the order $O(R^2)$ as $R \to \infty$ and $|\nabla u_n(\boldsymbol{y})| = O(R^{-l-2})$ as $R \to \infty$. The integral over the sphere $y = R$ is easy to calculate. We have

$$I_n = \delta_{n,\mathrm{e}01} \lim_{R \to \infty} \iint_{y=R} \hat{\boldsymbol{y}} u_{\mathrm{e}01}(\boldsymbol{y}) \mathrm{d}S = \delta_{n,\mathrm{e}01} a^2 \sqrt{\frac{4\pi}{3}} \hat{\boldsymbol{e}}_3 \qquad \text{(B1)}$$

The second integral of interest is

$$\iint_{\partial Y} \hat{\boldsymbol{\nu}}(\boldsymbol{y}) v_{\mathrm{e}01}(\boldsymbol{y}) \mathrm{d}S = \sqrt{\frac{3}{4\pi}} \iint_{\partial Y} \hat{\boldsymbol{\nu}}(\boldsymbol{y}) \frac{y_3}{a} \mathrm{d}S$$

$$= \sqrt{\frac{3}{4\pi}} \hat{\boldsymbol{e}}_3 \iiint_Y \frac{1}{a} \mathrm{d}v = \sqrt{\frac{3}{4\pi}} \hat{\boldsymbol{e}}_3 \frac{1}{a} \qquad \text{(B2)}$$

APPENDIX C. THE TRANSLATION MATRICES $P_{nn'}$

In this section, we review the entries of the translation matrix, $P_{nn'}$, in (A1). More details are found in [3].

If (d, η, α) are the spherical coordinates of $\boldsymbol{d}$, the matrix entries of $P_{nn'}(\boldsymbol{d})$ are [3, 6]

$$P_{\sigma m l \sigma' m' l'}(\boldsymbol{d}) = (-1)^{m'} B_{mlm'l'}(d, \eta) \left\{ \begin{array}{c} \cos(m - m')\alpha \\ (-1)^{\sigma'} \sin(m - m')\alpha \end{array} \right\}$$

$$+(-1)^{\sigma} B_{ml-m'l'}(d, \eta) \left\{ \begin{array}{c} \cos(m + m')\alpha \\ (-1)^{\sigma} \sin(m + m')\alpha \end{array} \right\} \left\{ \begin{array}{c} \sigma = \sigma' \\ \sigma \neq \sigma' \end{array} \right.$$

where $(-1)^{\sigma} = 1$ if $\sigma = $ e and $(-1)^{\sigma} = -1$ if $\sigma = $ o and where

$$B_{mlm'l'}(\boldsymbol{d}) = \lim_{k \to 0} \mathcal{B}_{mlm'l'}(\boldsymbol{d}) \frac{j_{l'}(ka)}{h_l^{(1)}(ka)} = \lim_{k \to 0} \mathcal{B}_{mlm'l'}(\boldsymbol{d}) \frac{\mathrm{i}(ka)^{l+l'+1}}{(2l'+1)!!(2l-1)!!}$$

The matrix $\mathcal{B}_{mlm'l'}(d, \eta)$ is detailed as [3]

$$\mathcal{B}_{mlm'l'}(d, \eta) = (-1)^{m+m'} \sqrt{\frac{(2-\delta_{m0})(2-\delta_{m'0})}{4}} \sum_{\lambda=|l-l'|}^{l+l'} (-1)^{(l'-l+\lambda)/2}(2\lambda+1)$$

$$\sqrt{\frac{(2l+1)(2l'+1)(\lambda-(m-m'))!}{(\lambda+m-m')!}}$$

$$\left(\begin{array}{ccc} l & l' & \lambda \\ 0 & 0 & 0 \end{array} \right) \left(\begin{array}{ccc} l & l' & \lambda \\ m & -m' & m'-m \end{array} \right) P_{\lambda}^{m-m'}(\cos\eta) h_{\lambda}^{(1)}(kd)$$

where $\left(\begin{array}{ccc} \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot \end{array} \right)$ is the Wigner 3-j symbol [6]. The limit process $k \to 0$ gives the final result.

