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Study Of Probabilistic Characteristics Of Local Field Fluctuations In Isotropic Two Phase Composites: Conductivity Type Problems

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STUDY OF PROBABILISTIC CHARACTERISTICS OF LOCAL FIELD FLUCTUATIONS IN ISOTROPIC TWO PHASE COMPOSITES: CONDUCTIVITY TYPE PROBLEMS

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CHAPTER 1 INTRODUCTION

Heterogenous materials comprise a family of widely tailorable materials that are abound in nature as well as man made products. The focus of this work is on a basic category of these materials: composites comprised of two isotropic phases. This category retains a rich set of widely studied materials due to their immense industrial utility. It includes materials with periodic characteristics such as laminated structures, as well as random materials which can only be described in statistical terms. Some examples of these random materials are fiber and particulate reinforced materials such as polymer matrix and metal matrix composites. This work considers the more narrow category of random composites, which can be further decomposed into macroscopically isotropic or anisotropic cases. This work gives solutions for the special case of macroscopically isotropic materials which provides the foundation for expanding to the more general anisotropic case.

The local fields for problems of elasticity (e.g. stresses, strains, etc.) within composites are the major interest in engineering since they drive critical processes such as fatigue and fracture. A similar, but much simpler problem, arises from problems of electrical conductivity. Since the results for electrical conductivity are also mathematically equivalent to heat conduction as well as dielectric polarization, these results are also useful for problems of thermal and electric breakdown as well as effective conductivity of composites. Although problems of conductivity do not have as many practical applications as those of elasticity, this work is focused on this simpler task, electric fields, bearing in mind the methods developed can be extended further to the case of elasticity problems.

The primary results in the field of heterogenous materials has been in methods for estimating the effective properties. For electrical conduction problems this is the effective conductivity, and for elastic problems, the effective elastic constants. Here a brief discussion is given with details given in Section 1.1. In the case we only have knowledge of the one point material probability density functions (PDF), which for two phase composites are the concentrations, effective properties for any two phase composite fall within the Voight and

Reuss bounds. For the focus of this work, statistically isotropic composites, effective properties fall in a much more narrow band, the Hashin-Shtrikman bounds [1]. Here, statistically isotropic composite is defined as the two material PDF depending only upon the spacing between two points of observation but not the direction. With these two sets of bounds, we do gain an estimate of the average field within each phase, but nothing on the distributions within each phase. To develop improved physics based models which depend on local fields such as fatigue, fracture, and thermal breakdown, the statistical characteristics of internal fields is required.

With full knowledge of the statistical characteristics of the internal fields we know not only the effective properties since they can be found through methods of homogenization using the first and second moments of the internal fields (e.g. see 2.58), but also now have the ability to develop the desired improved physics based models that depend on detailed knowledge of local fields. This work is focused on first understanding the nature of the internal field statistics within a particulate composite, then since the one and two point distributions can be found from experimental observations, developing a method using this additional data to not only make improvements upon the Hashin-Shtrikman bounds thereby providing an even more accurate estimate of effective properties, but also give an expanded level of insight into the internal fields.

In Chapter 2, probability distributions of electric field and electric potential in two-phase particulate composite materials with randomly placed spherical inclusions are found in the limit of small particle concentration by conducting a statistical superposition of the solution for a single spherical inclusion. This analytical solution provides detailed insight into the full statistics of internal fields within a composite of randomly placed spheres. Since this result arises from a statistical superposition of the solution of a single spherical inclusion, it also retains the feature of having a potential field.

Within this work, only steady state solutions are sought, and in the case for both conductivity and elasticity problems the true solution is a potential field. For elasticity the field is position, and for conductivity it is temperature or electric potential. Since true solutions have the feature of potentiality, but analytically computing the internal fields may not be possible for other microstructures, a method of approximating internal fields which retains the feature of potentiality is necessary to ensure a realistic estimate is developed.

In Chapter 3, a framework for considering a finite number of field fluctuations within a random statistically isotropic two phase composite is developed. It is the major achievement of this work that sufficient analytical simplifications of the microstructural and joint microstructure internal field statistics, as well as the potentiality and positive definiteness conditions, were reduced to only two unknowns in the case of three fluctuations and have been developed to easily allow consideration of additional fluctuations. Since constraints for potentiality were formed, the solution, probabilities and values of field fluctuations, can be determined using the variational principle of homogenization in statistical terms.

In Chapter 4 the variational principle of homogenization in statistical terms [2] is used to develop an approximate solution in the case of three field fluctuations in one phase and a homogenous field in the other phase. For future work, the approach is easily extended to additional field fluctuations, which will improve the resolution and prediction of the internal fields.

In Chapter 5, an outline of the process necessary to generalize the solutions developed from the two dimensional case to three is presented, and the application of the Hashin-Shtrikman variational principle [3] briefly discussed.

1.1 Homogenization of Two Phase Composites

This section summarizes important homogenization results in the case of a two phase composite with isotropic phases. For further details and discussion, see the reviews of this field by Torquato [4] and Berdichevsky [3]. Homogenization is a procedure which allows the precise description of the averaged properties of a heterogeneous media. For the case of conductivity, effective properties by homogenization are determined from the variational problem

$$a_{ij}^{\text{eff}} v_i v_j \leq \min_{u \in X} \frac{1}{|V|} \int_V a_{ij}(x)(v_i + u_i)(v_j + u_j)\, dx, \tag{1.1}$$

where x is a point in space, a_{ij} are conductivities, v_i average field over the composite, u_i field fluctuations within the composite, and X are some constraints (e.g. boundary conditions). Here summation over i and j is implied and they run values from 1 to the dimensionality of the problem (i runs $1, 2$ for two dimensional problems and $1, 2, 3$ for three dimensional problems).

Considering two phase composites comprised of isotropic phases

$$a_{ij}(x) = a_1 \quad \text{for } x \in V_1 \quad \text{and } a_{ij}(x) = a_2 \quad \text{for } x \in V_2 \quad \text{for any } i \text{ or } j,$$

the classical solutions are the Voight, Reuss, and the Hashin-Shtrikman [1] bounds on effective conductivity. The Voight and Reuss solutions provide an upper and lower bound on effective conductivity, respectively, for any composite and the Hashin-Shtrikman solution provides a more narrow set of bounds in the case of a macroscopically isotropic composite. First, the origin of the Voight and Reuss bounds will be introduced, then the Hashin-Shtrikman bounds which originate from an alternative means will be presented.

These solutions arise from the approximation of the field fluctuations in each phase of the composite by a homogenous field

$$f_1(\vec{u}) = c_1 \delta\left(\vec{u} - \vec{R}\right) \quad \text{and} \quad f_2(\vec{u}) = c_2 \delta\left(\vec{u} - \vec{Q}\right).$$

Here, $f_1(\vec{u})$ is the probability of observing field value $\vec{u}$ within the first phase and similarly for $f_2(\vec{u})$, $\vec{R}$ the field fluctuation in the first phase and similarly for $\vec{Q}$ in the second, where $\delta(x)$ the Kronecker delta function ($\delta(0) = 1$ else 0). Also c_1 and c_2 are the volume concentrations of phase one and two, respectively.

These solutions are all approximate since the field is not actually homogenous within each phase for any composite, except for the trivial degenerated case of a homogenous composite (i.e. Reuss Bound) or laminated composites which the flux is orthogonal to the laminations (i.e. the Voight Bound). In the case of a particulate composite this can be seen, e.g., in Chapter 2.

Continuing with the case of homogenous field fluctuations within a two phase composite, the field fluctuations by definition must vanish over the composite

$$c_1\vec{R} + c_2\vec{Q} = 0 \ \text{ then } \ \vec{Q} = -\frac{c_1}{c_2}\vec{R}.$$

Without loss of generality, let the direction of the applied field $\vec{v}$ be in the 1-direction

$$a_{11}^{\text{eff}} v_1 v_1 \le \min_{u\in X} a_1 c_1 \left(v_1 + R_1\right)^2 + a_2 c_2 \left(v_1 - \frac{c_1}{c_2} Q_1\right)^2 + a_1 c_1 \sum_{i=2}^{\text{dim}} \left(R_i\right)^2 + a_2 c_2 \sum_{i=2}^{\text{dim}} \left(\frac{c_1}{c_2} Q_i\right)^2,$$

where dim is the spatial dimension of the problem, c_1 is the volume concentration of the first phase, and similarly for c_2 in the second phase

$$c_1 = \frac{1}{|V|}\int\limits_{V_1} dx \ \text{ and } \ c_2 = \frac{1}{|V|}\int\limits_{V_2} dx.$$

As noted the Voight solution corresponds to the special case of a homogenous field throughout the entire composite (i.e. $\left|\vec{R}\right| = 0$), after dividing through by $v_1 v_1$ we have

$$a_{11}^{\text{eff}} \le a_1 c_1 + a_2 c_2,$$

Minimizing over the orthogonal direction (i.e. $R_i = 0$ for $i \neq 1$), dividing through by $(v_1)^2$, and dropping of subscripts results in the simple relationship

$$a^{\text{eff}} \le \min_{u\in X} \ a_1 c_1 \left(1 + \frac{\left|\vec{R}\right|}{|\vec{v}|}\right)^2 + a_2 c_2 \left(1 - \frac{c_1}{c_2}\frac{\left|\vec{R}\right|}{|\vec{v}|}\right)^2. \tag{1.2}$$

The Voight and Reuss solutions are the arithmetic and harmonic averages of the phase conductivities

$$a^{\text{eff}} = a_1c_1 + a_2c_2 \quad \text{and} \quad a^{\text{eff}} = \left(\frac{c_1}{a_1} + \frac{c_2}{a_2}\right)^{-1} = \frac{a_1a_2}{a_1c_2 + a_2c_1},$$

respectively. As noted the Voight solution corresponds to a homogenous field throughout the entire composite (i.e. $\left|\vec{R}\right| = 0$), and the Reuss solution when minimization is executed free from constraint. The field fluctuation by definition is zero for the Voight solution $\vec{R} = 0$, and for the Reuss

$$\frac{\left|\vec{R}\right|}{|\vec{v}|} = c_2\frac{a_2 - a_1}{a_1c_2 + a_2c_1}.$$

The Hashin-Shtrikman bounds [1] bring the Voight and Reuss bounds tighter by the constraint that the composite is macroscopically isotropic. To do this, they used the principal that the energy contained within the heterogenous composite is the same as the homogenous approximation, and then make an additional assumption that the field is homogenous within each phase. Under these assumptions, an example of a special periodic composite consisting of an infinite suspension of coated spheres is given to determine that for macroscopically isotropic two phase composites, a^{eff} has the bounds

$$a_1 + c_1\left(\frac{1}{a_2 - a_1} + \frac{c_2}{3a_1}\right)^{-1} \quad \text{and} \quad a_2 + c_2\left(\frac{1}{a_1 - a_2} + \frac{c_1}{3a_2}\right)^{-1}.$$

These same bounds can be found more rigorously though the Hashin-Shtrikman variational principle as briefly discussed in Section 5.2.

These important solutions will be used to test the extent that the approximations developed in Chapters 2 and 3 hold.

The methods developed in Chapter 3 yield in particular cases these classical results, but can also incorporate more subtle characteristics of microstructures like correlation functions. The Voight, Reuss, as well as Hashin-Shtrikman bounds correspond to the case where the field fluctuations are homogenous within each phase, and the methods developed in Chapter 3 generalize these results to the case of the field fluctuations being non-homogenous through an alternative method of homogenization.

1.2 Electric Field Fluctuations in Conductors with Spherical Inclusions

In Chapter 2, the probability distributions of electric field and electric potential in two-phase particulate composite materials with randomly placed spherical inclusions are found in the limit of small particle concentration.

The previous analytical results on the statistics of these type of internal fields arise from the computation of electric fields from randomly placed charges (ions), dipoles, and quadrupoles [5, 6, 7, 8], as well as similar results in related probability distributions: gravitational fields within a stellar system comprised of randomly placed masses [9, 10], stresses caused by point defects in crystallographic structures [11, 12], velocity distributions caused by vortices, temperature field in nuclear reactors, etc. (brief summary given in [13]). These solutions are all described by stable distributions [13] which include the special cases of Holtsmark, Cauchy, and normal distributions. In these previous solutions, the total field disturbance is found by summing the contribution of single defects over infinite space with a fixed number density.

These previous analytical solutions for point size defects are generalized to finite size particles in Chapter 2. The internal fields associated with point sized defects create a singular field value, while for particles field values are finite. Therefore distributions for finite sized particles are distinct from point defects due to the differing nature of the fields. Previous efforts for approaching the issue of finite sized defects have been only through numerical means in the two dimensional case [14, 15], but these results do not have generality; these previous results are only applicable to the concentrations and conductivities studied. In this work, a general analytical solution was found for particulate composites such that the solution is applicable to any concentration and conductivities.

The probability distributions were first computed over the entire composite and then in the matrix of the composite. Then, as a consequence, the distribution which occurs within particles is also known. The result found is approximate in the sense that particles were treated as independent and identically distributed, and the solution for the composite was simply taken as the summation of each particle's effect on the composite.

Asymptotic analysis and statistics of these distributions was found and results are compared against known bounds on effective properties and variances. Interestingly, distributions for electric field are found to be independent of particle size distribution. By comparing this result against the Hashin-Shtrikman bounds it was determined that this result is not valid for concentrations over 0.26.

As the work of Holtsmark provided a motivation for the study of a diverse set of physical issues with point sized defects, the approach given in this work can be applied further to other cases with finite sized defects. This includes both differing physics, such as the elastic case, as well as alternate microstructures such as the case of ellipsoidal particles in a matrix.

Next the work of Voight, Reuss, and Hashin-Shtrikman of approximating the field fluctuations within each phase as homogenous is generalized by allowing the field fluctuations to take additional values. To do this, a second means of homogenization was conducted: homogenization in probabilistic terms using the variational principle for probabilistic measure [2].

1.3 Internal Fields and Microstructure in Probabilistic Terms

In this Section the variational principle for homogenization in probabilistic terms for the case of an isotropic composite comprised of two isotropic phases is summarized. For further details and discussion on the variational principal see [3] and for discussion of the correlation functions in two phase composites [4].

The effective characteristics and statistics of the local fields can be found from the variational principle for homogenization in probabilistic terms

$$a_{ij}^{\mathrm{eff}} v_i v_j \leq \min_{f(a,u)\in X} \int a_{ij} \left(v_i + u_i\right) \left(v_j + u_j\right) f\left(a,u\right) da \; du \tag{1.3}$$

where a_{ij} are conductivities, v_i average electric field, u_i electric field fluctuations, $f\left(a,u\right)$ the joint one-point probability density of conductivities, and u_i electric field fluctuations, with the minimization conducted subject to some constraints X (e.g. potentiality, probabilties, etc.). Here, the composite microstructure and field fluctuations are defined statistically.

To uniquely describe a particular composite‘s microstructure, an infinite series of probability distributions describing the spacial distribution of material conductivity is required. The first description in this infinite series is the one point probability distribution which has been previously introduced, c_1 and c_2, the volume distribution of phases. Next, there are two point characteristics $f_{11}(\vec{\tau})$, $f_{12}(\vec{\tau})$, $f_{21}(\vec{\tau})$, and $f_{22}(\vec{\tau})$, where for example, $f_{12}(\vec{\tau})$ is defined as the probability of sampling two points separated by the vector $\vec{\tau}$ over the composite and having the first point in phase one and the second point in phase two. Higher characteristics, i.e. three, four, etc. point distributions, exist and comprise an infinite chain of statistical descriptors. These distributions are denoted as a chain of constraints since they are related to each other. For instance, the two point distribution must be compatible with the one point distribution

$$c_1 = f_{11}(\vec{\tau}) + f_{12}(\vec{\tau}) \quad \text{and} \quad c_2 = f_{21}(\vec{\tau}) + f_{22}(\vec{\tau}) \,. \tag{1.4}$$

Field fluctuations are also described by an additional infinite series of probabilities that are joint with microstructural characteristics. The joint one point probabilities $f_1(\vec{u})$ and $f_2(\vec{u})$ statistically describe the field fluctuations within each phase. For example, $f_1(\vec{u})$ is the probability of sampling the composite and having an observation in phase 1 with the field fluctuation $\vec{u}$. The joint two point probabilities $f_{11}(\vec{u};\vec{\tau},\vec{u}')$, $f_{12}(\vec{u};\vec{\tau},\vec{u}')$, and $f_{22}(\vec{u};\vec{\tau},\vec{u}')$ relate the field fluctuations and the material conductivity probabilities at two points separated by the vector $\vec{\tau}$. For example, $f_{12}(\vec{u};\vec{\tau},\vec{u}')$ defines the probability of having two points separated by distance $\vec{\tau}$ with the first point in phase 1 and with field fluctuation $\vec{u}$, as well as the second point in phase 2 with field fluctuation $\vec{u}'$. Again, these distributions are related to each other, the joint two point distributions must be compatible with the joint one point distribution

$$
\begin{aligned}
f_1(\vec{u}) &= \int \left(f_{11}(\vec{u};\vec{\tau},\vec{u}') + f_{12}(\vec{u};\vec{\tau},\vec{u}')\right) d\vec{u}' \\
\text{and} \quad f_2(\vec{u}) &= \int \left(f_{21}(\vec{u};\vec{\tau},\vec{u}') + f_{22}(\vec{u};\vec{\tau},\vec{u}')\right) d\vec{u}'.
\end{aligned}
$$

Additionally, the the joint probability distributions must be compatible with the microstructural characteristics

$$
\begin{aligned}
c_1 &= \int f_1(\vec{u})\, d\vec{u}, \quad c_2 = \int f_2(\vec{u})\, d\vec{u}, \\
f_{11}(\vec{\tau}) &= \int f_{11}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u} d\vec{u}', \quad f_{12}(\vec{\tau}) = \int f_{12}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u} d\vec{u}', \\
f_{21}(\vec{\tau}) &= \int f_{21}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u} d\vec{u}', \quad \text{and} \quad f_{22}(\vec{\tau}) = \int f_{22}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u} d\vec{u}'
\end{aligned}
$$

with similar relations for higher order correlation functions. And of course, all of these probability distributions must be must be non-negative for all values

$$
\begin{aligned}
0 &\leq c_1,\ 0 \leq c_2,\ 0 \leq f_1(\vec{\tau}),\ 0 \leq f_2(\vec{\tau}),0 \leq f_1(\vec{u}),\ 0 \leq f_2(\vec{u}), \\
0 &\leq f_{11}(\vec{\tau}),\ 0 \leq f_{12}(\vec{\tau}),\ 0 \leq f_{21}(\vec{\tau}),\ 0 \leq f_{22}(\vec{\tau}), \\
0 &\leq f_{11}(\vec{u};\vec{\tau},\vec{u}'),\ 0 \leq f_{12}(\vec{u};\vec{\tau},\vec{u}'),\ 0 \leq f_{21}(\vec{u};\vec{\tau},\vec{u}'),\ \text{and } 0 \leq f_{22}(\vec{u};\vec{\tau},\vec{u}').
\end{aligned}
\tag{1.5}
$$

There are also symmetries to probability distributions (e.g. $f_{12}(\vec{\tau}) = f_{21}(-\vec{\tau})$).

The composite to be studied is taken to lack long range correlation

$$
f_{11}(\vec{\tau}) = c_1 c_1, \quad f_{12}(\vec{\tau}) = c_1 c_2, \ \text{and} \ \ f_{22}(\vec{\tau}) = c_2 c_2 \ \text{as} \ |\vec{\tau}| \to \infty \tag{1.6}
$$

and be statistically invariant with respect to mirror image

$$
f_{12}(\vec{\tau}) = f_{12}(-\vec{\tau}) \quad \text{then} \quad f_{12}(\vec{\tau}) = f_{21}(\vec{\tau}) \tag{1.7}
$$

and phases separated by a surface

$$f_{11}(\vec{\tau}) = c_1, \quad f_{12}(\vec{\tau}) = 0, \text{ and } \; f_{22}(\vec{\tau}) = c_2 \text{ as } |\vec{\tau}| \to 0.$$

Then with (1.4), (1.6), and (1.7) the microstructures two point statistics can be described by a single function

$$\begin{aligned} f_{11}(\vec{\tau}) &= c_1c_1 + c_1c_2h_o(\vec{\tau}), \quad f_{12}(\vec{\tau}) = c_1c_2 - c_1c_2h_o(\vec{\tau}), \\ f_{21}(\vec{\tau}) &= f_{12}(\vec{\tau}), \quad \text{and} \;\; f_{22}(\vec{\tau}) = c_2c_2 + c_1c_2h_o(\vec{\tau}) \end{aligned}$$

where

$$h_o(\vec{\tau}) = 0 \;\text{ as }\; |\vec{\tau}| \to \infty, \;\; h_o(\vec{\tau}) = 1 \;\text{ as }\; |\vec{\tau}| \to 0, \;\; \text{and} \;\; 0 \le h_o(\vec{\tau}) \;\; \text{for all} \;\; \vec{\tau}. \tag{1.8}$$

While, one and two point distributions can be found from experimental observations, the higher order probability distributions are difficult to determine experimentally. Therefore, in this approximation the infinite chain of statistics will be truncated, which leads to a new constraint to impose positive definiteness of the joint two point probability. The condition of positive definiteness requires for any ϕ_1,ϕ_2

$$\begin{aligned} 0 \;\le\; & \int ((f_{11}(\vec{u};\vec{\tau},\vec{u}') - f_1(\vec{u})\, f_1(\vec{u}'))\, \phi_1(\vec{u})\, \phi_1(\vec{u}') \\ & +2\left(f_{12}(\vec{u};\vec{\tau},\vec{u}') - f_1(\vec{u})\, f_2(\vec{u}')\right) \phi_1(\vec{u})\, \phi_2(\vec{u}') \\ & +\left(f_{22}(\vec{u};\vec{\tau},\vec{u}') - f_2(\vec{u})\, f_2(\vec{u})\right) \phi_2(\vec{u})\, \phi_2(\vec{u}'))d\vec{u}d\vec{u}' \end{aligned} \tag{1.9}$$

The solution to the conductivity problem must be potential (i.e. for any realization $\Delta u = 0$). In statistical terms, this leads to the requirement that a function $B(\vec{\tau})$ exists and is related to probabilities by

$$-\frac{B(\vec{\tau})}{\partial\tau_i\partial\tau_j} = \int u_i u_j'(f_{11}(\vec{u};\vec{\tau},\vec{u}') - f_1(\vec{u})\,f_1(\vec{u}') + f_{12}(\vec{u};\vec{\tau},\vec{u}') - f_1(\vec{u})\,f_2(\vec{u}') \quad (1.10)$$
$$+f_{21}(\vec{u};\vec{\tau},\vec{u}') - f_2(\vec{u})\,f_1(\vec{u}') + f_{22}(\vec{u};\vec{\tau},\vec{u}') - f_2(\vec{u})\,f_2(\vec{u}'))d\vec{u}d\vec{u}',$$

and its Fourier transform is

$$B\left(\vec{k}\right)k_ik_j = \int u_i u_j'(f_{11}\left(\vec{u};\vec{k},\vec{u}'\right) - f_1(\vec{u})\,f_1(\vec{u}') + f_{12}\left(\vec{u};\vec{k},\vec{u}'\right) - f_1(\vec{u})\,f_2(\vec{u}') \quad (1.11)$$
$$+f_{21}\left(\vec{u};\vec{k},\vec{u}'\right) - f_2(\vec{u})\,f_1(\vec{u}') + f_{22}\left(\vec{u};\vec{k},\vec{u}'\right) - f_2(\vec{u})\,f_2(\vec{u}'))d\vec{u}d\vec{u}',$$

where

$$B\left(\vec{k}\right) = \int \mathrm{Exp}\left(i\ \vec{\tau}\cdot\vec{k}\right)B(\vec{\tau})\,d\vec{\tau} \geq 0. \quad (1.12)$$

Note that the average value of the field fluctuation $\vec{u}$ is zero

$$\int u_i\left(f_1(\vec{u}) + f_2(\vec{u})\right)d\vec{u} = 0$$

and the potentiality condition is then simply

$$B\left(\vec{k}\right)k_ik_j = \int u_i u_j'(f_{11}\left(\vec{u};\vec{k},\vec{u}'\right) + f_{12}\left(\vec{u};\vec{k},\vec{u}'\right) + f_{21}\left(\vec{u};\vec{k},\vec{u}'\right) + f_{22}\left(\vec{u};\vec{k},\vec{u}'\right))d\vec{u}d\vec{u}'.$$

Therefore, the solution to (1.3) with the constraints (1.4) through (1.12) is sought.

1.4 Non-Homogenous Fields: Two-Dimensional Case

As previously discussed, the classical results in homogenization for two phase composites comprised of isotropic phases provide bounds on the effective conductivity. These include the Voight and Reuss for all composites, as well as the more stringent Hashin-Shtrikman bounds for isotropic composites. These solutions were noted to approximate the field fluctuations in the composite by a homogenous field within each phase

$$f_1(\vec{u}) = \delta\left(\vec{u} - \vec{R}\right) \quad \text{and} \quad f_2(\vec{u}) = \delta\left(\vec{u} - \vec{Q}\right).$$

As shown in Chapter 2, the field for a particulate composite is not well represented by a homogenous one within each phase. Therefore, the previous solutions can be improved upon by making the local fields more realistic by increasing the number of allowable field fluctuations within each phase. Then, as the number of admissible field fluctuations increases, the approximation should improve and in the limit of infinite admissible values, be an exact result. A new procedure for finding these field fluctuations and their corresponding probabilities is introduced by conducting homogenization in probabilistic terms and using the Hashin-Shtrikman variational principle. First, the problem of electrical conductivity in a two dimensional composite is studied.