$$\mathcal{B}_{mlm'l'}(d, \eta) = (-1)^{m+m'+l'} \sqrt{\frac{(2-\delta_{m0})(2-\delta_{m'0})}{4}} \frac{(2l+2l'+1)!!}{(2l-1)!!(2l'+1)!!}$$

$$\sqrt{\frac{(2l+1)(2l'+1)(l+l'-(m-m'))!}{(l+l'+m-m')!}}$$

$$\left(\begin{array}{ccc} l & l' & l+l' \\ 0 & 0 & 0 \end{array} \right) \left(\begin{array}{ccc} l & l' & l+l' \\ m & -m' & m'-m \end{array} \right) P_{l+l'}^{m-m'}(\cos\eta) \left(\frac{a}{d} \right)^{l+l'+1}$$

where the Wigner 3-j symbol has the explicit expression [6]

$$\left(\begin{array}{ccc} l & l' & l+l' \\ m & -m' & m'-m \end{array} \right) = (-1)^{l-l'+m-m'}$$

$$\times \sqrt{\frac{(2l)!(2l')!(l+l'-(m-m'))!(l+l'+m-m')!}{(2l+2l'+1)!(l+m)!(l-m)!(l'+m')!(l'-m')!}}$$

The first explicit values for the lowest order terms used in this paper are

$$P_{\mathrm{e01e01}}(\boldsymbol{d}) = -2P_2(\cos\eta)\left(\frac{a}{d}\right)^3 = (1-3\cos^2\eta)\left(\frac{a}{d}\right)^3$$

and

$$\left\{\begin{array}{l} P_{\mathrm{e01e03}}(\boldsymbol{d}) = -4\sqrt{\frac{3}{7}}P_4(\cos\eta)\left(\frac{a}{d}\right)^5 \\[3mm] P_{\mathrm{e01e23}}(\boldsymbol{d}) = -\sqrt{\frac{1}{35}}P_4^2(\cos\eta)\cos 2\alpha\left(\frac{a}{d}\right)^5 \\[3mm] P_{\mathrm{e03e01}}(\boldsymbol{d}) = -4\sqrt{\frac{7}{3}}P_4(\cos\eta)\left(\frac{a}{d}\right)^5 \\[3mm] P_{\mathrm{e23e01}}(\boldsymbol{d}) = -\frac{1}{3}\sqrt{\frac{7}{5}}P_4^2(\cos\eta)\cos 2\alpha\left(\frac{a}{d}\right)^5 \end{array}\right\}$$

APPENDIX D. THE SUM $S_{nn'}$

An important matrix of frequent use in this paper is

$$S_{nn'} = \lim_{R\to\infty}\sum_{\substack{\mathbb{Z}^3\ni i\neq(000)\\|\boldsymbol{t}_i|\leq R}} P_{nn'}(\boldsymbol{t}_i) = \lim_{R\to\infty}\sum_{0<|(ijk)|\leq R} P_{nn'}(\boldsymbol{t}_{(ijk)}) \quad \text{(D1)}$$

where the translation $\boldsymbol{t}_{(ijk)} = i\hat{\boldsymbol{e}}_1 + j\hat{\boldsymbol{e}}_2 + k\hat{\boldsymbol{e}}_3$ and $|(ijk)| = (i^2 + j^2 + k^2)^{1/2}$.

The symmetry of the solution (even in $y_1 \to -y_1$ and $y_2 \to -y_2$, and odd in $y_3 \to -y_3$) implies that the only non-vanishing entries of $S_{nn'}$ occur when $l+l'$ is an even integer, $\sigma = \sigma'$, and $m+m'$ is an even integer. The dependence of a is, see Appendix C

$$S_{nn'} = O(a^{l+l'+1})$$

We denote the spherical polar and azimuth angles of the translation $\boldsymbol{t}_{(ijk)}$ by η_{ijk} and α_{ij}, respectively. In a cubic lattice we have

$$\cos\eta_{ijk} = \frac{k}{\sqrt{i^2+j^2+k^2}} \qquad \cos\alpha_{ij} = \frac{i}{\sqrt{i^2+j^2}}$$

We proceed by computing the first few elements of the matrix $S_{nn'}$. Several sums are of interests in this context. We get

$$a^{-3}S_{\text{e01e01}} = \lim_{R \to \infty} \sum_{0 < |(ijk)| \le R} \frac{i^2 + j^2 - 2k^2}{(i^2 + j^2 + k^2)^{5/2}}$$

$$= 8 \sum_{\substack{i,j,k=1 \\ i^2+j^2+k^2 \le R^2}} \frac{i^2 + j^2 - 2k^2}{(i^2 + j^2 + k^2)^{5/2}} + 4 \sum_{\substack{i,j=1 \\ i^2+j^2 \le R^2}} \frac{1}{(i^2 + j^2)^{3/2}}$$

$$+ 8 \sum_{\substack{i,k=1 \\ i^2+k^2 \le R^2}} \frac{i^2 - 2k^2}{(i^2 + k^2)^{5/2}} - 4 \sum_{k=1}^{R} \frac{1}{k^3} + 4 \sum_{i=1}^{R} \frac{1}{i^3}$$

$$= 4 \sum_{\substack{i,j=1 \\ i^2+j^2 \le R^2}} \frac{i^2 + j^2 + 2i^2 - 4j^2}{(i^2 + j^2)^{5/2}} = 0$$