To do this, N field fluctuations are taken in phase 1 and M field fluctuations in phase 2

$$f_1(\vec{u}) = \sum_{\mu} p_\mu \delta\left(\vec{u} - \vec{R}_\mu\right) \quad \text{and} \quad f_2(\vec{u}) = \sum_{\alpha} q_\alpha \delta\left(\vec{u} - \vec{Q}_\alpha\right), \tag{1.13}$$

where p_μ and q_α are corresponding probabilities of field fluctuations, variable μ runs values $1...N$, and α runs values $1...M$. By taking the field fluctuations as a limited number of unique values the integral relationships for compatibility, non-negativity of probabilities, potentiality of electric field, and positive definiteness of joint two point probability can now be greatly simplified. Also, the special case of having a homogenous field in one of the two phases is considered, leading to a great deal of further analytical simplifications. As for the other phase, it is found that a minimum of three values of field fluctuations are required to satisfy the condition of potentiality. Since only solutions with non-homogenous fields that statisfy potentiality are sought, the case of three field values is studied in detail.

To complete the formulation of the problem, the microstructural descriptor $h_o(\vec{\tau})$ must be defined. In this effort, it is taken as $\mathrm{Exp}|-\vec{\tau}|$ which satisfies the requirements of $h_o(\vec{\tau})$

but also is convenient for further simplifications. In order to make this problem solvable, correlations between field fluctuations also must be defined. They are taken to be correlated to the maximum extent that remains to allow the non-negativity of probabilities to be satisfied.

With the assumption that the electric field takes three values in one phase and is homogenous in the other, it is found that many of the constraints are collapsed and the problem reduces to satisfaction of non-negativity of probabilities. All of the constraints except the ones which relate the field fluctuations within the same phase can be written compactly. An analytical solution was found that satisfies all constraints. This solution was found to fall within the Hashin-Shtrikman bounds and, therefore is an improvement.

1.5 Discussion of Results

In Section 1.2, the methods to compute probability distributions in Chapter 2 were summarized and similarly in Section 1.4 for the methods to compute probability distributions in Chapter 3. In this Section the effective coefficients for these two methods will be compared to the Hashin-Shtrikman bounds.

For the case of a particulate composite, the first phase has been taken to be the matrix and the second phase the particles. For particles which are less conducting than the matrix, a_{eff}/a_1 follows the upper Hashin-Shtrikman bound; the plots for particular values $a_2/a_1 = 2/5$ and $a_2/a_1 = 10^{-3}$ are shown in Figs. 1.1a and 1.1b, respectively. For particles which are more conducting than the matrix, a_{eff}/a_1 initially follows the lower Hashin-Shtrikman bound for small concentration and remains within the bounds for particle concentrations less than 0.17; the plots for particular values $a_2/a_1 = 10^4$, $a_2/a_1 = 10$, and $a_2/a_1 = 2$ are shown in Fig. 1.2a, 1.2b, and 1.2c.

The leading term approximation for small concentrations was found to be $a_{\text{eff}}/a_1 \approx (1 - 3c^* \varkappa)$, where c^* is particle concentration, this is consistent with previous results (see e.g. [21]). It supports the validity of the approximation for small concentration and explains why a_{eff}/a_1 coincides with the upper Hashin-Shtrikman bound for $a_2/a_1 < 1$, and the lower

Hashin-Shtrikman bound for $a_2/a_1 > 1$.

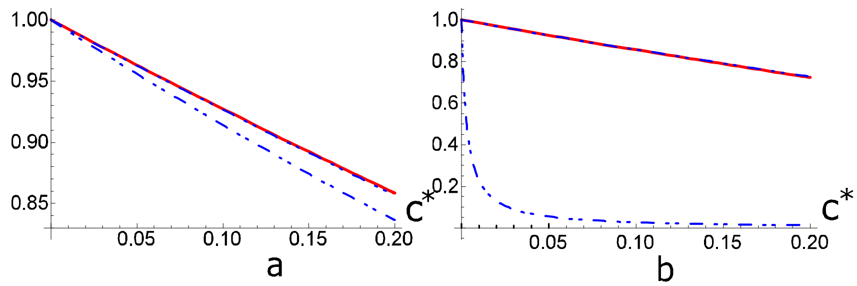

Figure 1.1: Effective coefficient a_{eff}/a_1 for composite with insulative particles (solid) and Hashin-Shtrikman bounds (point-dashed) ($a_2/a_1 = 2/5$ (a) and $a_2/a_1 = 10^{-3}$).

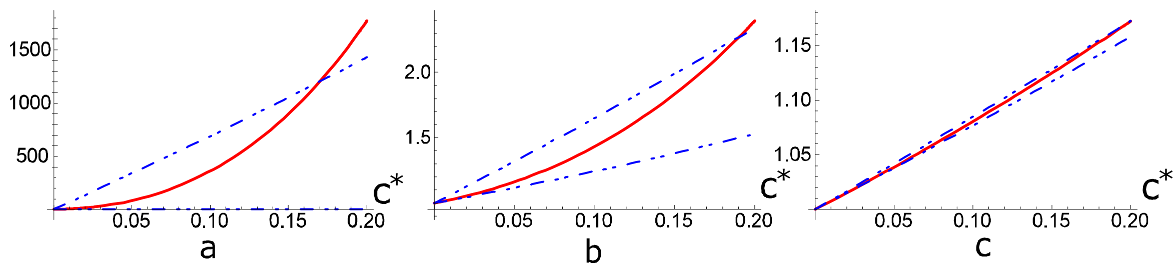

Figure 1.2: Effective coefficient a_{eff}/a_1 for composite with conductive particles (solid) and Hashin-Shtrikman bounds (point-dashed) ($a_2/a_1 = 10^4$ (a), $a_2/a_1 = 10$ (b), and $a_2/a_1 = 2$ (c)).

As for the results found in Chapter 3, they are valid over the entire range of concentrations. The probability distributions were computed with three field fluctuations in the first phase and one in the second phase. The selection for selecting the first phase to have multiple flucutations was arbitrary, the entire procedure is identical if the situation was reversed. Therefore, the procedure of Chapter 3 actually yields two results for a_{eff}/a_1, one corresponding to three fluctuations in the first phase and one corresponding to three in the second phase. These new results are compared against the Voight, Reuss, and Hashin-Shtrikman bounds in Figure 4.3 and fall within the Hashin-Shtrikman bounds for nearly all combinations of conductivity and phase concentrations.

The minimum of these two solutions is the upper bound for conductivity for the particular microstructure studied and it occurs when three field fluctuations are in the less conducting phase and one field value in the more conductive phase. Then, as shown in Figure 4.3 for

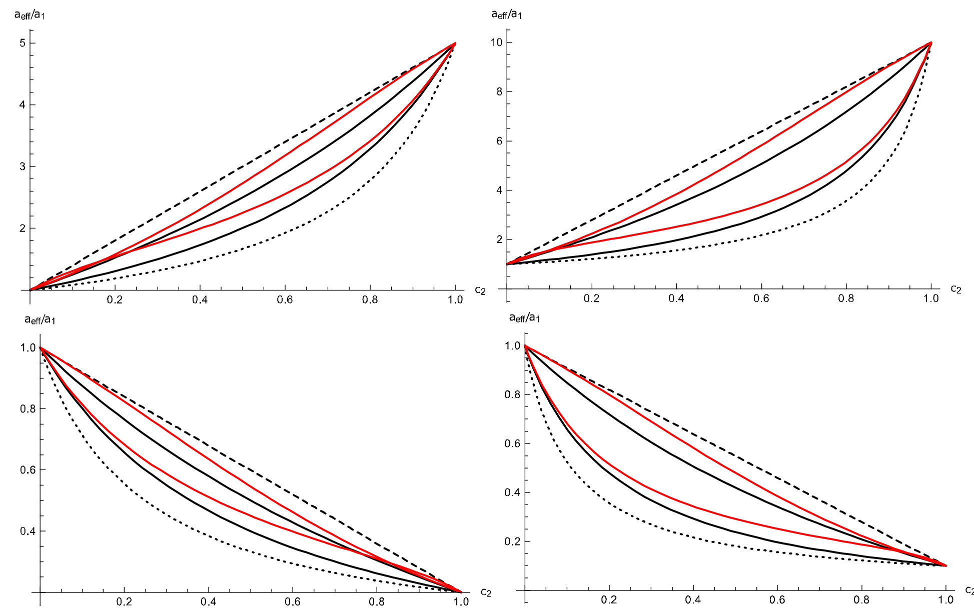

Figure 1.3: New result for a_{eff} (solid, grey) with Reuss (dashed), Voight, (dotted), and Hashin Shtrikman (solid, black) for four levels of contrast ($a_2/a_1 = 1/10$, top left), ($a_2/a_1 = 1/5$, top right), ($a_2/a_1 = 5$, bottom left), and ($a_2/a_1 = 10$, bottom right)

the particular microstructure studied ($h_o(\vec{\tau})$ =Exp$|-\vec{\tau}|$), a_{eff}/a_1 lies within a very narrow band: it is lower bound by the Hashin-Shtrikman bounds and upper bound by this new approximation.

Furthermore, this new result also provides what has not been previously available in approximations of this type: an understanding of field fluctuations. Since the Reuss, Voight, and Hashin-Shtrikman approximations all are homogenous within each phase, they necessarily have field fluctuations within each phase that are collinear with the applied field. However, e.g., as shown in Chapter 2 the field for a particulate composite is not well represented by a homogenous one within each phase and there also is a component of flux orthogonal to the applied field. In Figure 1.4 the field fluctuations for this new approximation is shown.

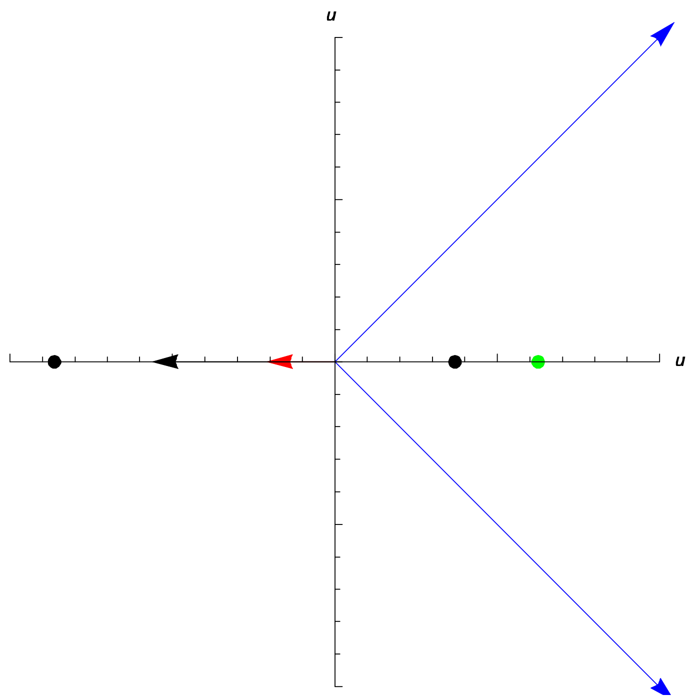

Figure 1.4: Points of concentration of fluctutations for the 3 vector approximation. The blue and red vectors are points of concentration in the first phase, and the black vector the point of concentration in the second phase. Black dots correspond to the Reuss solution.

CHAPTER 2 ELECTRIC FIELD FLUCTUATIONS IN CONDUCTORS WITH SPHERICAL INCLUSIONS

The chapter aims to find electric field and electric potential fluctuations in two-phase composites consisting of a matrix and randomly placed identically distributed spherical particles. This problem has a rich history. The study of the problem began about one hundred years ago by J. Holtsmark [5]. He was interested to assess if spectral line broadening observed in high pressure gases [6] can be explained by fluctuations of electric field, which are due to the presence of charges (ions), dipoles or quadrupoles.

To examine this proposition, the probability density of the magnitude of the electric field is to be computed for three cases: the random distribution of ions, dipoles and quadrupoles. This corresponds to determining probability distributions of sums of random variables,

$$\sum_a \frac{r_{ai}}{|r_a|^3} m, \quad \sum_a \frac{1}{|r_a|^3} \left(\delta_{ij} - \frac{r_{ai} r_{aj}}{|r_a|^2} \right) m_j, \tag{2.1}$$

$$\sum_a \frac{1}{|r_a|^5} \left(r_{aj}\delta_{ki} + r_{ai}\delta_{kj} - 3r_{ak}\delta_{ij} + 5\frac{r_{ai}r_{aj}r_{ak}}{|r_a|^2} \right) m_{jk},$$

where $r_a, a = 1, 2, ...,$ are points in three-dimensional space, which are distributed independently and homogeneously over space, r_{ai} coordinates of r_a, small Latin indices i, j, k run through values 1, 2, 3, $|r_a|$ the magnitude of r_a, δ_{ij} Kronecker's delta. The sums are the components of electric field at the origin caused by ions, dipoles and quadrupoles, respectively; m is the ion charge, m_i and m_{ij} reflect intensities of dipoles and quadrupoles. Holtsmark found that the probability density of the electric field $f(\nabla u)$ has the form

$$f(\nabla u) = \frac{1}{8\pi^3} \int_{R^3} e^{-i\vec{\rho}\cdot\nabla u - bn|\vec{\rho}|^H} d\vec{\rho}, \tag{2.2}$$

where variable of integration, vector $\overrightarrow{\rho}$, runs through three-dimensional space R^3, $\overrightarrow{\rho} \cdot \nabla u$ is the scalar product of $\overrightarrow{\rho}$ and ∇u, $|\overrightarrow{\rho}|$ length of $\overrightarrow{\rho}$, $d\overrightarrow{\rho} \equiv d\rho_1 d\rho_2 d\rho_3$, b a constant dependent on the charge magnitude, n is the number density of charges, and H is a constant which has the values 3/2, 1, and 3/4 for the ions, dipoles, and quadrupoles, respectively. Later on it was found that the Holtsmark distribution (2.2) is a member of a wide class of so-called stable distributions [13].

Writing integral (2.2) in spherical coordinates and integrating over angles one arrives at one-dimensional integral

$$f\left(\nabla u\right)=\frac{1}{2\pi^{2}\left|\nabla u\right|}\int_{0}^{\infty}\sin\left[\left|\nabla u\right|y\right]ye^{-bny^{H}}dy. \tag{2.3}$$

Formula (2.3) shows that probability density depends only on the magnitude of the electric field $|\nabla u|$. Denoting by $PD(X)$ the probability density of the magnitude of electric field, $X=\left|\nabla u\right|$, one finds from (2.3) after integration over all possible directions $\nabla u/\left|\nabla u\right|$ that

$$PD(X)=\frac{2}{\pi}X\int_{0}^{\infty}\sin\left[Xy\right]ye^{-bny^{H}}dy. \tag{2.4}$$

Formula (2.2) yields also the probability distributions of components of ∇u. They are all equal due to symmetry. For $x-$component of electric field u_x, when the $y-$ and $z-$ components are zero, probability density $PD\left(u_x\right)$ is:

$$PD\left(u_{x}\right)=\frac{1}{2\pi}\int_{-\infty}^{\infty}e^{-iyu_{x}-bn|y|^{H}}dy. \tag{2.5}$$

Probability density (2.5) is also a member of the family of stable distributions [13]. In the dipole case, $H=1$, formula (2.5) yields the symmetric Cauchy distribution,

$$PD\left(u_{x}\right)=\frac{1}{\pi}\frac{bn}{u_{x}^{2}+\left(bn\right)^{2}},$$

while for $H=2$ formula (2.5) becomes the symmetric normal distribution,

$$PD\left(u_{x}\right)=\frac{1}{\sqrt{4\pi bn}}\exp\left[\frac{-u_{x}^{2}}{4bn}\right].$$

Holtsmark applied the probability density found for quadrupoles to the broadening mechanism in hydrogen and nitrogen as well as to other cases. He found reasonable agreement to available experimental data on cesium spectral line broadening in hydrogen and nitrogen environments. Holtsmark's major focus was on the distribution of quadrupoles due to the limited experimental data for dipoles and ions. Further discussions on spectral line broadening mechanisms can be found in the review papers by Regemorter [7] and Stoneham [8].

Not surprisingly, due to the fundamental nature of the sums (2.1), Holtsmark distributions show up in other fields. In astrophysics, Chandrasekhar [9] considered the expected magnitude of gravitational force on point mass in a stellar system. The stars are approximated by identically and independently distributed point masses. The force on a single point mass is the sum of the first type (2.1). Chandrasekhar further showed in his review [10], that his original assumption of equal masses for all stars, which corresponds to Holtsmark's assumption of uniform charge magnitude, was not essential: if masses are distributed randomly with probability density $p(M)$, then probability density of the force is the same as in the case of uniform masses, with the value of the uniform mass M^*,

$$M^* = \left(\int_0^\infty M^{3/2} p(M)\, dM \right)^{2/3}.$$

The case of uniform masses is the special case of the mass distribution $p(M) = \delta(M - M^*)$.

In material science, Holtsmark distribution arises in studying of internal stresses caused by identically and independently distributed point defects [11]. The defects were modeled by spherical particles placed in a linear isotropic material. The displacement field u_i caused by these defects is determined then by Eshelby's solution [16] :

$$u_i = \pm K \frac{r_i}{|\vec{r}|^3},$$

where K is a parameter characterizing the intensity of the point defect and Poisson's ratio.

Total displacement is the sum of the first type (2.1) , total strains and stresses are the sums of the second type (2.1) . Zolotarev [11] found that the stresses follow the Cauchy distribution. Later Berlyand [12] studied this case in greater detail, and found the stress distribution to be a shifted Cauchy distribution for pressure and a symmetric Cauchy distribution for shear components. This shift was observed experimentally [18, 19] .

Problems of the type discussed are found in diverse fields including velocity distributions of vortices in two and three dimensions [20], temperature distributions in nuclear reactors, radiography, etc. (brief summary was given in [13]).

Holtsmark distributions have heavy tails. This means that moments of the electric field, i.e. the average values of $|\nabla u|^s$, are infinite for $s \geqslant 1$. For example, the variance of electric field (the average value of $|\nabla u|^2$) is infinite for Cauchy distribution. The origin of that is that singularities can come close to the point of observation and make the field at the point of observation infinite. The situation is different for conductors containing particles. Though the sums, probability of which is to be computed, are of the form (2.1), the singularities do not come closer to the point of observation than the particle radius. This changes the probability distributions considerably. Our goal is to find these distributions.

The major motivation for this work is that probability distributions of local fields (stresses, strains, currents, etc.) are needed in constructing the dynamic equations for microstructure evolution. To obtain probability distributions, some approximate methods are being developed (see, e.g. [14]) as well as in Chapters 3 and 5.2. One needs a proving ground to test the accuracy of approximations.

The problem under consideration can serve as such a ground, because the probability densities are computed analytically in the limit of small volume concentration. In fact, the computation follows Holtsmark's work with some minor deviations, which are due to finiteness of the particle radii. As in many other asymptotic problems, one can expect that the asymptotic results are applicable for not very small values of concentration. Therefore we provide results up to volume concentration 0.26.

Note that Cheng and Torquato [14] as well as Cule and Torquato [15] have found probability density of electric field fluctuations induced by an applied electric field for several two phase microstructures in the two-dimensional case.

Cheng and Torquato [14] computed the statistics of the collinear component of electric field by numerical approximation within a periodic unit cell for a large number of microstructure realizations. They considered composites with random non-overlapping discs, squares, and highly elongated ellipsoids within a matrix. For the case of discs, they found a probability distribution with two peaks, one arising from the matrix and the other from the inclusions.

Cule and Torquato [15] computed the probability distribution function of the electric field magnitude. They first found analytically the solution for a coated cylinder composite, similar to the coated sphere model of Hashin and Shtrikman [1]. This PDF was found to have a finite width with singular points at the limit values. One singular point arises from the homogenous field within the core of the cylinder, and the other from the maximum or minimum value. Additionally two singular points existed at intermediate values. These singular points were compared with the singularities found in calculations of the density of states of various applications of condensed matter physics. They also computed the cases of a grid of discs and as well a periodic composite comprised of a small unit cell of twenty random non-overlapping discs. The grid had similar features of singular points and finite width of the PDF, but with an additional intermediate singularity. For the periodic random composite case, the feature of a finite width appears to have been lost, but the intermediate singular points are subjectively retained. They state that in the limit of a random composite of the nature considered by Cheng and Torquato [14], these "local features" of finite width and singular points are smeared out and therefore are not expected for the case considered in this chapter.

Sections which follow are setting of the problem, analysis of the problem where it is shown that computation of probability densities is reduced to calculation of two functions $A(y)$ and

$B(y)$, find these functions numerically, describe the corresponding results for probability densities, and determine asymptotics of probability densities. A simple outcome of the calculations are values of effective coefficients and variances of electric field. Some technicalities are moved to Appendices.

Due to mathematical equivalence the results obtained for potential and electric field apply to electrical and heat conduction as well as dielectric polarization. Additionally, as the work of Holtsmark provided a motivation for the study of a diverse set of physical issues with point size defects, the approach given in this thesis can be applied further to other cases with finite sized defects. This includes both differing physics, such as the elastic case, as well as alternate microstructures such as the case of ellipsoidal particles in a matrix.

2.1 Setting of the Problem

Potential of electric field u is a solution of the equation

$$\frac{\partial}{\partial x_i} a\left(x\right) \frac{\partial u}{\partial x_i} = 0. \tag{2.6}$$

Here x is a point of three-dimensional unbounded space, x_i are Cartesian coordinates of x, summation over repeated indices is implied, $a\left(x\right)$ is conductivity of the composite which is assumed to be unbounded. The composite contains spherical particles in such a way that the number of particles N in any finite region V is proportional to the volume of V, $|V|$, and there is a limit

$$\lim_{|V|\to\infty} \frac{N}{|V|} = n.$$

Particles are placed in space randomly and independently. Radii of particles are also random but are such that the volume concentration c is finite

$$c = \lim_{|V|\to\infty} \sum_{a=1}^{N} \frac{4\pi}{3} \frac{R_a^3}{|V|}. \tag{2.7}$$

If radii of all particles are equal, then

$$c = \frac{4\pi R^3}{3} n. \tag{2.8}$$

Formula (2.7) assumes that particles do not overlap. When particles are placed statistically independent they do overlap, and one can introduce the volume concentration of particle phase c^*. Concentrations c and c^* are linked by the relation [4]

$$c = -\ln\left[1 - c^*\right]. \tag{2.9}$$

The conductivity $a(x)$ is a piecewise function: it is equal to a_1 in the matrix, and a_2 inside the particles. At particle boundaries both the potential $u(x)$ and the normal component of the flux $a(x)\,\partial u/\partial x_i$ are continuous

$$[u] = 0, \quad n_i \left[a(x)\frac{\partial u}{\partial x_i}\right] = 0. \tag{2.10}$$

Here $[\varphi]$ means the difference of the limit values of φ on the two sides of the particle boundary. The space average of the electric field is assumed to be a given constant. By space average of function $\varphi(x)$ we mean the limit

$$\langle \varphi \rangle = \lim_{|V| \to \infty} \frac{1}{|V|} \int\limits_V \varphi(x)\, dx.$$

Denoting by v_i the prescribed value of the average electric field, we have the condition

$$\left\langle \frac{\partial u}{\partial x_i} \right\rangle = v_i. \tag{2.11}$$

The boundary value problem in unbounded space $(2.6), (2.10), (2.11)$ has a unique solution for $\partial u/\partial x_i$.

Single particle solution. Electric potential in material without a particle is a linear

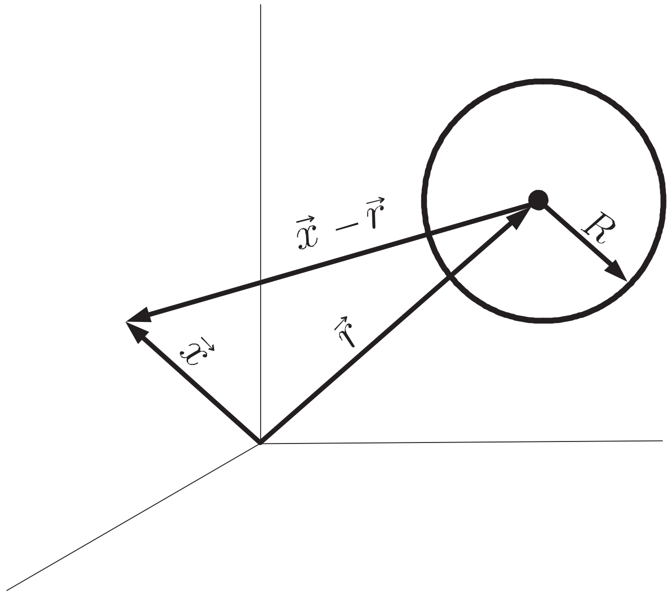

Figure 2.1: Notation for single inclusion problem

function $u = v_i x_i$. An arbitrary additive constant in potential is fixed by the condition $u(\{0,0,0\}) = 0$. One particle placed at the point r_i causes a disturbance of the electric potential

$$u = v_i x_i + \varkappa \begin{cases} (x_i - r_i)\, v_i & |x-r| \le R \\ \left(\frac{R}{|x-r|}\right)^3 (x_i - r_i)\, v_i & R \le |x-r| \end{cases}, \tag{2.12}$$

and the disturbance of the electric field

$$u_{,i} = v_i + \varkappa \begin{cases} v_i & |x-r| \le R \\ \left(\frac{R}{|x-r|}\right)^3 \left(\delta_{ij} - 3\frac{x_i - r_i}{|x-r|}\frac{x_j - r_j}{|x-r|}\right) v_j & R \le |x-r| \end{cases}. \tag{2.13}$$

Here comma in indices denotes spatial derivative, and $\varkappa$ is the constant

$$\varkappa \equiv \frac{a_1 - a_2}{2a_1 + a_2}.$$

The constant $\varkappa$ takes values in the range $[-1,\ 1/2]$. The limit cases $\varkappa = -1$ and $\varkappa = 1/2$ correspond to perfectly conductive particle ($a_2 = \infty$), and perfectly insulating particle ($a_2 = 0$), respectively. If $\varkappa = 0$, the media is homogeneous and no disturbance of the external field occurs.

Probability distributions. We seek probability densities of the random fields u and $\partial u/\partial x_i$ at a space point x.

In case of small c, electric field can be obtained by summation of electric fields generated by particles. Since the random fields are stationary, it is enough to consider probability distributions at one point. As such we take $x = \{0, 0, 0\}$. Due to linearity of the problem, u and $\partial u/\partial x_i$ are proportional to the magnitude of vector v_i. To simplify further relations we set $|\vec{v}|$ equal to unity. It is also convenient to make a shift for v_i and scale $u_{,i} - v_i$ by the constant $\varkappa$ and similarly scale u by $\varkappa$. Then results become independent of material characteristics and magnitude of the applied field. So, we will seek probability densities of random quantities

$$\eta_i = \frac{1}{\varkappa}\left(u_{,i} - v_i\right) \quad \text{and} \quad \xi = -\frac{1}{\varkappa}u. \tag{2.14}$$

It is convenient also to introduce a unit vector ζ_i and construct the probability density of the random number $\eta = \zeta_i \eta_i$; choosing different vectors ζ_i, we obtain the probability density of electric field in different directions.

After the probability densities of η_i and ξ are found the actual distribution of potential and electric field are obtained by scaling (see (2.46) and (2.60)).

By definition of probability density, probability density of $f_\xi(X)$ and $f_\eta(X)$ of random quantities ξ and η are

$$f_\xi(X) = M\delta\left(X - \sum_a \varphi_a(r_a, R^a)\right), \quad f_\eta(X) = M\delta\left(X - \sum_a \psi_a(r_a, R^a)\right), \tag{2.15}$$

where M stands for mathematical expectation, and $\varphi_a(r_a, R^a)$ and $\ _a(r_a, R^a)$ are the potential and electric field disturbance at the origin caused by the $a-$th particle

$$\varphi_a = \begin{cases} v_i r_i^a & |r^a| \leq R_a \\ \left(\frac{R_a}{|r^a|}\right)^3 v_i r_i^a & R_a \leq |r^a| \end{cases}, \tag{2.16}$$

$$\psi_a = \zeta_i \begin{cases} v_i & |r^a| \leq R_a \\ \left(\frac{R_a}{|r^a|}\right)^3 \left(\delta_{ij} - 3\frac{r_i^a}{|r^a|}\frac{r_j^a}{|r^a|}\right) v_j & R_a \leq |r^a| \end{cases}.$$

By mathematical expectation for an infinite system of particles, we mean in (2.15) the limits

$$f_\xi(X) = \lim_{N/|V| \to n, |V| \to \infty} M\delta\left(X - \sum_{a=1}^{N} \varphi_a\right), \tag{2.17}$$

$$f_\eta(X) = \lim_{N/|V| \to n, |V| \to \infty} M\delta\left(X - \sum_{a=1}^{N} {}_a\right),$$

where sums are taken over particles lying in the region V.

2.2 Computation of Probability Densities

Computation of the limits (2.17) follows to Lyapunov's idea for finding probability distributions of sums of independent random variables: $\delta-$function is to be replaced by its Fourier transform,

$$\delta(Y) \equiv \int_{-\infty}^{\infty} \frac{e^{iyY}}{2\pi} dy.$$

Then,

$$f_\xi(X) = \lim_{N/|V| \to n, |V| \to \infty} \int_{-\infty}^{\infty} e^{iyX} M \exp\left[-iy \sum_{a=1}^{N} \varphi_a\right] \frac{dy}{2\pi} \tag{2.18}$$

$$= \int_{-\infty}^{\infty} e^{iyX} \lim_{N/|V| \to n, |V| \to \infty} M \prod_{a=1}^{N} \exp\left[-iy\varphi_a\right] \frac{dy}{2\pi}.$$

Similarly,

$$f_\eta(X) = \int_{-\infty}^{\infty} e^{iyX} \lim_{N/|V| \to n, |V| \to \infty} M \prod_{a=1}^{N} \exp\left[-iy\psi_a\right] \frac{dy}{2\pi}. \tag{2.19}$$

Note that

$$M \exp\left[-iy\varphi_a\right] = \int\limits_V \exp\left[-iy\varphi_a\right] \frac{d^3r}{|V|}, \quad M \exp\left[-iy\psi_a\right] = \int\limits_V \exp\left[-iy\psi_a\right] \frac{d^3r}{|V|}.$$

Let all particles have the same radius, R. Then all functions φ_a and $\quad_a$ are the same: $\varphi_1 = \varphi_2 ... = \varphi$, and $\quad_1 = \psi_2 ... = \psi$. It is shown in Appendices A and B, that the limits in (2.18) and (2.19) can be found explicitly,

$$\lim_{N/|V| \to n, |V| \to \infty} \left(\int\limits_V \exp\left[-iy\varphi\right] \frac{d^3r}{|V|} \right)^N = \exp\left[-cA\left(yR\right)\right] \tag{2.20}$$

$$\lim_{N/|V| \to n, |V| \to \infty} \left(\int\limits_V \exp\left[-iy\psi\right] \frac{d^3r}{|V|} \right)^N = \exp\left[-cB\left(y\right)\right]. \tag{2.21}$$

Here $A\left(y\right)$ and $B\left(y\right)$ are the functions

$$\begin{aligned} A\left(y\right) &= 3\left(\frac{1}{y}\right)^3 \int\limits_0^y \left(1 - \frac{1}{m}\sin\left[m\right]\right) m^2 dm + \\ &\quad + 3\left|y\right|^{3/2} \int\limits_{1/\sqrt{|y|}}^{\infty} \left(1 - m^2 \sin\left[\frac{1}{m^2}\right]\right) m^2 dm \end{aligned} \tag{2.22}$$

$$\begin{aligned} B\left(y\right) &= \left(1 - e^{-iy\cos\alpha}\right) \\ &\quad + \frac{1}{4\pi} \int\limits_{0 \le \theta \le \pi} \int\limits_{0 \le \phi \le 2\pi} \int\limits_{0 \le s \le |y|} \left(|y| \frac{1 - \cos\left[s\left|C\right|\right]}{s^2} - iy\left(\frac{\sin\left[sC\right]}{s^2} - \frac{C}{s}\right)\right) ds \sin\theta d\theta d\phi, \end{aligned} \tag{2.23}$$

with

$$C = \cos\alpha - 3\cos\theta\left(\sin\alpha\cos\phi\sin\theta + \cos\alpha\cos\theta\right),$$

and α defined as the included angle between vectors ζ_i and external electric field v_i ($\cos\alpha = \zeta_i v_i$).

It is shown in Appendix B that $B\left(y\right)$ remains the same for any distribution of particle radii. The only change is the replacement of volume concentration c in (2.21) by effective volume concentration

$$\bar{c} = n\frac{4\pi}{3}\int_{0}^{\infty} R^3 p(R)\, dR \tag{2.24}$$

for random distribution of particle radii $f(R)$. The case of uniform radii (2.8) is the special case of the radius distribution $p(R) = \delta(R - R^*)$.

Functions $A(y)$ and $B(y)$ are easily found by numerical integration. After that, computation of $f_\xi(X)$ and $f_\eta(X)$ is reduced to another numerical integration,

$$f_\xi(X) = \int_{-\infty}^{\infty} e^{iyX - cA(yR)}\frac{dy}{2\pi}, \qquad f_\eta(X) = \int_{-\infty}^{\infty} e^{iyX - cB(y)}\frac{dy}{2\pi}. \tag{2.25}$$

We will compute three probability densities: probability density of the fields in matrix, in particles, and overall probability densities in composite. Probability density of a field in matrix is the conditional probability under the constraint that particles are not allowed to visit the point observation; this probability density is marked with index 1. The probability density in particles corresponds to placing the observation point inside particles; this probability density is marked with index 2. The overall probability density is the probability to observe a value of the field at any point of the composite.

Probability distributions within the particles can be determined after the probability distributions in the composite and matrix have been found. This can be done using an exact relation

$$f = (1 - c^*)\, f_1 + c^* f_2. \tag{2.26}$$

From (2.26) the probability density in the particles is

$$f_2 = f_1 + \frac{1}{c^*}(f - f_1). \tag{2.27}$$

First we present the results of computation for corresponding functions $A(y)$ and $B(y)$.

2.3 Function *A(y)*

To compute integrals in (2.22) numerically we find the values of integrals for $|y| \leqslant 20$ and for larger y we use asymptotics of integrals as $|y| \to \infty$.

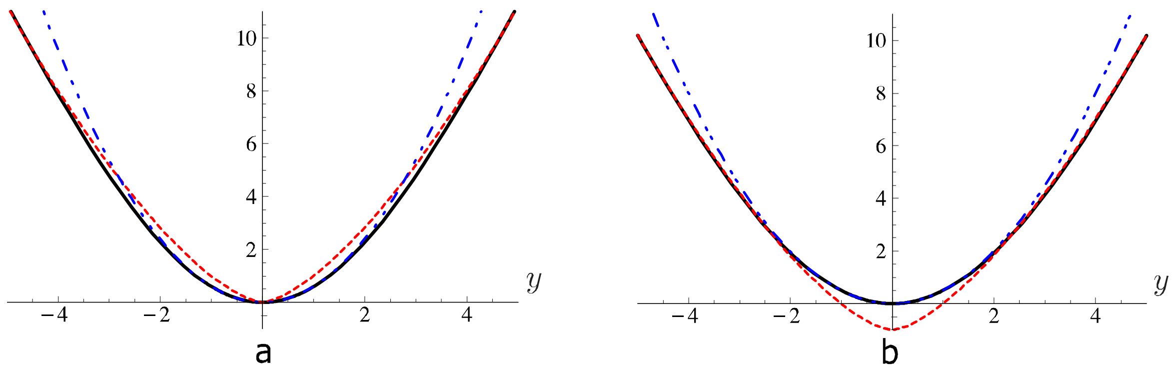

Figure 2.2: Function $A(y)$ (solid) in composite (a) along with its asymptotic approximations for small y ($|y|^{3/2}\ \ 2\sqrt{2\pi}/5$, point-dashed) and large y ($3y^2/5$, dashed), and $A_1(y)$ (solid) in matrix (b) along with its asymptotic approximations for small y ($|y|^{3/2}\ \ 2\sqrt{2\pi}/5 - 1$, point-dashed) and large y ($y^2/2$, dashed).

For large $|y|$ the first term in (2.22) tends to 1, while the integral in the second term is

$$\int\limits_{1/\sqrt{|y|}}^{\infty} \left(1 - m^2 \sin\left[\frac{1}{m^2}\right]\right) m^2 dm \tag{2.28}$$

$$= \int\limits_{0}^{\infty} \left(1 - m^2 \sin\left[\frac{1}{m^2}\right]\right) m^2 dm - \int\limits_{0}^{1/\sqrt{|y|}} \left(1 - m^2 \sin\left[\frac{1}{m^2}\right]\right) m^2 dm.$$

The first integral in (2.28) is equal to $2\sqrt{2\pi}/15$. In second integral since the upper limit goes to zero, the integrand should be evaluated for small m. We have in the leading approximation $\left(1 - m^2 \sin\left[1/m^2\right]\right) m^2 \asymp m^2$, yielding the value of the value of the second integral $|y|^{-3/2}/3$.

Thus, for large $|y|$

$$A\left(y\right) \asymp |y|^{3/2}\,\frac{2\sqrt{2\pi}}{5}. \tag{2.29}$$

For small $|y|$, expanding the integrand of the first integral in powers of m in (2.22) we get for this integral $y^5/30$. Then the first term in (2.22) is $y^2/10$. In the second integral, since the low limit goes to infinity, the integrand should be evaluated for large m. We have in the leading approximation $\left(1 - m^2 \sin\left[1/m^2\right]\right) m^2 \asymp 1/\left(6m^2\right)$. Thus the integral is $\sqrt{y}/6$ and the second term is $y^2/2$. Finally, for small y

$$A\left(y\right) \asymp \frac{3}{5} y^2. \tag{2.30}$$

Function $A(y)$ along with its asymptotics for large and small $|y|$ is shown in Fig. 2.2a.

Computing the probability distributions inside the matrix we constrain particle positions to be outside of the origin. Then in (2.17) a sphere at the origin with radius R is excluded from available positions of particle centers. This corresponds to dropping the first term in (2.22) and results in

$$A_1\left(y\right) = 3\left|y\right|^{3/2} \int\limits_{1/\sqrt{|y|}}^{\infty} \left(1 - m^2 \sin\left[\frac{1}{m^2}\right]\right) m^2 dm. \tag{2.31}$$

The asymptotics of $A_1\left(y\right)$ are found similarly to $A\left(y\right)$: for large $|y|$

$$A_1\left(y\right) \asymp \left|y\right|^{3/2} \frac{2\sqrt{2\pi}}{5} - 1 \tag{2.32}$$

and for small y

$$A_1\left(y\right) \asymp \frac{1}{2} y^2. \tag{2.33}$$

Function $A_1(y)$ along with its asymptotics for large and small $|y|$ is shown in Fig. 2.2b.

2.4 Function *B(y)*

In calculation of probability densities, we interpolate the numerical values of $B(y)$ for $|y| \leqslant 30$, and use the asymptotics of $B(y)$ for larger $|y|$.

We give the numerical illustrations for two values of α, $\alpha = 0$ $(C = 3\cos^2\theta - 1)$ and $\alpha = \pi/2$ $(C = 3\cos\theta\cos\phi\sin\theta)$. They correspond to consideration of the electric field components that are collinear and orthogonal to the direction of the external field. The corresponding functions and parameters, which arise, will be marked by symbols $\|$ and $\perp$, respectively. Function $A(y)$ does not depend on α, while function $B(y)$ does.

Consider first, function $B(y)$ for composite, when $\alpha = 0$. From (2.23)

$$\begin{aligned} B_{\parallel}(y) &= \left(1 - e^{-iy}\right) + \\ &\frac{1}{4\pi} \int\limits_{0\le\theta\le\pi} \int\limits_{0\le\phi\le 2\pi} \int\limits_{0\le s\le |y|} \left(|y| \frac{1-\cos[s|C|]}{s^2} - iy\left(\frac{\sin[sC]}{s^2} - \frac{C}{s}\right)\right) ds \sin\theta d\theta d\phi, \end{aligned} \tag{2.34}$$

with $C = 3\cos^2\theta - 1$.

Since

$$\int_0^{|y|} \frac{1-\cos[s|C|]}{s^2} ds \asymp \frac{\pi}{2}|C| - \frac{1}{|y|} \quad \text{as} \quad |y| \to \infty,$$

the integral in (2.34) for large $|y|$ behaves as

$$\begin{aligned} &\frac{1}{8}|y| \int\limits_{0\le\theta\le\pi} \int\limits_{0\le\phi\le 2\pi} |C| \sin\theta d\theta d\phi - 1 \\ &+ \frac{1}{4\pi} \int\limits_{0\le\theta\le\pi} \int\limits_{0\le\phi\le 2\pi} \int\limits_{0\le s\le |y|} \left(-iy\left(\frac{\sin[sC]}{s^2} - \frac{C}{s}\right)\right) ds \sin\theta d\theta d\phi. \end{aligned}$$

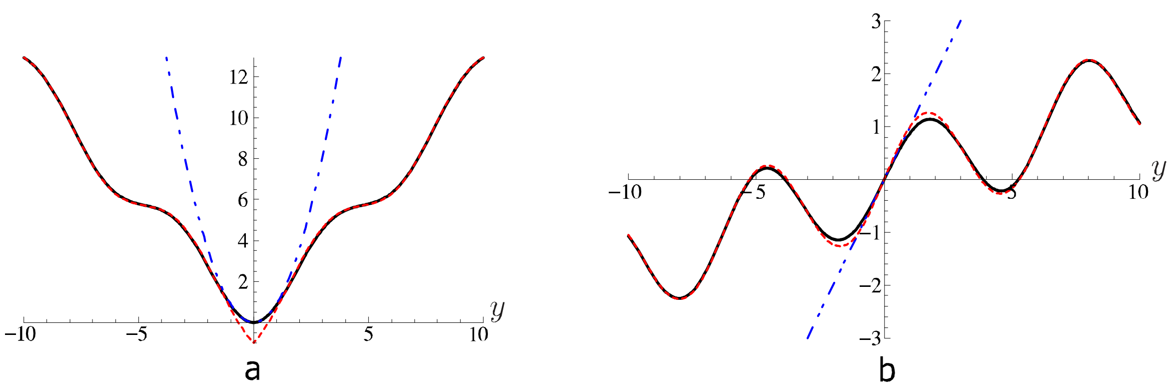

Figure 2.3: Real (a) and imaginary (b) parts of function $B_{\parallel}(y)$ (solid) along with its asymptotic approximations for small $y\,(iy + 9y^2/10$, point-dashed) and large y $\left(-\mathrm{Exp}(-iy) + iy/(2\pi) + 2\pi|y|/(3\sqrt{3}),\ \text{dashed}\right)$.

Therefore for large $|y|$

$$B_{\parallel}(y) \asymp -e^{-iy} + |y| \frac{2\pi}{3\sqrt{3}} + yi\frac{1}{2\pi}. \tag{2.35}$$

It was used here that

$$\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}|C|\sin\theta d\theta d\phi = \frac{16\pi}{3\sqrt{3}},\quad \int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}\int\limits_{0\le s\le\infty}\frac{\sin[sC]}{s^2}ds\sin\theta d\theta d\phi\approx-2,$$

$$\text{and}\quad \int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}C\sin\theta d\theta d\phi = 0.$$

The integral in (2.34) for small y can be approximated by the leading terms of the expansion of the integrand

$$\frac{1}{4\pi}\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}\int\limits_{0\le s\le|y|}\left(|y|\frac{1}{s^2}\frac{(s|C|)^2}{2}+iy\frac{1}{s^2}\frac{(sC)^3}{6}+...\right)ds\sin\theta d\theta d\phi,$$

and after integration over s the leading terms for $|y|\to0$ are

$$\frac{1}{8\pi}|y|^2\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}|C|^2\sin\theta d\theta d\phi+i\frac{1}{48\pi}y^3\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}C^3\sin\theta d\theta d\phi. \tag{2.36}$$

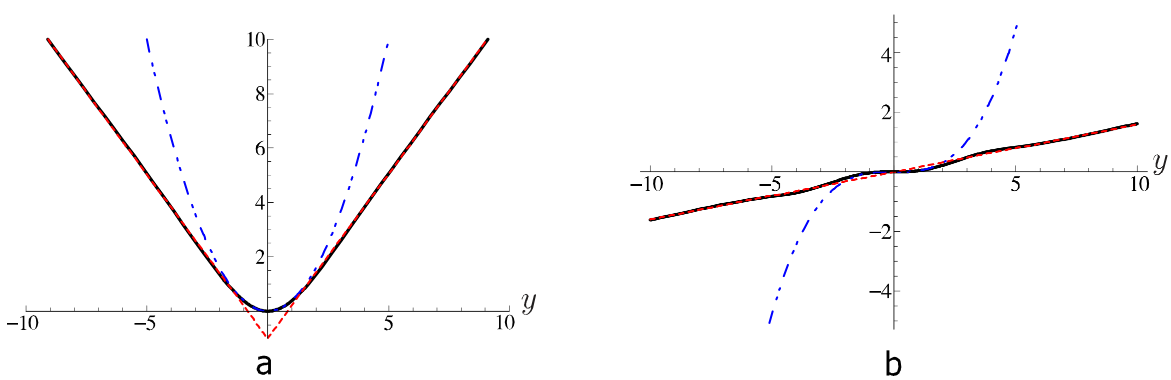

Figure 2.4: Real (a) and imaginary (b) parts of function $B^1_{\|}(y)$ (solid) along with its asymptotics for small y ($2y^2/5-4iy^3/105$, point-dashed) and large y ($iy/(2\pi)+2\pi|y|/(3\sqrt{3})-1$, dashed).

Therefore for small y

$$B_{\|}(y)\asymp iy+\frac{9}{10}y^2. \tag{2.37}$$

Here we used that

$$\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} C^2 \sin\theta d\theta d\phi = \frac{16\pi}{5}, \quad \int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} C^3 \sin\theta d\theta d\phi = \frac{64\pi}{35}.$$

Real and imaginary parts of $B_{\|}(y)$ for composite along with its asymptotics for large and small $|y|$ are shown in Fig. 2.3.

The corresponding function $B_{\|}^1(y)$ in the matrix is

$$B_{\|}^1(y) = \frac{1}{4\pi}\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi}\int\limits_{0\le s\le|y|}\left(|y|\frac{1-\cos\left[s\left|C\right|\right]}{s^2} - iy\left(\frac{\sin\left[sC\right]}{s^2} - \frac{C}{s}\right)\right) ds \sin\theta d\theta d\phi. \tag{2.38}$$

with $C = 3\cos^2\theta - 1$.

Large $|y|$ and small y asymptotics for $B_{\|}^1$ are found similarly to that of $B_{\|}$

$$B_{\|}^1(y) \asymp |y|\frac{2\pi}{3\sqrt{3}} - 1 + yi\frac{1}{2\pi} \quad \text{as} \quad |y| \to \infty. \tag{2.39}$$

$$B_{\|}^1(y) \asymp \frac{2}{5}y^2 + iy^3\frac{4}{105} \quad \text{as} \quad |y| \to 0. \tag{2.40}$$

Real and imaginary parts of $B_{\|}^1(y)$ for matrix along with its asymptotics for large and small $|y|$ are shown in Fig. 2.4.

Function $B(y)$ for $\alpha = \pi/2$ is a real valued function, and from (2.23)

$$B_{\perp}(y) = \frac{1}{4\pi}\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi}\int\limits_{0\le s\le|y|}\left(|y|\frac{1-\cos\left[s\left|C\right|\right]}{s^2} - iy\left(\frac{\sin\left[sC\right]}{s^2} - \frac{C}{s}\right)\right) ds \sin\theta d\theta d\phi \tag{2.41}$$

with $C = 3\cos\theta\cos\phi\sin\theta$.

For large $|y|$

$$B_{\perp}(y) \asymp |y| - 1. \tag{2.42}$$

For small y, $B_\perp$ behaves as the quadratic function

$$B_\perp(y) \asymp \frac{3}{10}y^2. \tag{2.43}$$

In (2.42) and (2.43) we used that

$$\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} |C|\sin\theta d\theta d\phi = 8, \quad \int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} \sin sC\sin\theta d\theta d\phi = 0,$$

$$\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} C^2\sin\theta d\theta = \frac{12\pi}{5}, \text{ and } \int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le 2\pi} C^3\sin\theta d\theta = 0.$$

$B_\perp(y)$ along with its asymptotics for large and small $|y|$ are shown in Fig. 2.5 .

It is easy to see that $B^1_\perp = B_\perp$.

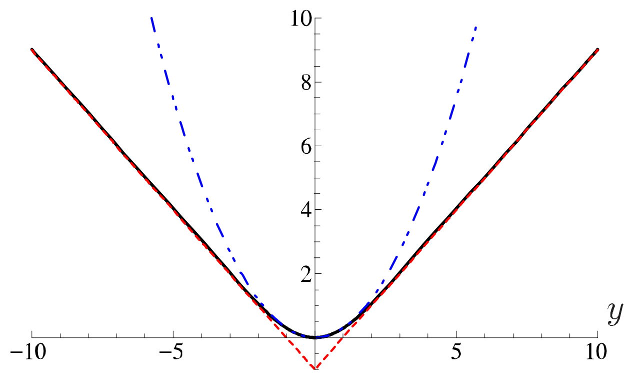

Figure 2.5: $B_\perp(y)$ (solid) along with its asymptotic approximations for small y $(3y^2/10$, point-dashed) and large y $(|y|-1,$ dashed$)$.