Therefore, $S_{\text{e01e01}} = 0$. Similarly, we have

$$\begin{cases} S_{\text{e01e23}} = \sqrt{\frac{45}{28}} a^5 \lim_{R \to \infty} \sum_{0 < |(ijk)| \le R} \frac{(i^2 - j^2)(i^2 + j^2 - 6k^2)}{(i^2 + j^2 + k^2)^{9/2}} = 0 \\[4mm] S_{\text{e23e01}} = \sqrt{\frac{35}{2}} a^5 \lim_{R \to \infty} \sum_{0 < |(ijk)| \le R} \frac{(i^2 - j^2)(i^2 + j^2 - 6k^2)}{(i^2 + j^2 + k^2)^{9/2}} = 0 \end{cases}$$

The first non-vanishing terms in powers of a are

$$a^{-5}S_{\text{e01e03}} = -\sqrt{\frac{3}{28}} \lim_{R \to \infty} \sum_{0 < |(ijk)| \le R} \frac{3i^4 + 6i^2j^2 + 3j^4 - 24i^2k^2 - 24j^2k^2 + 8k^4}{(i^2 + j^2 + k^2)^{9/2}}$$

$$= -\sqrt{\frac{3}{28}} \left\{ 8 \sum_{i,j,k=1}^{\infty} \frac{3i^4 + 6i^2j^2 + 3j^4 - 24i^2k^2 - 24j^2k^2 + 8k^4}{(i^2 + j^2 + k^2)^{9/2}} \right.$$

$$+ 4 \sum_{i,j=1}^{\infty} \frac{3i^4 + 6i^2j^2 + 3j^4}{(i^2 + j^2)^{9/2}} + 8 \sum_{i,k=1}^{\infty} \frac{3i^4 - 24i^2k^2 + 8k^4)}{(i^2 + k^2)^{9/2}}$$

$$\left. + 4 \sum_{i=1}^{\infty} \frac{3}{i^5} + 2 \sum_{k=1}^{\infty} \frac{8}{k^5} \right\}$$

which we can rearrange by symmetry to

$$a^{-5}S_{\text{e01e03}} = -\sqrt{\frac{3}{28}} \left\{ 112 \sum_{i,j,k=1}^{\infty} \frac{i^4 - 3i^2j^2}{(i^2 + j^2 + k^2)^{9/2}} \right.$$

$$+ 56 \sum_{i,j=1}^{\infty} \frac{2i^4 - 3i^2 j^2}{(i^2 + j^2)^{9/2}} + 28 \sum_{i=1}^{\infty} \frac{1}{i^5} \Bigg\}$$

The other term of the same magnitude is

$$a^{-5} S_{\text{e03e01}} = -\sqrt{\frac{7}{12}} \lim_{R \to \infty} \sum_{0 < |(ijk)| \leq R} \frac{3i^4 + 6i^2 j^2 + 3j^4 - 24i^2 k^2 - 24j^2 k^2 + 8k^4}{(i^2 + j^2 + k^2)^{9/2}}$$

From this result we have

$$S_{\text{e01e03}} S_{\text{e03e01}} = 196 a^{10} (S_1 + 2S_2 - S_3)^2 \qquad \text{(D2)}$$

The pertinent sums are

$$S_1 = \sum_{i,j,k=1}^{\infty} \frac{3i^2 j^2 - i^4}{(i^2 + j^2 + k^2)^{9/2}}, \quad S_2 = \sum_{i,j=1}^{\infty} \frac{3i^2 j^2 - 2i^4}{(i^2 + j^2)^{9/2}}, \quad S_3 = \sum_{i=1}^{\infty} \frac{1}{i^5}$$

The sums are positive and have approximate values as

$$S_1 \approx 0.021, \qquad S_2 \approx 0.033, \qquad S_3 \approx 1.037$$

and we get

$$S_{\text{e01e03}} S_{\text{e03e01}} \approx 155 a^{10}$$

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