2.5 Probability Density of Electric Potential

In computation of probability density of electric potential f_ξ, it is convenient to make a change of variable of integration, $y \to t = yR$, and introduce function

$$\Phi(Y) = \int\limits_{-\infty}^{\infty} e^{itY - cA(t)}\frac{dt}{2\pi},$$

then

$$f_\xi(X) = \frac{1}{R}\Phi\left(\frac{X}{R}\right). \tag{2.44}$$

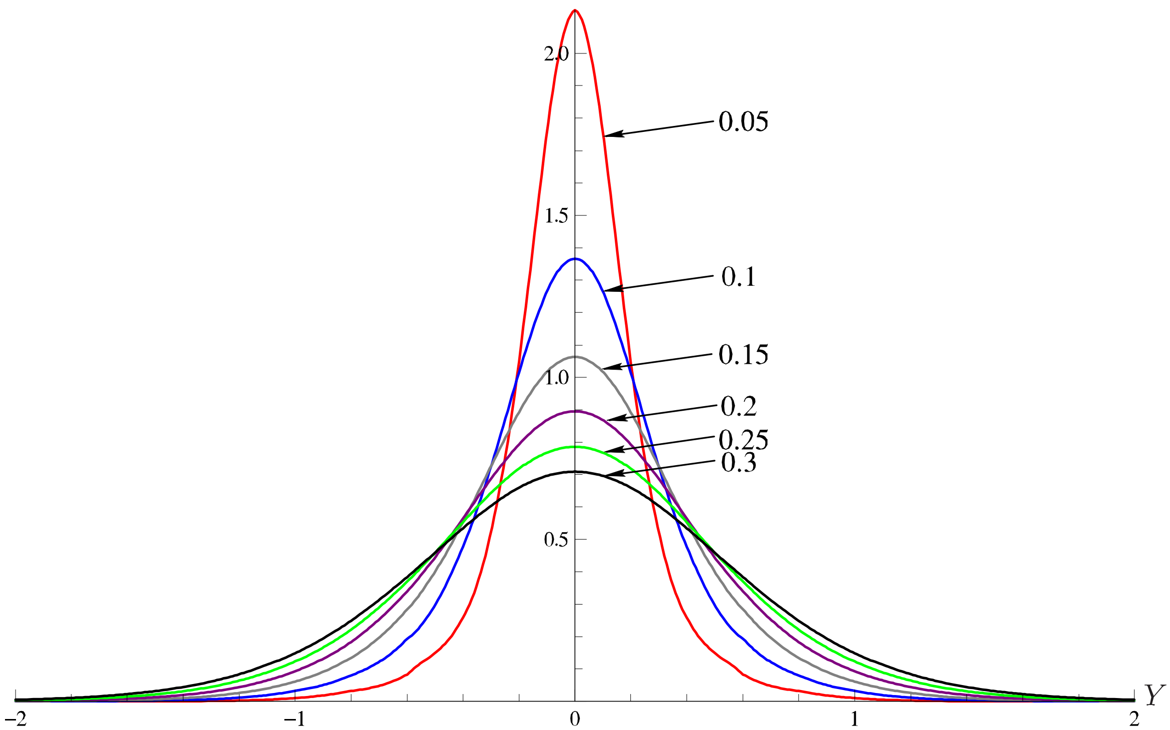

Figure 2.6: Probability density of electric potential in composite $\Phi(Y)$ for several values of concentration c.

The results of calculation for $\Phi(Y)$ are shown in Fig. 2.6, $\Phi_1(Y)$ in Fig. 2.7, and $\Phi_2(Y)$ in Fig. 2.8 for several values of concentration c up to 0.3. Note that the corresponding volume concentration $c^* \approx 0.26$, just below the percolation threshold found for uniform inclusion radii, $c^* \approx 0.29$ [21].

Because these distributions lack "heavy tails" the integrals for expected value

$$\mu = \int X f(X)\, dX,$$

variance

$$\sigma^2 = \int (X-\mu)^2 f(X)\, dX,$$

and excess kurtosis

$$\gamma^2 = \frac{1}{\sigma^4} \int (X-\mu)^4 f(X)\, dX - 3$$

obtained by numerical integration are fast converging.

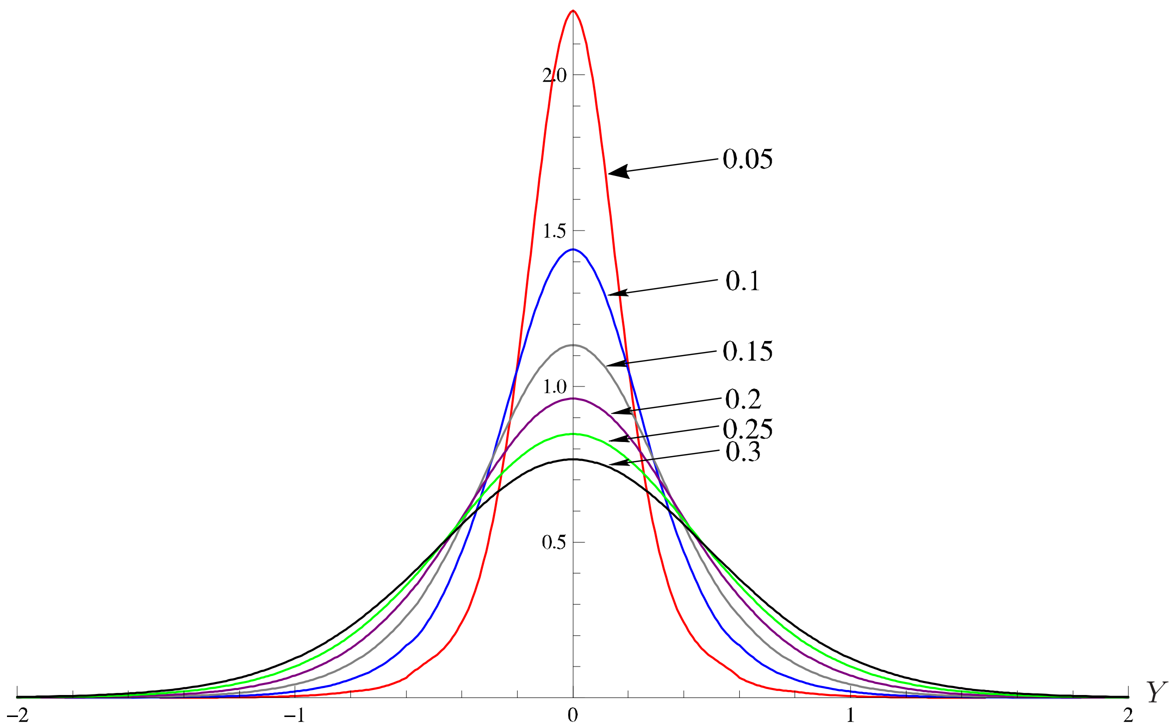

Figure 2.7: Probability density of electric potential in matrix $\Phi_1(Y)$ for several values of concentration c.

The functions Φ, Φ_1, and Φ_2 are even functions of Y and the corresponding expected values are zero. The values for variances and excess kurtosis are given in Table 2.1.

c	σ^2	γ^2	σ_1^2	γ_1^2	σ_2^2	γ_2^2
0.05	0.05887	2.9191	0.04881	2.4266	0.25508	-0.4108
0.10	0.11876	1.4566	0.09864	1.2289	0.31008	-0.1993
0.15	0.17942	0.9338	0.14943	0.7926	0.36477	-0.0997
0.20	0.23943	0.6707	0.19971	0.5805	0.41883	-0.0593
0.25	0.29881	0.5009	0.24966	0.4489	0.47190	-0.0528
0.30	0.35741	0.3735	0.29927	0.3535	0.52361	-0.0666

Table 2.1: Variances and excess kurtosis of electric potential (overall: σ^2 and γ^2, matrix: σ_1^2 and γ_1^2, particles: σ_2^2 and γ_2^2) for several values of concentration c.

Variances are fitted by the functions

$$\sigma^2 \approx \frac{6}{5}c, \quad \sigma_1^2 \approx c, \quad \text{and} \quad \sigma_2^2 \approx \frac{1}{5} + \frac{10}{9}c, \tag{2.45}$$

with errors not exceeding 3%.

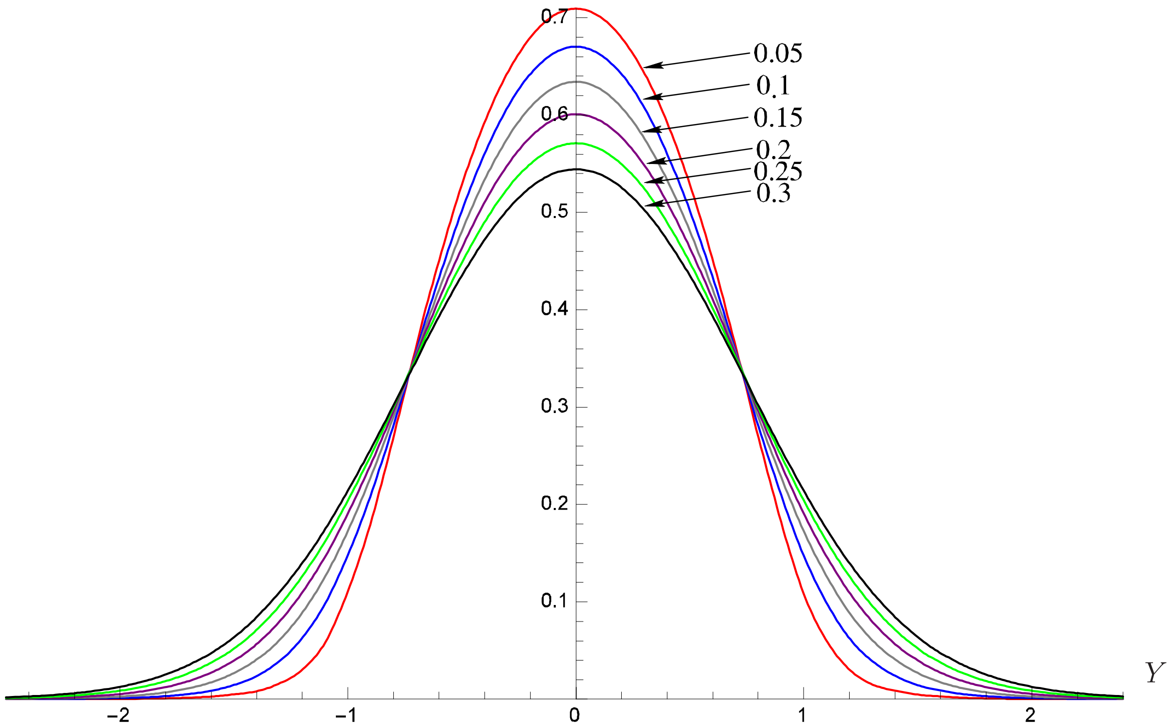

Figure 2.8: Probability density of electric potential in particles $\Phi_2(Y)$ for several values of concentration c.

Function (2.44) is the probability density of scaled potential $u/\varkappa$ for $|\vec{v}| = 1$. If $|\vec{v}| \neq 1$, then the probability density of potential u is

$$f(u) = \frac{1}{\varkappa |\vec{v}| R} \Phi\left(\frac{u}{\varkappa |\vec{v}| R}\right). \tag{2.46}$$

2.6 Probability Density of Electric Field Collinear with Applied Field

The probability densities of the electric field fluctuation collinear and orthogonal to the applied field are denoted by $f_{\parallel}(X)$ and $f_{\perp}(X)$, respectively.

The results of calculation for $f_{\parallel}(X)$ for several values of concentration c are shown in Fig. 2.9, $f_{\parallel}^{1}(X)$ in Fig. 2.10, and $f_{\parallel}^{2}(X)$ in Fig. 2.11, and the expected values, variance, and excess kurtosis are given in Table 2.2 .

Variances are well fitted by the functions

$$\left(\sigma_{\parallel}\right)^2 \approx \frac{9}{5}c - \frac{1}{2}c^2, \quad \left(\sigma_{\parallel}^{1}\right)^2 \approx \frac{4}{5}c - \frac{1}{3}c^2, \quad \text{and} \quad \left(\sigma_{\parallel}^{2}\right)^2 \approx \frac{4}{3}c - \frac{2}{5}c^2, \tag{2.47}$$

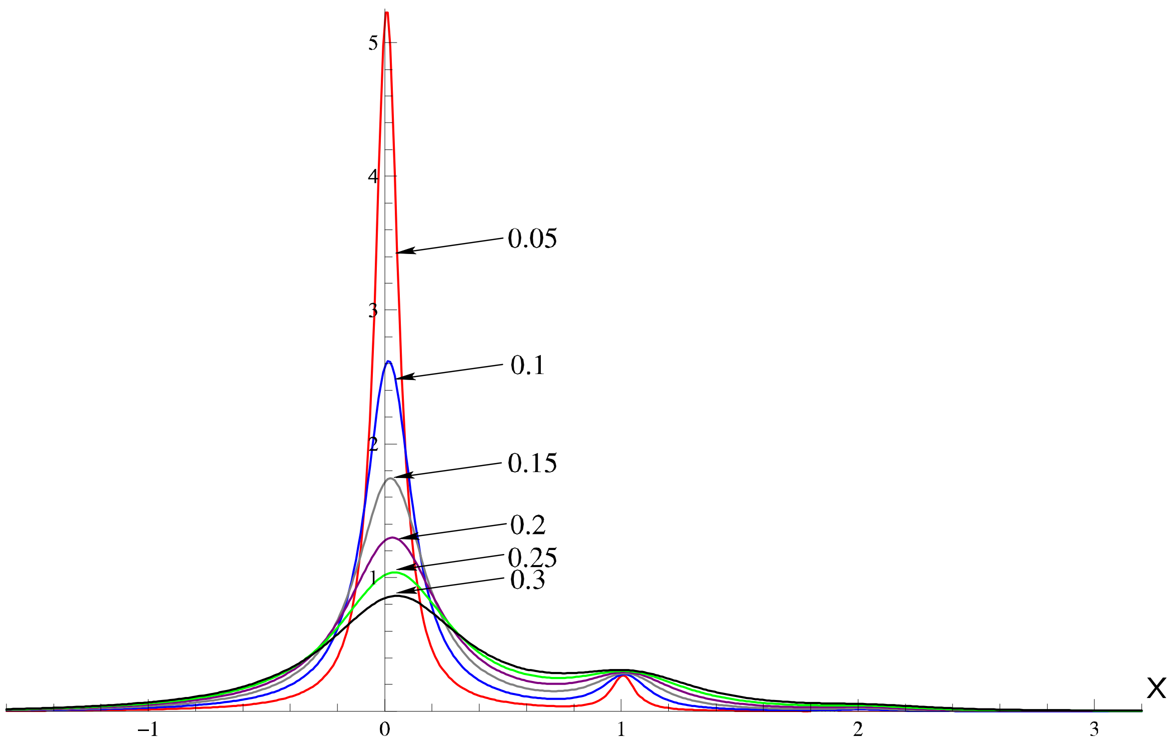

Figure 2.9: Probability density of collinear electric field component in composite $f_{\|}(X)$ for several values of concentration c.

c	$\mu_{\|}$	$(\sigma_{\|})^2$	$(\gamma_{\|})^2$	$\mu^1_{\|}$	$(\sigma^1_{\|})^2$	$(\gamma^1_{\|})^2$	$\mu^2_{\|}$	$(\sigma^2_{\|})^2$	$(\gamma^2_{\|})^2$
0.05	0.05140	0.08864	13.853	0.00010	0.03808	11.594	1.0315	0.06625	10.842
0.10	0.10226	0.17483	4.1344	0.00257	0.07508	5.3126	1.0495	0.12955	5.3826
0.15	0.15357	0.25937	2.5926	0.00469	0.11096	3.2257	1.0723	0.19224	3.3480
0.20	0.20466	0.34134	1.7703	0.00732	0.14573	2.1944	1.0953	0.25259	2.2863
0.25	0.25516	0.41999	1.2438	0.01043	0.17940	1.5860	1.1166	0.31017	1.6379
0.30	0.30479	0.49481	0.8709	0.01399	0.21200	1.1892	1.1360	0.36473	1.2034

Table 2.2: Average values, variances, and excess kurtosis of collinear electric field component (overall: $\mu_{\|}$, $(\sigma_{\|})^2$, and $(\gamma_{\|})^2$, matrix: $\mu^1_{\|}$, $(\sigma^1_{\|})^2$, and $(\gamma^1_{\|})^2$, particles: $\mu^2_{\|}$, $(\sigma^2_{\|})^2$, and $(\gamma^2_{\|})^2$) for several values of concentration c.

with error not exceeding 3%, while for expected values

$$\mu_{\|} \approx c, \quad \mu^1_{\|} \approx 0, \quad \text{and} \quad \mu^2_{\|} \approx 1 + \frac{1}{2}c. \tag{2.48}$$

Probability densities possess some interesting features. As seen in Fig. 2.9 the distribution of electric field in the composite has two strong peaks, one originating from the matrix

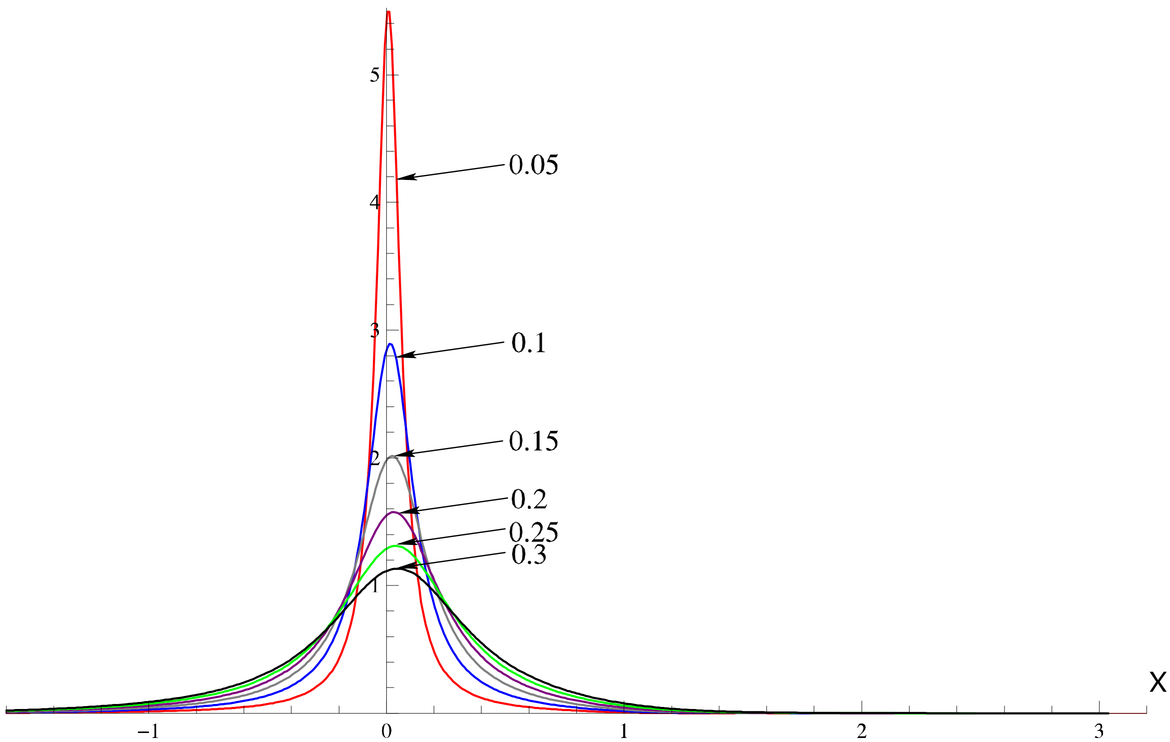

Figure 2.10: Probability density of collinear electric field component in matrix $f_{\|}^{1}(X)$ for several values of concentration c.

and the other from the particles. This is consistent with results for a similar microstructure in the two dimensional case [14]. The distribution in the particles (Fig. 2.11) has two distinct peaks. The major peak is associated with non-overlapping particles, and the peak at $X = 2$ results from overlapping of two particles. It is worth noting that these features hold also for random distribution of particle sizes since the size dependence enters only through volume concentration.

2.7 Probability Density of Electric Field Orthogonal to Applied Field

As was previously noted $B_{\perp}(y) = B_{\perp}^{1}(y)$, thus $f_{\perp}(X) = f_{\perp}^{1}(X) = f_{\perp}^{2}(X)$. This distribution is shown in Fig. 2.12 for several values of concentration c. The distribution $f_{\perp}$ is symmetric and the corresponding expected value is zero. The values for variance and excess kurtosis of the orthogonal component of electric field is given in Table 2.3. Again the distributions are noticeably non Gaussian.

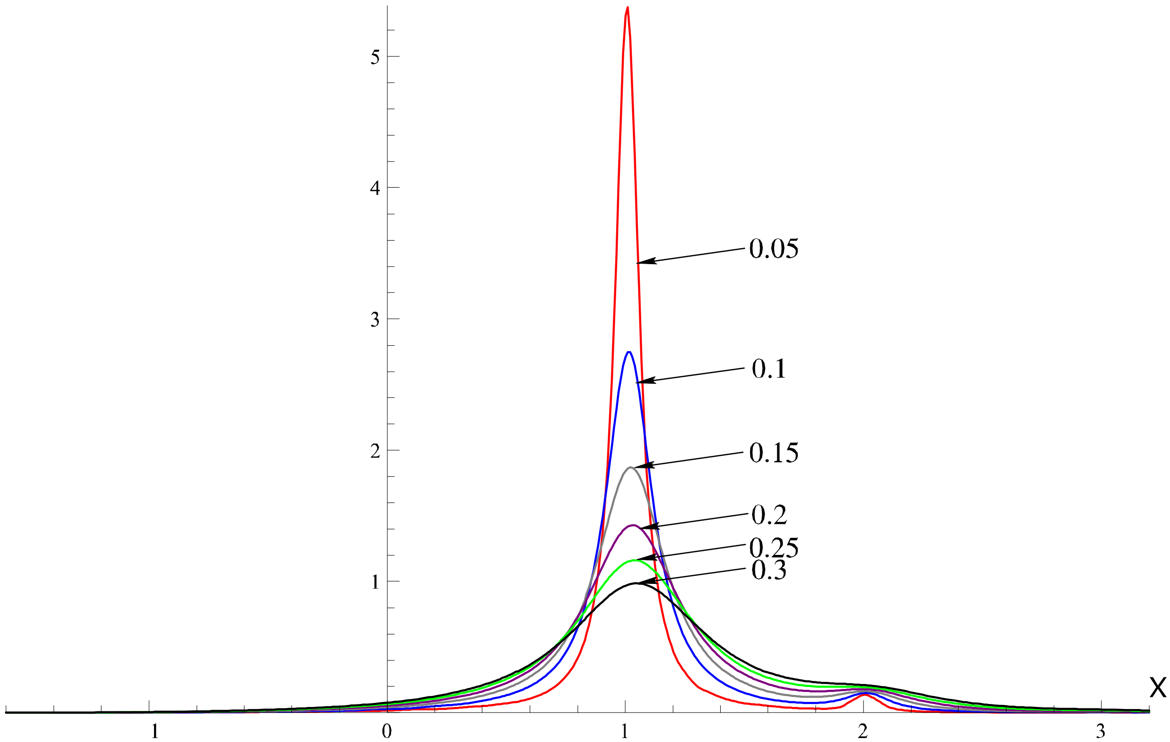

Figure 2.11: Probability density of collinear electric field component in particles ${f_{\|}}^{2}(X)$ for several values of concentration c.

Variance is fitted by the function

$$(\sigma_{\perp})^2 = \frac{3}{5}c - \frac{1}{8}c^2, \tag{2.49}$$

with error not exceeding 2%.

c	0.05	0.10	0.15	0.20	0.25	0.30
$(\sigma_{\perp})^2$	0.02984	0.05900	0.08737	0.11492	0.14156	0.16722
$(\gamma_{\perp})^2$	13.608	6.3946	3.9638	2.7325	1.9840	1.4791

Table 2.3: Variances and excess kurtosis of orthogonal electric field component (overall, matrix, and particles: $(\sigma_{\perp})^2$=$(\sigma^1_{\perp})^2$=$(\sigma^2_{\perp})^2$ and $(\gamma_{\perp})^2$=$(\gamma^1_{\perp})^2$=$(\gamma^2_{\perp})^2$) for several values of concentration c.

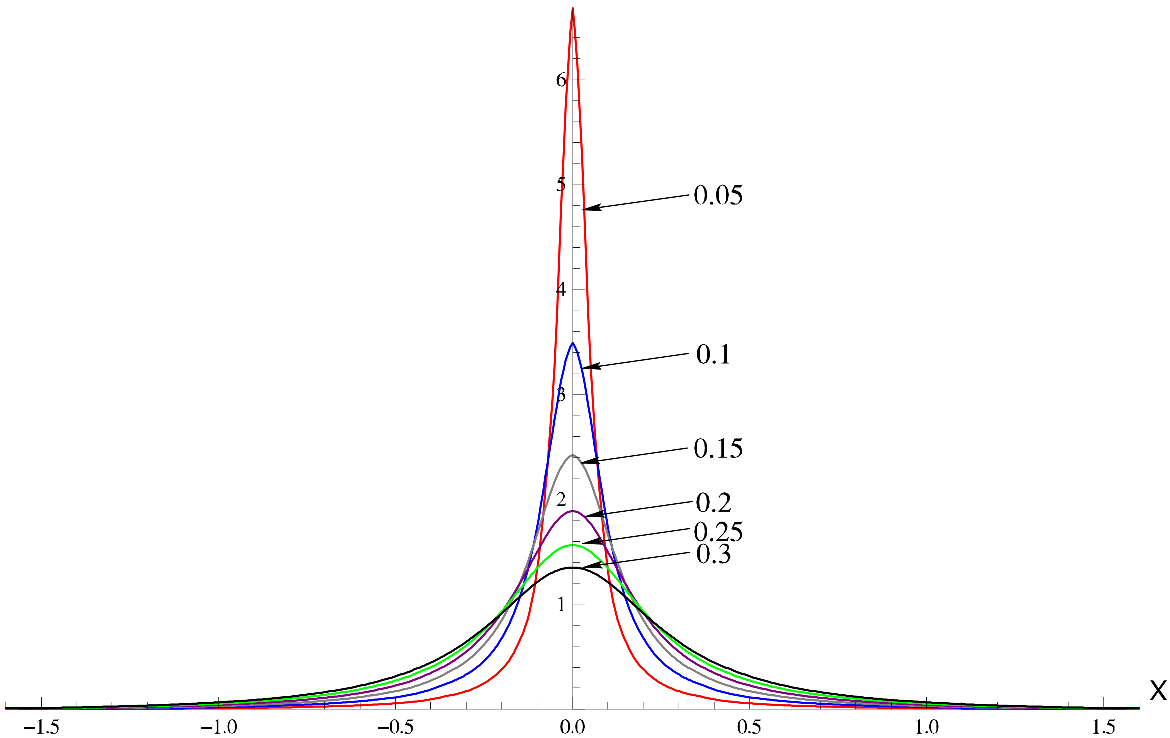

Figure 2.12: Probability density of orthogonal electric field component in composite, matrix, and particles $f_{\perp}(X) = f_{\perp}^{1}(X) = f_{\perp}^{2}(X)$ for several values of concentration c.

2.8 Asymptotics of Probability Densities

The distributions obtained decay like Gaussian distributions. Indeed, to find asymptotics of $\Phi(X)$ for large X let us make the substitution

$$y = w/X, \tag{2.50}$$

then

$$\Phi(X) = \int_{-\infty}^{\infty} \frac{e^{iyX-cA(y)}}{2\pi} dy = \frac{1}{X} \int_{-\infty}^{\infty} \frac{e^{iw-cA(w/X)}}{2\pi} dw. \tag{2.51}$$

Since $X \to \infty$, to evaluate integral in (2.51) we can use the asymptotics of $A(y)$ for small y. From (2.30) we have

$$\Phi(X) \asymp \frac{1}{X} \int_{-\infty}^{\infty} \frac{e^{iw-3c/5(w/X)^2}}{2\pi} dw.$$

Therefore, $\Phi(X)$ decays as Gaussian distribution with variance $6c/5$

$$\Phi(X) \asymp \sqrt{\frac{5}{12\pi c}} e^{-\frac{5}{12c}X^2} \quad \text{as} \quad |X| \to \infty, \tag{2.52}$$

Similarly

$$\Phi_1(X) \asymp \sqrt{\frac{1}{2\pi c}} e^{-\frac{1}{2c}X^2} \quad \text{as} \quad |X| \to \infty. \tag{2.53}$$

We have found in Section 6 that the variances for distributions $\Phi(X)$ and $\Phi_1(X)$ are $6c/5 - c^2/5$ and c, respectively. Large value asymptotics (2.52) and (2.53) suggest variances $6c/5$ and c. This would seem to be an indication that $\Phi(X)$ and $\Phi_1(X)$ are approximately Gaussian for small c. However this is not the case: the excess kurtosis (Table 2.1) was found to be non zero, and the distributions for small X are apparently non-Gaussian.

Similarly, the change of the variable of integration (2.50) in (2.25) shows that $f_{\|}(X)$, $f_{\|}^1(X)$, and $f_{\perp}(X)$ are determined by the asymptotics of $B_{\|}$, $B_{\|}^1$, and $B_{\perp}$ for small y (2.37), (2.40), and (2.43). We get the Gaussian decay

$$f_{\|}(X) \asymp \sqrt{\frac{5}{18\pi c}} e^{\frac{-5X^2}{18c}}, \quad f_{\|}^1(X) \asymp \sqrt{\frac{5}{8\pi c}} e^{-\frac{5X^2}{8c}}, \quad \text{and} \quad f_{\perp}(X) \asymp \sqrt{\frac{5}{6\pi c}} e^{-\frac{5X^2}{6c}} \quad \text{as} \quad |X| \to \infty. \tag{2.54}$$

These formulas correspond well to numerically found variances $(\sigma_{\|})^2 \approx 9c/5 - c^2/2$, $\left(\sigma_{\|}^1\right)^2 \approx 4c/5 - c^2/3$, and $(\sigma_{\perp})^2 \approx 3c/5 - c^2/8$. Again, $f_{\|}(X)$, $f_{\|}^1(X)$, and $f_{\perp}(X)$ appear to be nearly Gaussian for large X and non-Gaussian for finite X.

2.9 Effective Conductivity

The probability densities obtained allow us to find the effective conductivity (see, e.g. [3, 4])

$$a_{\text{eff}}^{ij} \bar{v}_i \bar{v}_j = \frac{1}{|V|} \int_V a^{ij}(r)\, \check{u}_{,i}(r)\, \check{u}_{,j}(r)\, d^3r, \tag{2.55}$$

where $\check{u}$ is the actual potential in the composite, $\bar{v}$ is the average value of electric field

$$\bar{v} = \langle \triangledown \check{u} \rangle = (1 - c^*) \langle \triangledown \check{u} \rangle_1 + c^* \langle \triangledown \check{u} \rangle_2, \tag{2.56}$$

and $\langle\ \ \rangle_1$ $\langle\ \ \rangle_2$ are space averages over phase 1 and phase 2, respectively.

For an isotropic composite with isotropic conductivity of each phase (2.55) simplifies to

$$a_{\text{eff}}\ \langle\triangledown\check{u}\rangle\cdot\langle\triangledown\check{u}\rangle = a_1(1-c^*)\int_{V_1}(\triangledown\check{u}\cdot\triangledown\check{u})\frac{d^3x}{|V_1|} + a_2c^*\int_{V_2}(\triangledown\check{u}\cdot\triangledown\check{u})\frac{d^3x}{|V_2|}. \tag{2.57}$$

We can compute a_{eff} using the statistics of the electric field obtained if we assume that the actual field can be approximated by the sums $\sum\varphi_a$ and $\sum\psi_a$ for small concentrations. Such calculation is instructive because it provides information on the validity of the approximation used: apparently, the approximation fails if the values of effective coefficient leaves the Hashin-Shtrikman bounds [17].

Let us show that the following relation holds:

$$\begin{aligned}\frac{a_{\text{eff}}}{a_1} &= (1-c^*)\frac{\varkappa^2\left(\left(\sigma_{\|}^1\right)^2+2\left(\sigma_{\perp}^1\right)^2\right)+1}{\left(1+\varkappa c^*\mu_{\|}^2\right)^2} \\ &\quad +\frac{a_2}{a_1}c^*\frac{\varkappa^2\left(\left(\sigma_{\|}^2\right)^2+\left(\mu_{\|}^2\right)^2+2\left(\sigma_{\perp}^2\right)^2\right)+2\varkappa\mu_{\|}^2+1}{\left(1+\varkappa c^*\mu_{\|}^2\right)^2}.\end{aligned} \tag{2.58}$$

Indeed, due to assumed ergodicity

$$\langle\triangledown u\cdot\triangledown u\rangle_1 = \int(\triangledown u\cdot\triangledown u)\,f^1d^3u \quad\text{and}\quad \langle\triangledown u\cdot\triangledown u\rangle_2 = \int(\triangledown u\cdot\triangledown u)\,f^2d^3u,$$

thus (2.57) is equivalent to

$$\begin{aligned}a_{\text{eff}}\ \langle\triangledown u\rangle\cdot\langle\triangledown u\rangle &= a_1(1-c^*)\left(\int_{-\infty}^{\infty}(u_1)^2f^1(u_1)\,du_1+2\int_{-\infty}^{\infty}(u_2)^2f^1(u_2)\,du_2\right) \\ &\quad +a_2c^*\left(\int_{-\infty}^{\infty}(u_1)^2f^2(u_1)\,du_1+2\int_{-\infty}^{\infty}(u_2)^2f^2(u_2)\,du_2\right).\end{aligned} \tag{2.59}$$

Using the dimensional results for probability density of electric field

$$f\left(u_1\right)=\frac{1}{\varkappa\left|\vec{v}\right|}f_{\parallel}\left(\frac{u_1-\left|\vec{v}\right|}{\varkappa\left|\vec{v}\right|}\right)\quad\text{and}\quad f\left(u_2\right)=\frac{1}{\varkappa\left|\vec{v}\right|}f_{\perp}\left(\frac{u_2}{\varkappa\left|\vec{v}\right|}\right),\tag{2.60}$$

we have

$$\begin{aligned}\frac{\left\langle u_1\right\rangle_1}{\left|\vec{v}\right|} &= \int\limits_{-\infty}^{\infty}\frac{u_1}{\left|\vec{v}\right|}f^1\left(u_1\right)du_1=\int\limits_{-\infty}^{\infty}\frac{u_1}{\left|\vec{v}\right|}\frac{1}{\varkappa\left|\vec{v}\right|}f_{\parallel}^1\left(\frac{u_1-\left|\vec{v}\right|}{\varkappa\left|\vec{v}\right|}\right)du_1=\varkappa\int\limits_{-\infty}^{\infty}\Psi f_{\parallel}^1\left(\Psi-\frac{1}{\varkappa}\right)d\Psi\\ &= \varkappa\int\limits_{-\infty}^{\infty}\left(\Psi+\frac{1}{\varkappa}\right)f_{\parallel}^1\left(\Psi\right)d\Psi=\left(\varkappa\mu_{\parallel}^1+1\right),\end{aligned}$$

and similarly

$$\frac{\left\langle u_2\right\rangle_1}{\left|\vec{v}\right|}=\varkappa\mu_{\perp}^1,\quad\frac{\left\langle u_1\right\rangle_2}{\left|\vec{v}\right|}=\left(\varkappa\mu_{\parallel}^2+1\right),\quad\text{and}\quad\frac{\left\langle u_2\right\rangle_2}{\left|\vec{v}\right|}=\varkappa\mu_{\perp}^2.$$

Since

$$\left\langle\bigtriangledown u\right\rangle\cdot\left\langle\bigtriangledown u\right\rangle=\left\langle u_1\right\rangle^2+2\left\langle u_2\right\rangle^2,$$

we obtain

$$\frac{\left\langle\bigtriangledown u\right\rangle\cdot\left\langle\bigtriangledown u\right\rangle}{\left|\vec{v}\right|^2}=\left(1+\varkappa\mu_{\parallel}^1+\varkappa c^*\left(\mu_{\parallel}^2-\mu_{\parallel}^1\right)\right)^2+2\varkappa^2\left(\mu_{\perp}^1+c^*\left(\mu_{\perp}^2-\mu_{\perp}^1\right)\right)^2.$$

Similarly

$$\begin{aligned}\frac{\left\langle\left(u_1\right)^2\right\rangle_1}{\left|\vec{v}\right|^2} &= \int\limits_{-\infty}^{\infty}\left(\frac{u_1}{\left|\vec{v}\right|}\right)^2f^1\left(u_1\right)du_1=\int\limits_{-\infty}^{\infty}\left(\frac{u_1}{\left|\vec{v}\right|}\right)^2\frac{1}{\varkappa\left|\vec{v}\right|}f_{\parallel}^1\left(\frac{u_1-\left|\vec{v}\right|}{\varkappa\left|\vec{v}\right|}\right)du_1\\ &= \varkappa^2\int\limits_{-\infty}^{\infty}\left(\Psi+\frac{1}{\varkappa}\right)^2f_{\parallel}^1\left(\Psi\right)d\Psi\\ &= \varkappa^2\int\limits_{-\infty}^{\infty}\Psi^2f_{\parallel}^1\left(\Psi\right)d\Psi+2\varkappa\int\limits_{-\infty}^{\infty}\Psi f_{\parallel}^1\left(\Psi\right)d\Psi+\int\limits_{-\infty}^{\infty}f_{\parallel}^1\left(\Psi\right)d\Psi\\ &= \varkappa^2\left(\left(\sigma_{\parallel}^1\right)^2+\left(\mu_{\parallel}^1\right)^2\right)+2\varkappa\mu_{\parallel}^1+1,\end{aligned}$$

$$\frac{\left\langle (u_2)^2 \right\rangle_1}{|\vec{v}|^2} = \int_{-\infty}^{\infty} \left(\frac{u_2}{|\vec{v}|}\right)^2 f^1(u_2)\, du_2 = \int_{-\infty}^{\infty} \left(\frac{u_2}{|\vec{v}|}\right)^2 \frac{1}{\varkappa |\vec{v}|} f_\perp^1 \left(\frac{u_2}{\varkappa |\vec{v}|}\right) du_2 = \varkappa^2 \int_{-\infty}^{\infty} \Psi^2 f_\perp^1(\Psi)\, d\Psi$$
$$= \varkappa^2 \left(\left(\sigma_\perp^1\right)^2 + \left(\mu_\perp^1\right)^2 \right),$$

$$\frac{\left\langle (u_1)^2 \right\rangle_2}{|\vec{v}|^2} = \varkappa^2 \left(\left(\sigma_\parallel^2\right)^2 + \left(\mu_\parallel^2\right)^2 \right) + 2\varkappa \mu_\parallel^2 + 1, \quad \text{and} \quad \frac{\left\langle (u_2)^2 \right\rangle_2}{|\vec{v}|^2} = \varkappa^2 \left(\left(\sigma_\perp^2\right)^2 + \left(\mu_\perp^2\right)^2 \right).$$

The term $|\vec{v}|^2$ is a common factor in (2.59) and can be dropped. Since $\mu_\perp^1 = \mu_\perp^2 = 0$ and $\mu_\parallel^1 \approx 0$, we have arrived at (2.58).

Comparison of (2.58) with the Hashin-Shtrikman bounds [17]

$$1 + \frac{c^*}{a_1} \left(\frac{1}{a_2 - a_1} + \frac{1 - c^*}{3a_1} \right)^{-1} \quad \text{and} \quad \frac{a_2}{a_1} + \frac{1 - c^*}{a_1} \left(\frac{1}{a_1 - a_2} + \frac{c^*}{3a_2} \right)^{-1} \tag{2.61}$$

is shown in Figs 2.13 and 2.14.

For insulative particles $\varkappa > 0$, (2.58) follows the upper Hashin-Shtrikman bound; the plots for particular values $a_2/a_1 = 2/5$ ($\varkappa = 1/4$) and $a_2/a_1 = 10^{-3}$ ($\varkappa \approx 1/2$) are shown in Figs. 2.13a and 2.13b, respectively.

For conducting particles $\varkappa < 0$, (2.58) coincides with the lower Hashin-Shtrikman bound for small c^* and remains within the bounds for sufficiently small c^*. In examples shown in Fig. 2.14a, 2.14b, and 2.14c $(a_2/a_1 = 10^4 \ (\varkappa \approx -1)$, $a_2/a_1 = 10 \ (\varkappa = -3/4)$, and $a_2/a_1 = 2 \ (\varkappa = -1/4))$ bounds are not violated for $c^* < .17$.

For small concentration, the leading term approximation of (2.58) in c^*, $a_{\text{eff}}/a_1 \approx (1 - 3c^*\varkappa)$, is consistent with previous results (see e.g. [17]). This result supports the validity of the approximation for small concentration and explains why (2.58) coincides with the upper Hashin-Shtrikman bound for $\varkappa > 0$, and the lower Hashin-Shtrikman bound for $\varkappa < 0$.

Let us compare our results for variances within the composite with known bounds [22, 23] : the second central moment of the electric field magnitude normalized to the average field magnitude squared can be found from the statistics in the composite

$$\frac{\langle (\nabla u - \langle \nabla u \rangle) \cdot (\nabla u - \langle \nabla u \rangle) \rangle}{\langle \nabla u \rangle \cdot \langle \nabla u \rangle} = \varkappa^2 \frac{\left(\sigma_{\parallel}\right)^2 + 2\left(\sigma_{\perp}\right)^2}{\left(1 + \varkappa c^* \mu_{\parallel}^2\right)^2} \tag{2.62}$$

or from statistics within each phase since

$$\begin{aligned} \frac{\langle (\nabla u - \langle \nabla u \rangle) \cdot (\nabla u - \langle \nabla u \rangle) \rangle_1}{\langle \nabla u \rangle \cdot \langle \nabla u \rangle} &= \varkappa^2 \frac{\left(\sigma_{\parallel}^1\right)^2 + \left(\mu_{\parallel} - \mu_{\parallel}^1\right)^2 + 2\left(\sigma_{\perp}^1\right)^2}{\left(1 + \varkappa c^* \mu_{\parallel}^2\right)^2} \\ \frac{\langle (\nabla u - \langle \nabla u \rangle) \cdot (\nabla u - \langle \nabla u \rangle) \rangle_2}{\langle \nabla u \rangle \cdot \langle \nabla u \rangle} &= \varkappa^2 \frac{\left(\sigma_{\parallel}^2\right)^2 + \left(\mu_{\parallel} - \mu_{\parallel}^2\right)^2 + 2\left(\sigma_{\perp}^2\right)^2}{\left(1 + \varkappa c^* \mu_{\parallel}^2\right)^2}. \end{aligned} \tag{2.63}$$

Note that there is less than 1.1% difference between the results computed from the composite and the results within each phase over the range of concentrations considered.

We know that for the degenerated homogenous composite case (i.e. $a_1 = a_2$ or $a_1 \neq a_2$ and $c^* = 0$) variances must be zero since the PDF of electric field is delta distributed; indeed this solution holds in this case. Additionally, a valid solution should not violate the Beran bounds [22]

$$\frac{\langle a \rangle - a_{\text{eff}}}{a_1} \quad \text{and} \quad \frac{\langle a \rangle - a_{\text{eff}}}{a_2}, \tag{2.64}$$

nor the lower Lipton bound [23]

$$\frac{3\left(\langle a \rangle - a_{\text{HS}}^+\right)^2}{\left(a_1 - a_2\right)^2 \left(1 - c^*\right) c^*}. \tag{2.65}$$

In (2.65), a_{HS}^+ denotes the greater of the two Hashin-Shtrikman bounds on effective properties (2.61).

These bounds (2.64, 2.65) are compared with our results (2.62) in Fig. 2.15a, 2.15b, and 2.15c for $a_2/a_1 = 2/5$ ($\varkappa = 1/4$), $a_2/a_1 = 2$ ($\varkappa = -1/4$), and $a_2/a_1 = 10$ ($\varkappa = -3/4$) using relation (2.58) for a_{eff}. Fig. 2.15 shows that for insulating particles $\varkappa > 0$ (Fig. 2.15a) and for conducting particles in the high contrast case (Fig. 2.15c) these bounds do not appear to further constrain the validity the relations obtained. However, for conducting particles $\varkappa < 0$, in the low contrast case (Fig.2.15b) the approximation fails against the (2.64) upper bound for values of concentration $c^* > .10$.

These higher than expected variances appear to arise from the uncorrected particle to particle interactions. With the approach of this thesis if two individual particles perfectly overlap, which is simply a single particle, the field disturbances are double the actual. This was noted to be apparent from the plot of $f_{\|}^{2}(X)$ in Fig. 2.11, and would increase the values of $\left(\sigma_{\|}^{2}\right)^{2}$ and $\mu_{\|}^{2}$. This issue also applies to $f_{\|}^{1}(X)$ and $f_{\perp}(X)$ thereby increasing the values of $\left(\sigma_{\|}^{1}\right)^{2}$ and $(\sigma_{\perp})^{2}$.

Next, let us consider the variances within each phase

$$\frac{\langle(\nabla u - \langle\nabla u\rangle_1)\cdot(\nabla u - \langle\nabla u\rangle_1)\rangle_1}{\langle\nabla u\rangle_1\cdot\langle\nabla u\rangle_1} = \varkappa^2\left(\left(\sigma_{\|}^{1}\right)^2 + 2\left(\sigma_{\perp}^{1}\right)^2\right) \tag{2.66}$$

$$\frac{\langle(\nabla u - \langle\nabla u\rangle_2)\cdot(\nabla u - \langle\nabla u\rangle_2)\rangle_2}{\langle\nabla u\rangle_2\cdot\langle\nabla u\rangle_2} = \varkappa^2\frac{\left(\sigma_{\|}^{2}\right)^2 + 2\left(\sigma_{\perp}^{2}\right)^2}{\left(\varkappa\mu_{\|}^{2}+1\right)^2} \tag{2.67}$$

which should not violate the Beran upper bounds [24]

$$\frac{\left\langle\left(\nabla u - \langle\nabla u\rangle_+\right)\cdot\left(\nabla u - \langle\nabla u\rangle_+\right)\right\rangle_+}{\langle\nabla u\rangle_+\cdot\langle\nabla u\rangle_+} \le \frac{a_-}{c_-}\frac{1}{\left(a_{\text{eff}}-a_-\right)^2}\left(\langle a\rangle - a_{\text{eff}}\right)\left(a_{\text{eff}}\left\langle\frac{1}{a}\right\rangle - 1\right) \tag{2.68}$$

$$\frac{\left\langle\left(\nabla u - \langle\nabla u\rangle_-\right)\cdot\left(\nabla u - \langle\nabla u\rangle_-\right)\right\rangle_-}{\langle\nabla u\rangle_-\cdot\langle\nabla u\rangle_-} \le \frac{a_+}{c_+}\frac{1}{\left(a_{\text{eff}}-a_+\right)^2}\left(\langle a\rangle - a_{\text{eff}}\right)\left(a_{\text{eff}}\left\langle\frac{1}{a}\right\rangle - 1\right).$$

Here a_+ denotes the greater of a_1 and a_2 and c_+ the concentration of this phase, and similarly for a_-.

These bounds (2.68) are compared with our results (2.66, 2.67) in Figs. 2.16 and 2.17 for $a_2/a_1 = 2/5$ ($\varkappa = 1/4$), $a_2/a_1 = 2$ ($\varkappa = -1/4$), and $a_2/a_1 = 10$ ($\varkappa = -3/4$) using relation (2.58) for a_{eff}.

First consider the variances within the matrix. In the insulative particle case (Fig. 2.16a) our result (2.66) nearly coincides with the upper bound (2.68) and in the high contrast conducting particle case, this bound does not provide further constraint (Fig. 2.16c). However, for the low contrast conducting particle case (Fig. 2.16b) the approximation fails against the (2.68) upper bound for small values of concentration $c^* > .1$.

For the variances within the particles (2.67) the insulative particle (Fig. 2.17a) and low contrast conducting particle (Fig. 2.17b) cases the bounds (2.68) do not provide further constraint. However, for the high contrast conductive particle case (Fig. 2.17c) variances in the particles fail against the bounds (2.68) for very small values of concentration $c^* > .05$. The variance in the particles (2.67) enters in the effective coefficient (2.58) and variances in the composite (2.62) with the factor

$$\frac{\langle \nabla u \rangle_2 \cdot \langle \nabla u \rangle_2}{\langle \nabla u \rangle \cdot \langle \nabla u \rangle} = \frac{\left(\varkappa \mu_{\|}^2 + 1\right)^2}{\left(\varkappa c^* \mu_{\|}^2 + 1\right)^2};$$

for the high contrast conductive particle case this factor takes very small values, and explains why the violation of this bound was not apparent from the effective coefficient (Fig. 2.14b) nor variance in the composite (Fig. 2.13c).

Our results were compared with bounds on effective coefficient as well as variances in the composite, matrix, and particles. These bounds constrain the validity of this approximation in the case of conductive particles. These results do not violate bounds in the low contrast case $a_2/a_1 = 2$ for $c^* < .10$ and for the high contrast case $a_2/a_1 = 10$ concentrations $c^* < .05$. It is expected if the particle-particle interactions were corrected, the validity of these results could be expanded.

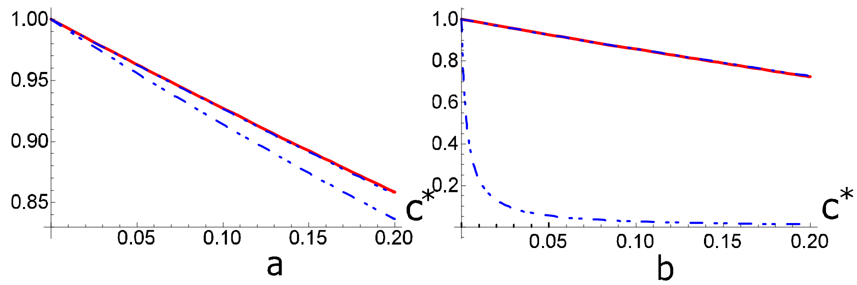

Figure 2.13: Effective coefficient a_{eff}/a_1 for composite with insulative particles (solid) and Hashin-Shtrikman bounds (point-dashed) ($\varkappa = 1/4$ (a) and $\varkappa \approx 1/2$ (b)).

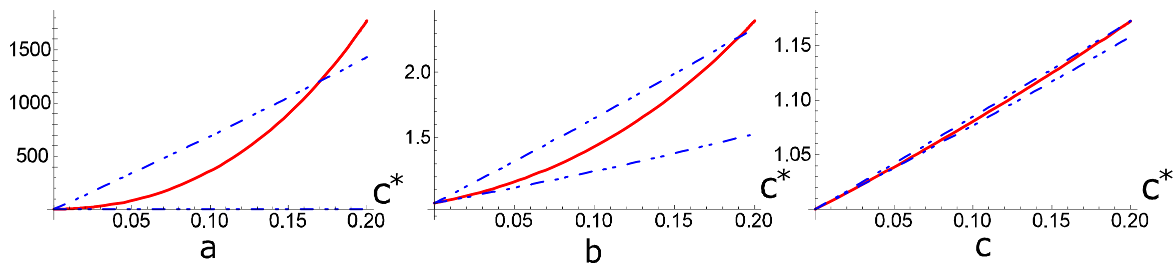

Figure 2.14: Effective coefficient a_{eff}/a_1 for composite with conductive particles (solid) and Hashin-Shtrikman bounds (point-dashed) (($\varkappa \approx -1$ (a), $\varkappa = -3/4$ (b), and $\varkappa = -1/4$ (c)).

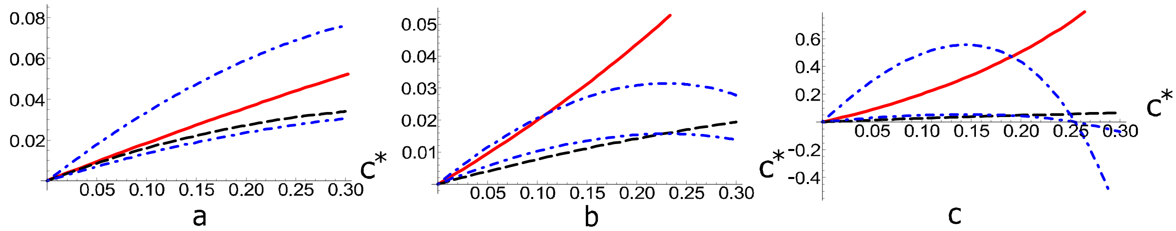

Figure 2.15: Normalized second central moment of electric field magnitude in the composite (solid), Beran 1968 bounds (point-dashed), and Lipton bounds (dashed) ($\varkappa = 1/4$ (a), $\varkappa = -1/4$ (b), and $\varkappa = -3/4$ (c)).

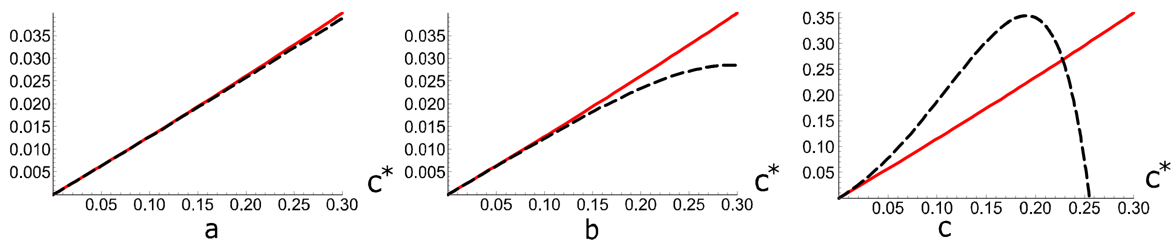

Figure 2.16: Normalized second central moment of electric field magnitude in the matrix (solid) and Beran 1980 bounds (dashed) ($\varkappa = 1/4$ (a), $\varkappa = -1/4$ (b), and $\varkappa = -3/4$ (c)).

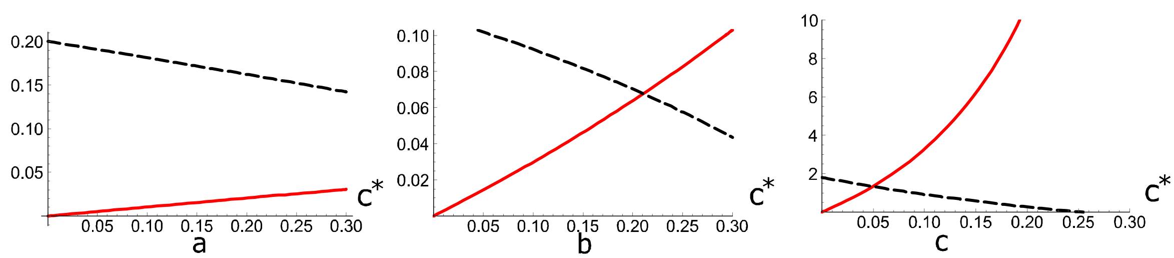

Figure 2.17: Normalized second central moment of electric field magnitude within inclusions (solid) and Beran 1980 bounds (dashed) ($\varkappa = 1/4$ (a), $\varkappa = -1/4$ (b), and $\varkappa = -3/4$ (c)).

CHAPTER 3 APPROXIMATION OF LOCAL FIELDS IN TWO PHASE COMPOSITES

The material conductivity is assumed to be statistically invariant with respect to mirror image and translations, as well as lacking in long range correlation as described in Section 1.3. Take $f\left(a,\vec{u}\right)$ with N field fluctuations in phase 1 and M field fluctuations in phase 2 then

$$f_1\left(\vec{u}\right)=\sum_{\mu} p_\mu \delta\left(\vec{u}-\vec{R}_\mu\right) \quad \text{and} \quad f_2\left(\vec{u}\right)=\sum_{\alpha} q_\alpha \delta\left(\vec{u}-\vec{Q}_\alpha\right), \tag{3.1}$$

where p_μ and q_α are probabilities of the corresponding field fluctuation, variable μ runs values $1...N$, and α runs values $1...M$. Obviously, the field fluctuation probabilities are non-negative, and are constrained by the volume concentration of phases

$$\begin{aligned} 0 \leq\ & \sum_{\mu} p_\mu \equiv c_1,\ 0 \leq p_\mu \leq 1 \ \text{ for each } \mu, \\ 0 \leq\ & \sum_{\alpha} q_\alpha \equiv c_2,\ 0 \leq q_\alpha \leq 1 \ \text{ for each } \alpha, \\ \text{and }\ 1 =\ & c_1 + c_2. \end{aligned} \tag{3.2}$$

Since the average value of the field fluctuations vanish over the composite, it follows that

$$\sum_{\mu} p_\mu \vec{R}_\mu + \sum_{\alpha} q_\alpha \vec{Q}_\alpha = 0. \tag{3.3}$$

Without a loss of generality, let the joint two point probabilities which define the correlation between field fluctuations spatially over $\vec{\tau}$, be defined as the sum of the one point probabilities and some unknown functions $p_{\mu\nu}(\vec{\tau}), s_{\mu\beta}(\vec{\tau})$, and $q_{\alpha\beta}(\vec{\tau})$

$$\begin{aligned} f_{11}(\vec{u};\vec{\tau},\vec{u}') &= f_1(\vec{u})\, f_1(\vec{u}') + \sum_{\mu,\nu} p_{\mu\nu}(\vec{\tau})\, \delta\left(\vec{u}-\vec{R}_\mu\right) \delta\left(\vec{u}'-\vec{R}_\nu\right) \\ &= \sum_{\mu,\nu} \left(p_\mu p_\nu + p_{\mu\nu}(\vec{\tau})\right) \delta\left(\vec{u}-\vec{R}_\mu\right) \delta\left(\vec{u}'-\vec{R}_\nu\right) \\ f_{12}(\vec{u};\vec{\tau},\vec{u}') &= f_1(\vec{u})\, f_2(\vec{u}') + \sum_{\mu,\alpha} s_{\mu\alpha}(\vec{\tau})\, \delta\left(\vec{u}-\vec{R}_\mu\right) \delta\left(\vec{u}'-\vec{Q}_\alpha\right) \\ &= \sum_{\mu,\alpha} \left(p_\mu q_\alpha + s_{\mu\alpha}(\vec{\tau})\right) \delta\left(\vec{u}-\vec{R}_\mu\right) \delta\left(\vec{u}'-\vec{Q}_\alpha\right) \\ f_{21}(\vec{u};\vec{\tau},\vec{u}') &= f_{12}(\vec{u};\vec{\tau},\vec{u}') \\ f_{22}(\vec{u};\vec{\tau},\vec{u}') &= f_2(\vec{u})\, f_2(\vec{u}') + \sum_{\alpha,\beta} q_{\alpha\beta}(\vec{\tau})\, \delta\left(\vec{u}-\vec{Q}_\alpha\right) \delta\left(\vec{u}'-\vec{Q}_\beta\right) \\ &= \sum_{\alpha,\beta} \left(q_\alpha q_\beta + q_{\alpha\beta}(\vec{\tau})\right) \delta\left(\vec{u}-\vec{Q}_\alpha\right) \delta\left(\vec{u}'-\vec{Q}_\beta\right) \end{aligned} \tag{3.4}$$

where variable ν runs values $1...N$ and β runs values $1...M$. Then the compatibility condition of joint probabilities with microstructural characteristics

$$\begin{aligned} &\int f_{11}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u}d\vec{u}' = c_1c_1 + \sum_{\mu,\nu} p_{\mu\nu}(\vec{\tau}) = f_{11}(\vec{\tau}) = c_1c_1 + c_1c_2h_o(\vec{\tau}), \\ &\int f_{12}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u}d\vec{u}' = c_1c_2 + \sum_{\mu,\alpha} s_{\mu\alpha}(\vec{\tau}) = f_{12}(\vec{\tau}) = c_1c_2 - c_1c_2h_o(\vec{\tau}), \\ \text{and } &\int f_{22}(\vec{u};\vec{\tau},\vec{u}')\, d\vec{u}d\vec{u}' = c_2c_2 + \sum_{\alpha,\beta} q_{\alpha\beta}(\vec{\tau}) = f_{22}(\vec{\tau}) = c_2c_2 + c_1c_2h_o(\vec{\tau}), \end{aligned}$$

leads to the relationships

$$\sum_{\mu,\nu} \hat{p}_{\mu\nu}\left(\vec{k}\right) = c_1c_2\hat{h}_o\left(\vec{k}\right), \quad \sum_{\mu,\alpha} \hat{s}_{\mu\alpha}\left(\vec{k}\right) = -c_1c_2\hat{h}_o\left(\vec{k}\right), \quad \text{and} \quad \sum_{\alpha,\beta} \hat{q}_{\alpha\beta}\left(\vec{k}\right) = c_1c_2\hat{h}_o\left(\vec{k}\right) \tag{3.5}$$

which are written in terms of Fourier transforms defined as

$$\hat{h}_o\left(\vec{k}\right) = \int \mathrm{Exp}\left(i\ \vec{\tau}\cdot\vec{k}\right) h_o(\vec{\tau})\, d\vec{\tau}. \tag{3.6}$$

The inverse Fourier transform can be found by

$$h_o(\vec{\tau}) = \int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)d\vec{k}. \tag{3.7}$$

Here the important concept that the solutions which hold for all vectors $\vec{\tau}$ in physical space also hold in the transformed $\vec{k}$ space, and vice versa, is used.

The compatibility condition of one and two point joint distributions

$$\int\left(f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right)+f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right)\right)d\vec{u}' = f_1\left(\vec{u}\right) \quad \text{and} \quad \int\left(f_{21}\left(\vec{u};\vec{\tau},\vec{u}'\right)+f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right)\right)d\vec{u}' = f_2\left(\vec{u}\right)$$

provide further constraints

$$\sum_{\nu} p_{\mu\nu}\left(\vec{\tau}\right) + \sum_{\alpha} s_{\mu\alpha}\left(\vec{\tau}\right) = 0 \;\text{ for each } \mu \text{ and } \sum_{\mu} s_{\mu\beta}\left(\vec{\tau}\right) + \sum_{\alpha} q_{\alpha\beta}\left(\vec{\tau}\right) = 0 \;\text{ for each } \beta$$

and writing in terms of Fourier transforms

$$\sum_{\nu}\hat{p}_{\mu\nu}\left(\vec{k}\right) + \sum_{\alpha}\hat{s}_{\mu\alpha}\left(\vec{k}\right) = 0 \;\text{ for each } \mu \text{ and } \sum_{\mu}\hat{s}_{\mu\beta}\left(\vec{k}\right) + \sum_{\alpha}\hat{q}_{\alpha\beta}\left(\vec{k}\right) = 0 \;\text{ for each } \beta. \tag{3.8}$$

The remaining conditions for non-negativeness of probability are

$$0 \le f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right),\; 0 \le f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right),\; \text{and } 0 \le f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right)$$

which must hold for all values of $\vec{u}$, $\vec{\tau}$, and $\vec{u}'$.

Here, rather than the full infinite series to describe the correlations within the composite, the description has been truncated to the one and two point statistics. This truncation introduces a new constraint to ensure that the joint 2 point probability is positive definite in terms of Fourier transforms; the inequality

$$0 \le \sum_{\mu,\nu}\varphi_\mu\varphi_\nu\hat{p}_{\mu\nu}\left(\vec{k}\right) + 2\sum_{\mu,\alpha}\varphi_\mu\phi_\alpha\hat{s}_{\mu\alpha}\left(\vec{k}\right) + \sum_{\alpha,\beta}\phi_\alpha\phi_\beta\hat{q}_{\alpha\beta}\left(\vec{k}\right) \tag{3.9}$$

must hold for any vector $\vec{\varphi}$ and $\vec{\phi}$.

Lastly, there is a constraint due to the potentiality condition in terms of Fourier transforms:

$$\hat{B}\left(\vec{k}\right)k_ik_j = \sum_{\mu,\nu} R_{\mu i}R_{\nu j}\hat{p}_{\mu\nu}\left(\vec{k}\right) + \sum_{\mu,\alpha}\left(R_{\mu i}Q_{\alpha j} + R_{\mu j}Q_{\alpha i}\right)\hat{s}_{\mu\alpha}\left(\vec{k}\right) + \sum_{\alpha,\beta} Q_{\alpha i}Q_{\beta j}\hat{q}_{\alpha\beta}\left(\vec{k}\right) \quad (3.10)$$

where

$$0 \le \hat{B}\left(\vec{k}\right) \quad \text{for all} \quad \vec{k}.$$

3.1 Simplified Case of One Phase having a Homogenous Field

The constraints can be further simplified, and we will find $\hat{B}\left(\vec{k}\right)$, $\hat{p}_{\mu\nu}\left(\vec{k}\right)$, $\hat{s}_{\mu 1}\left(\vec{k}\right)$, and $\hat{q}_{11}\left(\vec{k}\right)$. Consider the case of a homogenous field in phase 2 and with (3.1), (3.2), and (3.3)

$$\begin{aligned} \vec{Q} &= -\frac{1}{c_2}\sum_{\mu} p_\mu \vec{R}_\mu, \quad 0 \le \sum_{\mu} p_\mu = c_1, \quad 0 \le p_\mu \le 1 \ \text{ for each } \mu, \qquad (3.11) \\ \text{and} \quad 0 &\le q_1 = c_2 = 1 - c_1, \end{aligned}$$

where the subscript for vector $\vec{Q}_1$ has been dropped. This simplifies one point probabilities to

$$f_1(\vec{u}) = \sum_{\mu} p_\mu \delta\left(\vec{u} - \vec{R}_\mu\right) \quad \text{and} \quad f_2(\vec{u}) = c_2\delta\left(\vec{u} - \vec{Q}\right),$$

and two point probabilities to

$$\begin{aligned} 0 \le \frac{f_{11}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} &= \sum_{\mu,\nu}\left(\frac{p_{\mu\nu}(\vec{\tau})}{c_1c_2} + \frac{p_\mu p_\nu}{c_1c_2}\right)\delta\left(\vec{u} - \vec{R}_\mu\right)\delta\left(\vec{u}' - \vec{R}_\nu\right) \qquad (3.12) \\ 0 \le \frac{f_{12}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} &= \sum_{\mu}\left(\frac{s_{\mu 1}(\vec{\tau})}{c_1c_2} + \frac{p_\mu}{c_1}\right)\delta\left(\vec{u} - \vec{R}_\mu\right)\delta\left(\vec{u}' - \vec{Q}\right) \\ 0 \le \frac{f_{21}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} &= \frac{f_{12}(\vec{u};0,\vec{u}')}{c_1c_2} \\ 0 \le \frac{f_{22}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} &= \left(\frac{q_{11}(\vec{\tau})}{c_1c_2} + \frac{c_2}{c_1}\right)\delta\left(\vec{u} - \vec{Q}\right)\delta\left(\vec{u}' - \vec{Q}\right). \end{aligned}$$

With (3.5)

$$\sum_{\mu,\nu}\hat{p}_{\mu\nu}\left(\vec{k}\right)=c_1c_2\hat{h}_o\left(\vec{k}\right),\quad \sum_{\mu}\hat{s}_{\mu 1}\left(\vec{k}\right)=-c_1c_2\hat{h}_o\left(\vec{k}\right),\quad \text{and}\quad \hat{q}_{11}\left(\vec{k}\right)=c_1c_2\hat{h}_o\left(\vec{k}\right) \tag{3.13}$$

and (3.8)

$$\sum_{\nu}\hat{p}_{\mu\nu}\left(\vec{k}\right)+\hat{s}_{\mu 1}\left(\vec{k}\right)=0 \;\text{ for each } \mu \text{ and } \sum_{\mu}\hat{s}_{\mu 1}\left(\vec{k}\right)+\hat{q}_{11}\left(\vec{k}\right)=0 \tag{3.14}$$

$\hat{q}_{11}\left(\vec{k}\right)$ is known, the second constraint in (3.13) is redundant, and the fourth term in (3.12) is always satisfied due to the definition of $0\leq h_o\left(\vec{\tau}\right)$ (1.8).

Positive definiteness (3.9) takes the form

$$0\leq \sum_{\mu,\nu}\varphi_\mu\varphi_\nu\hat{p}_{\mu\nu}\left(\vec{k}\right)+2\sum_{\mu}\varphi_\mu\phi_1\hat{s}_{\mu 1}\left(\vec{k}\right)+\phi_1^2c_1c_2\hat{h}_o\left(\vec{k}\right),$$

which after rearrangement is

$$0\leq \sum_{\mu,\nu}\varphi_\mu\varphi_\nu\hat{p}_{\mu\nu}+c_1c_2\hat{h}_o\left(\phi_1+\frac{1}{c_1c_2\hat{h}_o}\sum_{\mu}\varphi_\mu\hat{s}_{\mu 1}\right)^2-\frac{1}{c_1c_2\hat{h}_o}\left(\sum_{\mu}\varphi_\mu\hat{s}_{\mu 1}\right)^2.$$

By minimization of the right side over ϕ_1 the constraint follows

$$0\leq\left(\hat{p}_{\mu\nu}\left(\vec{k}\right)-\frac{1}{c_1c_2\hat{h}_o\left(\vec{k}\right)}\hat{s}_{\mu 1}\left(\vec{k}\right)\hat{s}_{\nu 1}\left(\vec{k}\right)\right)\varphi_\mu\varphi_\nu. \tag{3.15}$$

Repeated indices implies summation.

The potentiality condition (3.10) is

$$\hat{B}\left(\vec{k}\right)k_ik_j=\left(\hat{p}_{\mu\nu}\left(\vec{k}\right)-\frac{1}{c_2}\left(p_\mu\hat{s}_{\nu 1}\left(\vec{k}\right)+p_\nu\hat{s}_{\mu 1}\left(\vec{k}\right)\right)+\frac{1}{\left(c_2\right)^2}p_\mu p_\nu c_1c_2\hat{h}_o\left(\vec{k}\right)\right)R_{\mu i}R_{\nu j}. \tag{3.16}$$

3.2 Compatibility of Probability Distributions

By introduction of vector ξ_μ where μ runs $1, ..., N$ and all components of ξ_μ are equal to unity, tensors $\hat{p}_{\mu\nu}\left(\vec{k}\right)$ and $\hat{s}_\mu\left(\vec{k}\right)$ can be expressed in the general bilinear form comprised of parts orthogonal and collinear to ξ_μ :

$$\hat{p}_{\mu\nu}\left(\vec{k}\right) = p'_{\mu\nu}\left(\vec{k}\right) + p'_\mu\left(\vec{k}\right)\xi_\nu + \xi_\mu p'_\nu\left(\vec{k}\right) + \bar{p}\left(\vec{k}\right)\xi_\mu\xi_\nu \quad \text{and} \quad \hat{s}_\mu\left(\vec{k}\right) = s'_\mu\left(\vec{k}\right) + \bar{s}\left(\vec{k}\right)\xi_\mu.$$

By definition, the orthogonal parts

$$p'_{\mu\nu}\left(\vec{k}\right)\xi_\mu = 0, \quad p'_{\mu\nu}\left(\vec{k}\right)\xi_\nu = 0, \quad p'_\mu\left(\vec{k}\right)\xi_\mu = 0, \quad \text{and} \quad s'_\mu\left(\vec{k}\right)\xi_\mu = 0.$$

Since remaining constraints in (3.13) can be written

$$\xi_\mu\xi_\nu\hat{p}_{\mu\nu}\left(\vec{k}\right) = N^2\bar{p}\left(\vec{k}\right) = c_1c_2\hat{h}_o\left(\vec{k}\right) \quad \text{and} \quad \xi_\mu\hat{s}_\mu\left(\vec{k}\right) = N\bar{s}\left(\vec{k}\right) = -c_1c_2\hat{h}_o\left(\vec{k}\right)$$

we then know

$$\bar{p}\left(\vec{k}\right) = \frac{1}{N^2}c_1c_2\hat{h}_o\left(\vec{k}\right) \quad \text{and} \quad \bar{s}\left(\vec{k}\right) = -\frac{1}{N}c_1c_2\hat{h}_o\left(\vec{k}\right).$$

With the first constraint in (3.14)

$$Np'_\mu\left(\vec{k}\right) + N\bar{p}\left(\vec{k}\right)\xi_\mu = -s'_\mu\left(\vec{k}\right) - \bar{s}\left(\vec{k}\right)\xi_\mu,$$

we also know

$$p'_\mu\left(\vec{k}\right) = -\frac{1}{N}s'_\mu\left(\vec{k}\right).$$

Then, $\hat{p}_{\mu\nu}\left(\vec{k}\right)$ and $\hat{s}_\mu\left(\vec{k}\right)$ which comply to constraints (3.13,3.14) are written

$$\hat{p}_{\mu\nu}\left(\vec{k}\right) = p'_{\mu\nu}\left(\vec{k}\right) - \frac{1}{N}s'_\mu\left(\vec{k}\right)\xi_\nu - \frac{1}{N}s'_\nu\left(\vec{k}\right)\xi_\mu + \frac{1}{N^2}c_1c_2\hat{h}_o\left(\vec{k}\right)\xi_\mu\xi_\nu \tag{3.17}$$

$$\text{and} \quad \hat{s}_\mu\left(\vec{k}\right) = s'_\mu\left(\vec{k}\right) - \frac{1}{N}c_1c_2\hat{h}_o\left(\vec{k}\right)\xi_\mu.$$

Now, consider positive definiteness (3.15) with the similar decomposition of vector $\vec{\varphi}$

$$\varphi_\mu = \varphi'_\mu + \varphi\xi_\mu \quad \text{where } \varphi'_\mu\xi_\mu = 0,$$

with (3.17) , after simplifications we have

$$0 \leq \left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right) s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} \right) \varphi'_\mu\varphi'_\nu. \tag{3.18}$$

The potentiality condition (3.16) with (3.17) is

$$\begin{aligned}
\hat{B}\left(\vec{k}\right) k_ik_j = & \left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right) s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} \right) R_{\mu i}R_{\nu j} \\
& + \left(\frac{s'_\mu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} - \left(\frac{1}{c_2}p_\mu + \frac{1}{N}\xi_\mu \right) \right) \\
& \text{x} \left(\frac{s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} - \left(\frac{1}{c_2}p_\nu + \frac{1}{N}\xi_\nu \right) \right) c_1c_2\hat{h}_o\left(\vec{k}\right) R_{\mu i}R_{\nu j}
\end{aligned}$$

and with the similar decomposition of the field fluctuations $\vec{R}_\mu$

$$R_{\mu i} = R'_{\mu i} + \xi_\mu\bar{R}_i \quad \text{where} \quad \bar{R}_i \equiv \frac{1}{N}\xi_\mu R_{\mu i} \text{ and } \quad R'_{\mu i}\xi_\mu = 0 \quad \text{for each } i,$$

potentiality is after simplifications

$$\begin{aligned}
\hat{B}\left(\vec{k}\right) k_ik_j \quad = \quad & \left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right) s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} \right) R'_{\mu i}R'_{\nu j} \\
& + \left(\frac{s'_\mu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} R'_{\mu i} - \frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right) \right) \\
& \text{x} \left(\frac{s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} R'_{\nu j} - \frac{1}{c_2}\left(p_\nu R'_{\nu j} + \bar{R}_j\right) \right) c_1c_2\hat{h}_o\left(\vec{k}\right).
\end{aligned} \tag{3.19}$$

3.3 General Solution of Field Fluctuation Correlations

In Appendix D, the positive definiteness and potentiality conditions are shown to be constraints between $p'_{\mu\nu}\left(\vec{k}\right)$ and $s'_{\mu}\left(\vec{k}\right)$. With these general solutions, the field realized for any composite now is a potential one, or more compactly the potentiality condition is achieved. These general solutions which ensure the potentiality condition is achieved are

$$\begin{aligned}\left(p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_{\mu}\left(\vec{k}\right)s'_{\nu}\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j} &= \Psi\left(\vec{k}\right)k_ik_j \\ \text{and}\quad \frac{s'_{\mu}\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}R'_{\mu i}-\frac{1}{c_2}\left(p_{\mu}R'_{\mu i}+\bar{R}_i\right) &= \Phi\left(\vec{k}\right)k_i\end{aligned}\tag{3.20}$$

where

$$0\leq\hat{B}\left(\vec{k}\right)=\Psi\left(\vec{k}\right)+c_1c_2\hat{h}_o\left(\vec{k}\right)\left(\Phi\left(\vec{k}\right)\right)^2.$$

We are after particular solutions which allow determination of $s_{\mu 1}\left(\vec{\tau}\right)$ and $p_{\mu\nu}\left(\vec{\tau}\right)$; the particular solutions to (3.20) are

$$\begin{aligned}p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_{\mu}\left(\vec{k}\right)s'_{\nu}\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)} &= \Psi\left(\vec{k}\right)Y^i_{\mu}k_iY^j_{\nu}k_j \\ \text{and}\quad s'_{\mu}\left(\vec{k}\right) &= c_1c_2\hat{h}_o\left(\vec{k}\right)\left(\Phi\left(\vec{k}\right)Y^i_{\mu}k_i-X_{\mu}\right)+\tilde{s}_{\mu}\left(\vec{k}\right)\end{aligned}\tag{3.21}$$

where fulfillment of the potentiality condition (i.e. general solutions (3.20)) requiring

$$\begin{aligned}Y^i_{\mu}R'_{\mu j} &= \delta^i_j,\quad Y^i_{\mu}\xi_{\mu}=0, \\ X_{\mu}R'_{\mu i} &= -\frac{1}{c_2}\left(p_{\mu}R'_{\mu i}+\bar{R}_i\right),\quad X_{\mu}\xi_{\mu}=0, \\ \tilde{s}_{\mu}\left(\vec{k}\right)R'_{\mu i} &= 0,\quad \text{and}\quad s'_{\mu}\left(\vec{k}\right)\xi_{\mu}=0;\end{aligned}\tag{3.22}$$

it will be shown later that $\tilde{s}_{\mu}\left(\vec{k}\right)=0$, and it is assumed here.

Then, with (3.17) and the inverse Fourier transform (3.7) of (3.21)

$$\frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2} = \frac{s'_{\mu}(\vec{\tau})}{c_1 c_2} - \frac{1}{N}\xi_\mu h_o(\vec{\tau}) = Y^i_\mu \gamma_i - \left(X_\mu + \frac{1}{N}\xi_\mu\right) h_o(\vec{\tau})$$

and

$$\begin{aligned}\frac{p_{\mu\nu}(\vec{\tau})}{c_1 c_2} &= \frac{p'_{\mu\nu}(\vec{\tau})}{c_1 c_2} - \frac{\xi_\nu}{N}\frac{s'_\mu(\vec{\tau})}{c_1 c_2} - \frac{\xi_\mu}{N}\frac{s'_\nu(\vec{\tau})}{c_1 c_2} + \frac{\xi_\mu \xi_\nu}{N^2} h_o(\vec{\tau}) \\ &= \left(\Gamma_{ij} + \gamma_{ij}\right) Y^i_\mu Y^j_\nu - Y^i_\mu \gamma_i \left(X_\nu + \frac{1}{N}\xi_\nu\right) - \left(X_\mu + \frac{1}{N}\xi_\mu\right) Y^j_\nu \gamma_j + \\ &\left(X_\mu + \frac{1}{N}\xi_\mu\right)\left(X_\nu + \frac{1}{N}\xi_\nu\right) h_o(\vec{\tau})\end{aligned}$$

where

$$\begin{aligned}\Gamma_{ij}(\vec{\tau}) &\equiv \frac{1}{c_1 c_2}\int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right)\Psi\left(\vec{k}\right) k_i k_j dV_k, \\ \gamma_{ij}(\vec{\tau}) &\equiv \int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)\Phi^2\left(\vec{k}\right) k_i k_j dV_k, \\ \text{and}\quad \gamma_i(\vec{\tau}) &\equiv \int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)\Phi\left(\vec{k}\right) k_i dV_k.\end{aligned}$$

A simple case is when

$$\Psi\left(\vec{k}\right) = \psi\; c_1 c_2 \hat{h}_o\left(\vec{k}\right)\Phi^2\left(\vec{k}\right) \quad \text{then } \Gamma_{ij}(\vec{\tau}) = \psi\;\gamma_{ij}(\vec{\tau});$$

it corresponds the reasonable assumption that the intra phase correlations $p'_{\mu\nu}\left(\vec{k}\right)$ are only functions of microstructural characteristics $c_1 c_2 \hat{h}_o\left(\vec{k}\right)$ and the interphase correlations $s'_\mu\left(\vec{k}\right)$. With this definition, the positive definiteness condition reduces to

$$0 \leq \psi.$$

From this point on we will only consider the degenerated case, of $\psi = 0$ which gives

$$p'_{\mu\nu}\left(\vec{k}\right) = \frac{s'_{\mu}\left(\vec{k}\right) s'_{\nu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)};$$

then the positive definiteness condition is always satisfied and the potentiality condition has now been reduced to (3.22). Remaining constraints are now only related to the non-negativity of probabilities $(3.11), (3.12)$ and potentiality (3.22).

3.4 Symmetry of Internal Fields in Two Dimensional Case

Motivated by the results of Chapter 2, where PD of electric field in a composite comprised of spherical inclusions was found, we take the reasonable assumption that the PD must be symmetric in the direction orthogonal to the applied field. Consequently we take field fluctuations directed along or in equiprobable pairs symmetric to the applied field direction, which provides this feature in the case of two dimensional space. Without loss of generality we take $|\vec{v}|$ in the $1-$direction. It then a consequence that $p_{\mu} R'_{\mu 2}$ and $\bar{R}_2$ must be zero, which simplifies the constraints in the second line of (3.22)

$$X_{\mu} R'_{\mu 1} = -\frac{1}{c_2}\left(p_{\mu} R'_{\mu 1} + \bar{R}_1\right) \quad \text{and} \quad X_{\mu} R'_{\mu 2} = 0. \tag{3.23}$$

3.5 Debye Microstructural Statistics

The microstructure selected for study is the Debye type, which is commonly encountered in a wide range of engineering materials and corresponds to microstructures comprised of randomly placed inclusions such as the particulate type microstructure studied in Chapter 2 (see e.g. [4]):

$$h_o\left(\vec{\tau}\right) = \mathrm{Exp}[-\left|\vec{\tau}\right|] \quad \text{and} \quad \hat{h}_o\left(\vec{k}\right) = \frac{2\pi}{\left(1 + \left|\vec{k}\right|^2\right)^{3/2}},$$

where $dV_k = \frac{1}{(2\pi)^2} dk_1 dk_2$ for the two dimensional case.

3.6 Selected Statistical Characteristics of the Field Fluctuation

We must also select statistical characteristics of the field fluctuation correlations $f_{11}(\vec{u};\vec{\tau},\vec{u}')$ and $f_{12}(\vec{u};\vec{\tau},\vec{u}')$ which also defines the correlation function of the potential field $B(\vec{\tau})$. Consider the second equation of (3.20) the right hand side of must be even based on definitions, therefore Φ must be an odd function. We take it as the function which has the slowest rate of decay allowing for the convergence of the integrals of $\gamma_{ij}(\vec{\tau})$ and $\gamma_i(\vec{\tau})$:

$$\Phi\left(\vec{k}\right)=\frac{\alpha_i\ k_i}{1+\left|\vec{k}\right|^2}.$$

The use of the slowest rate of decay in Fourier space was chosen because we expect that the fluctuations should be correlated very strongly locally.Then

$$\begin{aligned}
\gamma_j(\vec{\tau}) &= \int \mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)\frac{2\pi}{\left(1+|k|^2\right)^{3/2}}\frac{\alpha_i\ k_i}{1+\left|\vec{k}\right|^2}k_j dV_k \qquad (3.24)\\
&= -\alpha_i\frac{\partial^2}{\partial\tau_i\partial\tau_j}\int\frac{\mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)}{\left(1+|k|^2\right)^{5/2}}\frac{1}{2\pi}dk_1dk_2\\
&= -\alpha_i\frac{\partial^2}{\partial\tau_i\partial\tau_j}\int\frac{\mathrm{Exp}\left(-i\ |\vec{\tau}|\ |k|\ \mathrm{Cos}(\theta)\right)}{\left(1+|k|^2\right)^{5/2}}|k|\,d|k|\,\frac{1}{2\pi}d\theta\\
&= -\frac{1}{3}\alpha_i\frac{\partial^2}{\partial\tau_i\partial\tau_j}\mathrm{Exp}\left(-|\vec{\tau}|\right)\left(1+|\vec{\tau}|\right)\\
&= -\frac{1}{3}\alpha_i\left(\frac{\tau_i\tau_j}{|\vec{\tau}|}-\delta_{ij}\right)\mathrm{Exp}\left(-|\vec{\tau}|\right)
\end{aligned}$$

and by similar means we find

$$\gamma_{ij}(\vec{\tau}) = \int \mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right) \frac{2\pi}{\left(1+|k|^2\right)^{3/2}} \frac{\alpha_i\ k_i}{1+\left|\vec{k}\right|^2} \frac{\alpha_j\ k_j}{1+\left|\vec{k}\right|^2} dV_k \tag{3.25}$$

$$= \frac{1}{15}\alpha_m\alpha_n \frac{\partial^2}{\partial\tau_i\partial\tau_j}\frac{\partial^2}{\partial\tau_m\partial\tau_n}\mathrm{Exp}\left(-|\vec{\tau}|\right)\left(3+3|\vec{\tau}|+|\vec{\tau}|^2\right)$$

$$= \frac{1}{15}\alpha_m\alpha_n \frac{\partial^2}{\partial\tau_i\partial\tau_j}\mathrm{Exp}\left(-|\vec{\tau}|\right)\left(\tau_m\tau_n-\delta_{mn}\left(1+|\vec{\tau}|\right)\right)$$

$$= \frac{1}{15}\alpha_m\alpha_n\mathrm{Exp}\left(-|\vec{\tau}|\right)\left(\left(\left(1+\frac{1}{|\vec{\tau}|}\right)\frac{\tau_m\tau_n}{|\vec{\tau}|^2}-\frac{\delta_{mn}}{|\vec{\tau}|}\right)\tau_i\tau_j\right.$$
$$\left.+\left(\delta_{ij}\delta_{mn}+\delta_{im}\delta_{jn}+\delta_{in}\delta_{jm}\right)-\frac{1}{|\vec{\tau}|}\left(\tau_m\tau_n\delta_{ij}+\tau_j\tau_m\delta_{in}+\tau_j\tau_n\delta_{im}+\tau_i\tau_m\delta_{jn}+\tau_i\tau_n\delta_{jm}\right)\right)$$

The correlation function of the potential field can now be found

$$\frac{B(\vec{\tau})}{c_1c_2} = \alpha_1\alpha_1\frac{\partial^2}{\partial\tau_1\partial\tau_1}\int\frac{\mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)}{\left(1+\left|\vec{k}\right|^2\right)^{7/2}}\frac{1}{2\pi}dk_1dk_2 \tag{3.26}$$

$$= \alpha_1\alpha_1\frac{\partial^2}{\partial\tau_1\partial\tau_1}\frac{1}{15}\mathrm{Exp}\left(-|\vec{\tau}|\right)\left(3+3|\vec{\tau}|+|\vec{\tau}|^2\right)$$

$$= \alpha_1\alpha_1\frac{1}{15}\left(\tau_1\tau_1-|\vec{\tau}|-1\right)\mathrm{Exp}\left(-|\vec{\tau}|\right).$$

3.7 Non-Negativity of Joint Two Point Probabilities

From (3.12) we are left with the constraints

$$0 \le \frac{f_{11}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} = \sum_{\mu,\nu}\left(\frac{p_{\mu\nu}(\vec{\tau})}{c_1c_2}+\frac{p_\mu p_\nu}{c_1c_2}\right)\delta\left(\vec{u}-\vec{R}_\mu\right)\delta\left(\vec{u}'-\vec{R}_\nu\right)$$
$$0 \le \frac{f_{12}(\vec{u};\vec{\tau},\vec{u}')}{c_1c_2} = \sum_{\mu}\left(\frac{s_{\mu1}(\vec{\tau})}{c_1c_2}+\frac{p_\mu}{c_1}\right)\delta\left(\vec{u}-\vec{R}_\mu\right)\delta\left(\vec{u}'-\vec{Q}\right);$$

the last two terms have been dropped due to redundancy ($f_{12}(\vec{u};\vec{\tau},\vec{u}') = f_{21}(\vec{u};\vec{\tau},\vec{u}')$ since $s_{\mu1}(\vec{\tau}) = s_{\mu1}(-\vec{\tau})$) and satisfaction of the last constraint (due to non-negativity of $h_o(\vec{\tau})$). For $f_{11}(\vec{u};\vec{\tau},\vec{u}')$ we take each of the $\vec{R}_\mu$ as unique otherwise the problem is degenerate to a lower number of vectors. For example if we were considering three vectors in the first phase, but two were identical, then this can be easily shown to be mathematically equivalent to considering two vectors in the first phase. Therefore we must satisfy the conditions for $f_{11}(\vec{u}; \vec{\tau}, \vec{u}')$ to ensure non negativity of probabilities

$$0 \leq \frac{p_{\mu\nu}(\vec{\tau})}{c_1 c_2} + \frac{p_\mu p_\nu}{c_1 c_2} \quad \text{for each } \mu \text{ and } \ \nu. \tag{3.27}$$

For $f_{12}(\vec{u}; \vec{\tau}, \vec{u}')$ to ensure non negativity of probabilities given the uniqueness of $\vec{R}_\mu$ we have

$$0 \leq \frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2} + \frac{p_\mu}{c_1}. \tag{3.28}$$

The non-negativity of PD in (3.28) can be simplified further by writing $\vec{\tau}$ in polar coordinates and it will be shown that this constraint can be reduced to a constraint of $|\vec{\tau}|$ only. The first term in (3.28) can be further simplified; consider in polar coordinates and the double angle trigonometric identities

$$\begin{aligned}
\alpha_i Y_\mu^j \frac{\tau_i \tau_j}{|\vec{\tau}|^2} &= \frac{1}{|\vec{\tau}|^2}\left(\alpha_1 Y_\mu^1 (\tau_1)^2 + \left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)\tau_1\tau_2 + \alpha_2 Y_\mu^2 (\tau_2)^2\right) \\
&= \alpha_1 Y_\mu^1 \frac{1+\cos 2\theta}{2} + \left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)\frac{\sin 2\theta}{2} + \alpha_2 Y_\mu^2 \frac{1-\cos 2\theta}{2} \\
&= \frac{1}{2}\left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right) + \frac{1}{2}\sqrt{\left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)^2 + \left(\alpha_1 Y_\mu^1 - \alpha_2 Y_\mu^2\right)^2}\sin(2\theta + \Theta).
\end{aligned}$$

Here, Θ is a phase shift which arises from the linear combination of $\sin 2\theta$ and $\cos 2\theta$. Since the constraint on probability (3.28) must hold for all $\vec{\tau}$, the strongest form of this constraint is desired for any Θ and θ. Since the first term in (3.28) enters with a negative coefficient, we maximize to find $\sin(2\theta + \Theta) = 1$ and then

$$\begin{aligned}
\alpha_i Y_\mu^j \left(\frac{\tau_i \tau_j}{|\vec{\tau}|} - \delta_{ij}\right)\mathrm{Exp}(-|\vec{\tau}|) = \left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right)\left(\frac{1}{2}|\vec{\tau}| - 1\right)\mathrm{Exp}(-|\vec{\tau}|) \\
+ \sqrt{\left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)^2 + \left(\alpha_1 Y_\mu^1 - \alpha_2 Y_\mu^2\right)^2}\frac{1}{2}|\vec{\tau}|\,\mathrm{Exp}(-|\vec{\tau}|).
\end{aligned}$$

Inserting this relation into (3.28) we have the constraint as a function of $|\vec{\tau}|$ only

$$
\begin{aligned}
0 \leq & -\frac{1}{3}\left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right)\left(\frac{1}{2}|\vec{\tau}| - 1\right)\mathrm{Exp}\left(-|\vec{\tau}|\right) - \left(X_\mu + \frac{1}{N}\xi_\mu\right)\mathrm{Exp}\left(-|\vec{\tau}|\right) + \frac{p_\mu}{c_1} \\
& -\frac{1}{3}\sqrt{\left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)^2 + \left(\alpha_1 Y_\mu^1 - \alpha_2 Y_\mu^2\right)^2}\frac{1}{2}|\vec{\tau}|\,\mathrm{Exp}\left(-|\vec{\tau}|\right)
\end{aligned} \tag{3.29}
$$

or more compactly for the probability densities in (3.28) we have

$$
0 \leq F_\mu\left(|\vec{\tau}|\right) = A_\mu + B_\mu\ \mathrm{Exp}\left(-|\vec{\tau}|\right) - C_\mu\frac{1}{2}|\vec{\tau}|\,\mathrm{Exp}\left(-|\vec{\tau}|\right) \tag{3.30}
$$

where

$$
\begin{aligned}
A_\mu &= \frac{p_\mu}{c_1} \geq 0 \\
B_\mu &= \frac{1}{3}\left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right) - \left(X_\mu + \frac{1}{N}\xi_\mu\right) \\
C_\mu &= \frac{1}{3}\sqrt{\left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)^2 + \left(\alpha_1 Y_\mu^1 - \alpha_2 Y_\mu^2\right)^2} + \frac{1}{3}\left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right) \geq 0.
\end{aligned} \tag{3.31}
$$

Considering the following relationship

$$
0 \leq \left(\left(\alpha_1 Y_\mu^2 + \alpha_2 Y_\mu^1\right)^2 + \left(\alpha_1 Y_\mu^1 - \alpha_2 Y_\mu^2\right)^2\right) - \left(\alpha_1 Y_\mu^1 + \alpha_2 Y_\mu^2\right)^2 = \left(\alpha_1 Y_\mu^2 - \alpha_2 Y_\mu^1\right)^2,
$$

the first term in C_μ, which is always positive, is always equal to or greater than the absolute value of the second term, therefore C_μ is always non-negative as noted in (3.31).

There are three possible minimizers to $F_\mu\left(|\vec{\tau}|\right): 0,\ \infty$, and depending on the values of B_μ and C_μ possibly an intermediate point denoted $\tilde{\tau}_\mu$. Let us show why these three conditions ensure the non-negativeness in (3.30). For derivatives we have

$$
F_\mu'\left(|\vec{\tau}|\right) = -\left(B_\mu + C_\mu\frac{1}{2}\left(1 - |\vec{\tau}|\right)\right)\mathrm{Exp}\left(-|\vec{\tau}|\right)
$$

where prime indicates differentiation with respect to $|\vec{\tau}|$. Considering values $-\infty \leq |\vec{\tau}| \leq \infty$, the function $F_\mu(|\vec{\tau}|)$ has stationary point $(F'_\mu(|\vec{\tau}|) = 0)$ for finite $|\vec{\tau}|$ only at the point denoted denoted $\tilde{\tau}_\mu$,

$$\tilde{\tau}_\mu = 1 + 2\frac{B_\mu}{C_\mu}.$$

The second derivative with respect to $|\vec{\tau}|$ is

$$F''_\mu(|\vec{\tau}|) = \left(B_\mu + C_\mu\left(1 - \frac{1}{2}|\vec{\tau}|\right)\right) \operatorname{Exp}(-|\vec{\tau}|),$$

and at $\tilde{\tau}_\mu$

$$F''_\mu(\tilde{\tau}_\mu) = \frac{1}{2}C_\mu \operatorname{Exp}(-\tilde{\tau}_\mu).$$

At the point $\tilde{\tau}_\mu$, the first derivative is zero, so this is a stationary point and the second derivative is non-negative since $0 \leq C_\mu$; therefore, the stationary point $\tilde{\tau}_\mu$ corresponds to a minimum.

In the case $0 = F_\mu(0)$, then $B_\mu = -A_\mu = -p_\mu/c_1$ and the stationary point $\tilde{\tau}_\mu$ cannot not have a positive value

$$\tilde{\tau}_\mu = 1 + 2\frac{B_\mu}{C_\mu} = 1 - 2\frac{p_\mu}{c_1}\frac{1}{C_\mu} \leq 0 \quad \text{thus} \quad C_\mu \leq 2\frac{p_\mu}{c_1}.$$

This conditional statement also arrises from alternative reasoning: if $0 = F_\mu(0)$ and there is only one stationary point for all $|\vec{\tau}|$, to satisfy the condition $0 \leq F_\mu(|\vec{\tau}|)$ for $0 \leq |\vec{\tau}|$, the first derivative of $F_\mu(|\vec{\tau}|)$ with respect to $|\vec{\tau}|$ at $|\vec{\tau}| = 0$ must be non-negative

$$0 \leq F'_\mu(0) = -\left(B_\mu + \frac{C_\mu}{2}\right) = \frac{p_\mu}{c_1} - \frac{C_\mu}{2} \quad \text{thus} \quad C_\mu \leq 2\frac{p_\mu}{c_1}.$$

Note also shown that C_μ is non-negative, therefore

$$0 \leq C_\mu \leq 2\frac{p_\mu}{c_1} \quad \text{if} \quad 0 = F_\mu(0) \quad \text{for each} \quad \mu \tag{3.32}$$

3.8 Two Dimensional Debye Material

In summary for the two dimensional case with the Debye material and the correlation of fluctuations selected, a random composite must have non-negative one point probabilities

$$0 \leq c_1 = \sum_{\mu} p_\mu \leq 1, \quad c_2 = 1 - c_1, \quad 0 \leq p_\mu \text{ for each } \mu \tag{3.33}$$

and two point probabilities $(3.28, 3.30)$. The fulfillment of potentiality requires

$$\begin{aligned} \xi_\mu R'_{\mu i} &= 0, \; \xi_\mu X_\mu = 0, \quad X_\mu R'_{\mu 1} = -\frac{1}{c_2}\left(p_\mu R'_{\mu 1} + \bar{R}_1\right), \\ X_\mu R'_{\mu 2} &= 0, \quad Y^i_\mu R'_{\mu j} = \delta^i_j, \quad \text{and} \quad \xi_\mu Y^i_\mu = 0; \end{aligned} \tag{3.34}$$

all other required constraints have been satisfied through the structure of this solution.

3.9 Number of Field Fluctuations to Satisfy Potentiality Condition

To satisfy the potentiality condition in the two dimensional case, two values of field fluctuation in the first phase are insufficient except for the degenerate case of a uniform field for each phase. For the case of two dimensions a single vector orthogonal to $\vec{k}$ exists and is denoted $\vec{k}^*$ (i.e. $k_i k^*_i = 0)$. Then, consider the case when $\vec{R}_1 = const\ \vec{R}_2$, where the potentiality condition has the form

$$\hat{B}\left(\vec{k}\right) k_i k_j = T\left(\vec{k}\right) R_{1i} R_{1j} :$$

contracting with $k^*_i k^*_j$ we have

$$0 = T\left(\vec{k}\right) \left(R_{1i} k^*_i\right)^2,$$

which must hold for all $\vec{k}$. Since $T\left(\vec{k}\right)$ is a non zero function of $\vec{k}$, and this equation should hold for arbitrary values of $\vec{k}^*$, the only solution is a homogenous field, $R_{1i} = 0$. It is shown in Appendix C that when $\vec{R}_1 \neq const\ \vec{R}_2$ the same conclusion occurs.

We then know $R'_{\mu 1} \neq const\ R'_{\mu 2}$, and with the conditions $\tilde{s}_\mu R'_{\mu i} = 0$ and $\tilde{s}_\mu \xi_\mu = 0$ we have the solution that $\tilde{s}_\mu\left(\vec{k}\right) = 0$.

Note this same argument holds for the case of homogenous fields within each phase. Again, the field in the second phase $\vec{Q}_1$ can be expressed as a function of the field in the first phase $\vec{R}_1$ using (3.11).

Therefore from the perspective of fulfilling the potentiality condition in the two dimensional case, more than two field fluctuations in the first phase are required for a two phase composite; as shown in the following section, three is sufficient.

3.10 Three Field Fluctuations in Phase 1 with a Homogenous Field in Phase 2

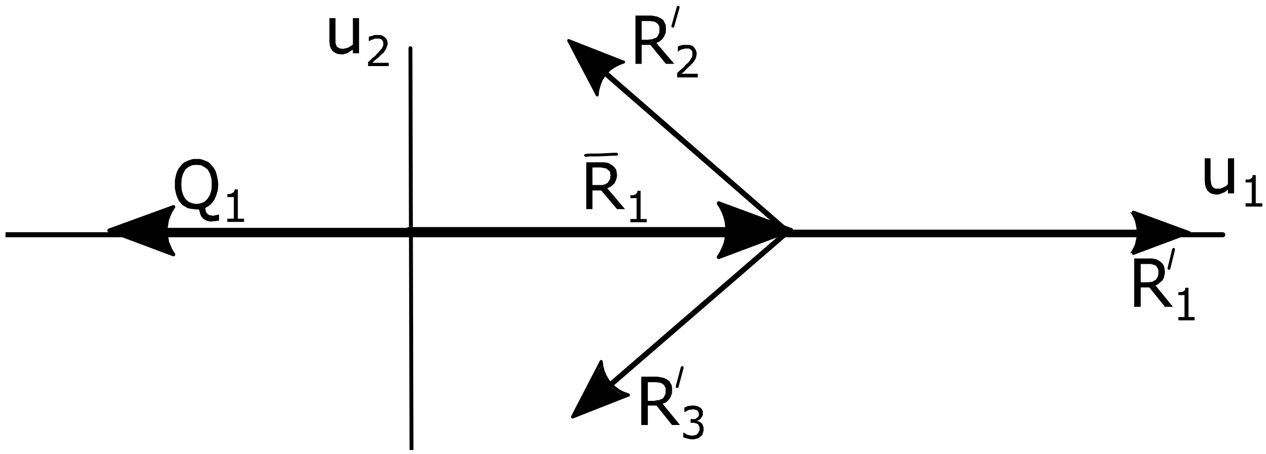

Figure 3.1: Diagram for $N\mu = 3$, three vectors in the first phase $\vec{R}_\mu$ and one in second $\vec{Q}$

In the case of three field fluctuations as shown in Fig. 3.1, the constraints relating to potentiality and vector probabilities are fulfilled

$$p_2 = p_3 = \frac{1}{2}\left(c_1 - p_1\right), \quad R'_{12} = 0, \quad R'_{22} = -R'_{32}, \quad R'_{21} = R'_{31} = -\frac{1}{2}R'_{11},$$
$$X_1 = \frac{2}{3}\frac{1}{c_2}\left(\frac{1}{2}\left(c_1 - 3p_1\right) - \frac{\bar{R}_1}{R'_{11}}\right), \quad X_2 = X_3 = -\frac{1}{2}X_1,$$
$$Y_1^1 = \frac{2}{3}\frac{1}{R'_{11}}, \quad Y_2^1 = Y_3^1 = -\frac{1}{2}Y_1^1, \; Y_1^2 = 0, \quad \text{and} \quad Y_2^2 = -Y_3^2 = \frac{1}{2}\frac{1}{R'_{22}}$$

with unknowns p_1, R'_{11}, R'_{22}, $\bar{R}_1$, and α_i with only constraints remaining relating to the non-negativeness of PD $(3.27, 3.30)$. The vectors $\vec{R}_1$ and $\vec{R}_2$ as well as the components R_{22} and R_{32} must be non zero, otherwise this case degenerates to two fluctuations in the first phase and potentiality cannot be satisfied.

In the case of three field fluctuations in the first phase, the constraints (3.27) for $|\vec{\tau}| = 0$

$$\begin{aligned}
0 \leq \;\; F_1(0) = & \quad A_1 + B_1 &= \frac{1}{3}\alpha_1 Y_1^1 + \left(\frac{p_1}{c_1} - X_1 - \frac{1}{3}\right), \\
0 \leq 2F_2(0) = & \; 2(A_2 + B_2) &= -\frac{1}{3}\alpha_1 Y_1^1 - \left(\frac{p_1}{c_1} - X_1 - \frac{1}{3}\right) + \frac{1}{3}\alpha_2 \frac{1}{R'_{22}}, \quad \text{and} \\
0 \leq 2F_3(0) = & \; 2(A_3 + B_3) &= -\frac{1}{3}\alpha_1 Y_1^1 - \left(\frac{p_1}{c_1} - X_1 - \frac{1}{3}\right) - \frac{1}{3}\alpha_2 \frac{1}{R'_{22}}
\end{aligned}$$

are collapsed upon inspection (i.e. $0 = F_1(0) = F_2(0) = F_3(0)$) and the solutions follow

$$B_\mu = -A_\mu = -\frac{p_\mu}{c_1}, \quad \alpha_1 = -\frac{3}{Y_1^1}\left(\frac{p_1}{c_1} - X_1 - \frac{1}{3}\right), \quad \text{and} \quad \alpha_2 = 0. \tag{3.35}$$

This conclusion has the rational consequence that the probability of observing two different field fluctuations at a point on the boundary between the two phases is zero.

With (3.31) and (3.32) the constraint $0 \leq F_\mu(|\vec{\tau}|)$ for any $0 \leq |\vec{\tau}|$ is reduced to to constraints on C_μ

$$\begin{aligned}
0 \;\; \leq \;\; & C_1 = \frac{2}{9}\left|\frac{\alpha_1}{R'_{11}}\right| + \frac{2}{9}\frac{\alpha_1}{R'_{11}} \leq 2\frac{p_1}{c_1} \\
\text{and} \;\; 0 \;\; \leq \;\; & C_2 = C_3 = \frac{1}{9}\left|\frac{\alpha_1}{R'_{11}}\right| \sqrt{\frac{9}{4}\left(\frac{R'_{11}}{R'_{22}}\right)^2 + 1} - \frac{1}{9}\frac{\alpha_1}{R'_{11}} \leq 1 - \frac{p_1}{c_1}
\end{aligned} \tag{3.36}$$

where $|\;\;|$ indicates the absolute value (i.e. $|x| = \sqrt{x^2}$ since $R'_{11} \neq 0$ and $R'_{22} \neq 0$).

Also it will also be helpful to write $\bar{R}_1$ as a function of α_1 to make relationships more compact later

$$\frac{\bar{R}_1}{R'_{11}} = \frac{1}{2}\left(1 - 3\frac{p_1}{c_1}\right) - 3c_2\frac{\alpha_1}{R'_{11}}. \tag{3.37}$$

Since $\alpha_2 = 0$ we have

$$\gamma_j(\vec{\tau}) = -\frac{1}{3}\alpha_1\left(\frac{\tau_j \tau_1}{|\vec{\tau}|} - \delta_{j1}\right) \mathrm{Exp}\left(-|\vec{\tau}|\right)$$

and

$$\begin{aligned}\gamma_{ij}(\vec{\tau}) &= \frac{1}{15}\alpha_1\alpha_1 \mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)\left(\left(\delta_{ij}+2\delta_{1i}\delta_{j1}\right)\right.\\ &\quad + \left(\left(1+\frac{1}{\left|\vec{\tau}\right|}\right)\frac{\tau_1\tau_1}{\left|\vec{\tau}\right|^2}-\frac{1}{\left|\vec{\tau}\right|}\right)\tau_i\tau_j - \frac{1}{\left|\vec{\tau}\right|}\left(\tau_1\tau_1\delta_{ij}+2\tau_j\tau_1\delta_{i1}+2\tau_i\tau_1\delta_{j1}\right))\end{aligned}$$

then

$$\begin{aligned}0 &\le \frac{s_{\mu 1}(\vec{\tau})+p_\mu c_2}{c_1c_2}\\ &= \left(\frac{1}{3}\alpha_1\left(Y_\mu^1-\left(Y_\mu^1\tau_1+Y_\mu^2\tau_2\right)\frac{\tau_1}{\left|\vec{\tau}\right|}\right)-\left(X_\mu+\frac{1}{N}\xi_\mu\right)\right)\mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)+\frac{p_\mu}{c_1}\end{aligned}$$

and

$$\begin{aligned}0 &\le \frac{p_{\mu\nu}(\vec{\tau})+p_\mu p_\nu}{c_1c_2}\\ &= \frac{1}{15}\left(\alpha_1\right)^2\mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)\left(Y_\mu^i\tau_iY_\nu^j\tau_j\left(\left(1+\frac{1}{\left|\vec{\tau}\right|}\right)\frac{\tau_1\tau_1}{\left|\vec{\tau}\right|^2}-\frac{1}{\left|\vec{\tau}\right|}\right)\right)\\ &+\frac{1}{15}\left(\alpha_1\right)^2\mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)\left(\left(Y_\mu^kY_\nu^k+2Y_\mu^1Y_\nu^1\right)-\frac{\tau_1}{\left|\vec{\tau}\right|}\left(\tau_1Y_\mu^kY_\nu^k+2\tau_jY_\mu^1Y_\nu^j+2\tau_iY_\mu^iY_\nu^1\right)\right)\\ &+\frac{1}{3}\alpha_1\mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)\left(\left(Y_\mu^j\frac{\tau_1\tau_j}{\left|\vec{\tau}\right|}-Y_\mu^1\right)\left(X_\nu+\frac{1}{N}\xi_\nu\right)+\left(X_\mu+\frac{1}{N}\xi_\mu\right)\left(Y_\nu^j\frac{\tau_1\tau_j}{\left|\vec{\tau}\right|}-Y_\nu^1\right)\right)\\ &+\left(X_\mu+\frac{1}{N}\xi_\mu\right)\left(X_\nu+\frac{1}{N}\xi_\nu\right)\mathrm{Exp}\left(-\left|\vec{\tau}\right|\right)+\frac{p_\mu p_\nu}{c_1c_2}.\end{aligned}$$

3.11 Composites with Statistically Continuous Material Characteristics

The material statistics developed thus far do not prevent the case of two points of observation within the first phase having two different field values at the limit they approach and coincide. In a statistical description an additional condition between the two point probability and the one point probability is introduced

$$\begin{aligned} f_{11}(\vec{u},0;\vec{u}') &= f_1(\vec{u})\,\delta(\vec{u}-\vec{u}'), \\ f_{12}(\vec{u},0;\vec{u}') &= 0, \\ \text{and}\quad f_{22}(\vec{u},0;\vec{u}') &= f_2(\vec{u})\,\delta(\vec{u}-\vec{u}') \quad \text{as } |\vec{r}|\to 0 \end{aligned} \tag{3.38}$$

and this is denoted as the condition of statistically continuous material characteristics, which holds for all materials.

Briefly consider the case of three vectors in the preceding section. On the boundary between the phases, (3.38) requires

$$0 = \frac{f_{12}(\vec{u};0,\vec{u}')}{c_1 c_2} = \sum_\mu \left(\frac{s_{\mu 1}(0)}{c_1 c_2} + \frac{p_\mu}{c_1}\right)\delta\left(\vec{u}-\vec{R}_\mu\right)\delta\left(\vec{u}'-\vec{Q}\right)$$

which was achieved due to the collapsed constraints at zero

$$0 = \frac{s_{\mu 1}(0)}{c_1 c_2} + \frac{p_\mu}{c_1} \quad \text{for each } \mu.$$

The new conditions which arise only from (3.38) are correlations within the first phase which ensure a point of observation cannot have two different field values which forces the probability of having an observed field value to be the probability of that same field value occurring:

$$\begin{aligned} \frac{p_{12}(0)+p_1p_2}{c_1c_2} &= \frac{p_{13}(0)+p_1p_3}{c_1c_2} = 0, \\ \frac{p_{23}(0)+p_2p_3}{c_1c_2} &= \frac{p_{32}(0)+p_3p_2}{c_1c_2} = 0, \\ \frac{p_{11}(0)+p_1p_1}{c_1c_2} &= \frac{p_1}{c_1c_2}, \\ \text{and}\quad \frac{p_{22}(0)+p_2p_2}{c_1c_2} &= \frac{p_{33}(0)+p_3p_3}{c_1c_2} = \frac{p_2}{c_1c_2}. \end{aligned} \tag{3.39}$$

Returning to general solutions independent of problem dimension and number of vectors in the first phase, from (3.20) we have

$$\begin{aligned} \frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2} &= Y_\mu^i \gamma_i(\vec{\tau}) - \left(X_\mu + \frac{1}{N}\xi_\mu\right) h_o(\vec{\tau}) \quad \text{and} \\ \frac{p_{\mu\nu}(\vec{\tau})}{c_1 c_2} &= \gamma_{ij}(\vec{\tau}) Y_\mu^i Y_\nu^j - Y_\mu^i \gamma_i(\vec{\tau}) \left(X_\nu + \frac{1}{N}\xi_\nu\right) - \left(X_\mu + \frac{1}{N}\xi_\mu\right) Y_\nu^j \gamma_j(\vec{\tau}) \\ &\quad + \left(X_\mu + \frac{1}{N}\xi_\mu\right)\left(X_\nu + \frac{1}{N}\xi_\nu\right) h_o(\vec{\tau}) \end{aligned} \tag{3.40}$$

where $\gamma_{ij}(\vec{\tau})$ and $\gamma_i(\vec{\tau})$ are unknown functions of $\hat{h}_o$ microstructure and Φ field fluctuation correlation

$$\begin{aligned} \gamma_{ij}(\vec{\tau}) &\equiv \int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right) \hat{h}_o\left(\vec{k}\right) \Phi^2\left(\vec{k}\right) k_i k_j dV_k \\ \text{and} \quad \gamma_i(\vec{\tau}) &\equiv \int \mathrm{Exp}\left(-i\,\vec{\tau}\cdot\vec{k}\right) \hat{h}_o\left(\vec{k}\right) \Phi\left(\vec{k}\right) k_i dV_k. \end{aligned}$$

Only constraints due to potentiality

$$X_\mu R'_{\mu i} = -\frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right), \quad X_\mu \xi_\mu = 0, \quad Y_\mu^i R'_{\mu j} = \delta_j^i, \quad \text{and} \quad Y_\mu^i \xi_\mu = 0 \tag{3.41}$$

as well as non-negativity of probabilities

$$0 \le \frac{p_\mu}{c_1} + \frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2} \quad \text{and} \quad 0 \le \frac{p_\mu p_\nu}{c_1 c_2} + \frac{p_{\mu\nu}(\vec{\tau})}{c_1 c_2} \tag{3.42}$$

remain.

The second constraint in (3.38) $f_{12}(\vec{u}, 0; \vec{u}') = 0$ requires

$$0 = \frac{p_\mu}{c_1} + \frac{s_{\mu 1}(0)}{c_1 c_2} \quad \text{for any} \quad \mu$$

giving a general solution of X_μ

$$X_\mu = Y_\mu^i \gamma_i(0) + \frac{1}{c_1} p_\mu - \frac{1}{N}\xi_\mu \tag{3.43}$$

and simplifications in (3.40) arise at the limit $|\vec{\tau}| \to 0$,

$$\frac{p_{\mu\nu}(0)}{c_1 c_2} = \frac{p_\mu p_\nu}{c_1 c_1} + \left(\gamma_{ij}(0) - \gamma_i(0)\gamma_j(0)\right) Y^i_\mu Y^j_\nu$$

since $h_o(0) = 1$.

Constraint $f_{11}(\vec{u}, 0; \vec{u}') = f_1(\vec{u})\,\delta(\vec{u} - \vec{u}')$ requires

$$\begin{aligned} \frac{p_\mu \xi_\nu}{c_1 c_2} &= \frac{p_\mu p_\nu}{c_1 c_2} + \frac{p_{\mu\nu}(0)}{c_1 c_2} = \frac{1}{c_2}\frac{p_\mu p_\nu}{c_1 c_1} + \left(\gamma_{ij}(0) - \gamma_i(0)\gamma_j(0)\right) Y^i_\mu Y^j_\nu \quad \text{if} \quad \mu = \nu \\ 0 &= \frac{p_\mu p_\nu}{c_1 c_2} + \frac{p_{\mu\nu}(0)}{c_1 c_2} = \frac{1}{c_2}\frac{p_\mu p_\nu}{c_1 c_1} + \left(\gamma_{ij}(0) - \gamma_i(0)\gamma_j(0)\right) Y^i_\mu Y^j_\nu \quad \text{if} \quad \mu \neq \nu \end{aligned}$$

or upon rearrangement

$$\begin{aligned} \gamma_{ij}(0)\, Y^i_\mu Y^j_\nu &= \gamma_i(0)\gamma_j(0)\, Y^i_\mu Y^j_\nu - \frac{1}{c_2}\frac{p_\mu p_\nu}{c_1 c_1} + \frac{p_\mu \xi_\nu}{c_1 c_2} \quad \text{if} \quad \mu = \nu \\ \gamma_{ij}(0)\, Y^i_\mu Y^j_\nu &= \gamma_i(0)\gamma_j(0)\, Y^i_\mu Y^j_\nu - \frac{1}{c_2}\frac{p_\mu p_\nu}{c_1 c_1} \qquad\qquad \text{if} \quad \mu \neq \nu. \end{aligned}$$

Contracting $\gamma_{ij}(0)\, Y^i_\mu Y^j_\nu$ with $R'_{\mu k} R'_{\nu l}$

$$\gamma_{ij}(0)\, Y^i_\mu Y^j_\nu R'_{\mu k} R'_{\nu l} = \gamma_i(0)\gamma_j(0)\, Y^i_\mu Y^j_\nu R'_{\mu k} R'_{\nu l} - \frac{1}{c_2}\frac{p_\mu}{c_1} R'_{\mu k} \frac{p_\nu}{c_1} R'_{\nu l} + \sum_\mu \frac{p_\mu}{c_1 c_2} R'_{\mu k} R'_{\mu l}$$

gives $\gamma_{ij}(0)$ since $Y^i_\mu R'_{\mu k} = \delta^i_k$

$$\gamma_{ij}(0) = \gamma_i(0)\gamma_j(0) - \frac{1}{c_2}\frac{p_\mu}{c_1} R'_{\mu i} \frac{p_\nu}{c_1} R'_{\nu j} + \sum_\mu \frac{p_\mu}{c_1 c_2} R'_{\mu i} R'_{\mu j}. \tag{3.44}$$

Without loss of generality we can specify the average field value in the first phase to be in the 1-dir and if we also introduce the requirement of symmetry of field fluctuations and probabilities about the applied field direction we have

$$p_\mu R'_{\mu i} = \bar{R}_i = 0 \quad \text{for } i \neq 1. \tag{3.45}$$

Combining the first term in potentiality condition (3.41) with the contraction of X_μ (3.43) and $R'_{\mu i}$

$$-\frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right) = X_\mu R'_{\mu i} = \gamma_j\left(0\right) Y^j_\mu R'_{\mu i} + \frac{p_\mu}{c_1} R'_{\mu i} - \frac{1}{N} R'_{\mu i}\xi_\mu$$

allows for the solution of $\gamma_i\left(0\right)$ since $R'_{\mu i}\xi_\mu = 0$ and $Y^j_\mu R'_{\mu i} = \delta^j_i$

$$\gamma_i\left(0\right) = -\frac{1}{c_2}\left(\frac{1}{c_1} p_\mu R'_{\mu i} + \bar{R}_i\right)$$

and with (3.45)

$$\gamma_1\left(0\right) = -\frac{1}{c_2}\left(\frac{1}{c_1} p_\mu R'_{\mu 1} + \bar{R}_1\right) \quad \text{and } \gamma_i\left(0\right) = 0 \text{ for } i \neq 1.$$

3.12 Statistically Continuous Material Characteristics: General Results

By specifying the condition of material characteristics being statistically continuous and introducing the requirement of symmetry of field fluctuations and probabilities about the applied field direction (assumed to be the 1−dir) we have reduced the entire problem to some simple constraints independent of dimensions and number of vectors. We know from the problem specification for large values of $|\vec{\tau}|$

$$\gamma_i\left(\vec{\tau}\right) = \gamma_{ij}\left(\vec{\tau}\right) = 0 \quad \text{as} \quad |\vec{\tau}| \rightarrow \infty$$

and conditions have been developed in the limit $|\vec{\tau}| \rightarrow 0$ from (3.38)

$$\begin{aligned}
\gamma_1\left(0\right) &= -\frac{1}{c_2}\left(\frac{1}{c_1} p_\mu R'_{\mu 1} + \bar{R}_1\right), \quad \gamma_i\left(0\right) = 0 \quad \text{for} \quad i \neq 1, \\
\gamma_{11}\left(0\right) &= \gamma_1\left(0\right)\gamma_1\left(0\right) - \frac{1}{c_2}\frac{p_\mu}{c_1} R'_{\mu 1}\frac{p_\nu}{c_1} R'_{\nu 1} + \sum_\mu \frac{p_\mu}{c_1 c_2} R'_{\mu 1} R'_{\mu 1} \\
\text{and} \quad \gamma_{ij}\left(0\right) &= \sum_\mu \frac{p_\mu}{c_1 c_2} R'_{\mu i} R'_{\mu j} \quad \text{for} \quad i \neq 1 \quad \text{or} \quad j \neq 1.
\end{aligned} \tag{3.46}$$

where

$$\gamma_{ij}(\vec{\tau}) \equiv \int \mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)\Phi^2\left(\vec{k}\right)k_ik_jdV_k$$
$$\text{and}\quad \gamma_i(\vec{\tau}) \equiv \int \mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)\Phi\left(\vec{k}\right)k_idV_k.$$

The first two conditions in potentiality constraints (3.41) are satisfied due to (3.45) and (3.43) leaving

$$Y_\mu^i R'_{\mu j} = \delta_j^i \quad\text{and}\quad \xi_\mu Y_\mu^i = 0.$$

We also have non-negativity of probabilities now just for positive $|\vec{\tau}|$

$$0 \le \frac{p_\mu}{c_1} + \frac{s_{\mu 1}(\vec{\tau})}{c_1c_2} \text{ and } 0 \le \frac{p_\mu p_\nu}{c_1c_2} + \frac{p_{\mu\nu}(\vec{\tau})}{c_1c_2} \text{ for } 0 < |\vec{\tau}|. \tag{3.47}$$

By introduction of the condition of material characteristics being statistically continuous, unknown parameters X_μ have been eliminated and functions $s_{\mu 1}(\vec{\tau})$ and $p_{\mu\nu}(\vec{\tau})$ are simplified

$$\begin{aligned}
\frac{s_{\mu 1}(\vec{\tau})}{c_1c_2} &= Y_\mu^i\gamma_i(\vec{\tau}) - \left(Y_\mu^1\gamma_1(0) + \frac{1}{c_1}p_\mu\right)h_o(\vec{\tau}) \quad\text{and} && (3.48)\\
\frac{p_{\mu\nu}(\vec{\tau})}{c_1c_2} &= \gamma_{ij}(\vec{\tau})Y_\mu^iY_\nu^j - Y_\mu^i\gamma_i(\vec{\tau})\left(Y_\mu^1\gamma_1(0) + \frac{1}{c_1}p_\mu\right) - \left(Y_\mu^1\gamma_1(0) + \frac{1}{c_1}p_\mu\right)Y_\nu^j\gamma_j(\vec{\tau})\\
&\quad + \left(Y_\mu^1\gamma_1(0) + \frac{1}{c_1}p_\mu\right)\left(Y_\mu^1\gamma_1(0) + \frac{1}{c_1}p_\mu\right)h_o(\vec{\tau}).
\end{aligned}$$

3.12.1 Statistically Continuous Material Characteristics: Two Dimensional Debye

Again taking a Debye material with

$$h_o(\vec{\tau}) = \mathrm{Exp}[-|\vec{\tau}|] \quad\text{and}\quad \hat{h}_o\left(\vec{k}\right) = \frac{2\pi}{\left(1+\left|\vec{k}\right|^2\right)^{3/2}}$$

with

$$\Phi\left(\vec{k}\right) = \frac{\alpha_i \; k_i}{1 + \left|\vec{k}\right|^2}$$

and considering the two dimensional case, $i, j = 1..2$, $\gamma_i\left(\vec{\tau}\right)$ and $\gamma_{ij}\left(\vec{\tau}\right)$ were simplified at the limit $\left|\vec{\tau}\right| \rightarrow 0$

$$\gamma_i\left(0\right) = \frac{1}{3}\alpha_i \text{ and } \; \gamma_{ij}\left(0\right) = \frac{1}{15}\alpha_m\alpha_n\left(\delta_{ij}\delta_{mn} + \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}\right). \tag{3.49}$$

With the two terms $\gamma_i\left(0\right)$ in (3.46) we can solve for α_i

$$\alpha_1 = -\frac{3}{c_2}\left(\frac{1}{c_1}p_\mu R'_{\mu 1} + \bar{R}_1\right) \text{ and } \; \alpha_i = 0 \; \text{ for } \; i \neq 1$$

and α_i known, $\gamma_{ij}\left(0\right)$ in (3.49) and (3.46) are

$$\begin{aligned}
\frac{1}{5}\alpha_1\alpha_1 &= \gamma_{11}\left(0\right) = \frac{1}{9}\alpha_1\alpha_1 - \frac{1}{c_2}\frac{p_\mu}{c_1}R'_{\mu 1}\frac{p_\nu}{c_1}R'_{\nu 1} + \sum_\mu \frac{p_\mu}{c_1 c_2}R'_{\mu 1}R'_{\mu 1} \\
0 &= \gamma_{12}\left(0\right) = \gamma_{21}\left(0\right) = \sum_\mu \frac{p_\mu}{c_1 c_2}R'_{\mu 1}R'_{\mu 2} \\
0 &= \gamma_{21}\left(0\right) = \gamma_{12}\left(0\right) \\
\frac{1}{15}\alpha_1\alpha_1 &= \gamma_{22}\left(0\right) = \sum_\mu \frac{p_\mu}{c_1 c_2}R'_{\mu 2}R'_{\mu 2}.
\end{aligned}$$

Summary

Upon simplifications the problem is reduced to p_μ, $R'_{\mu i}$, and Y^i_μ subject to the constraints

$$\begin{aligned}
0 &\leq c_1 = \sum_\mu p_\mu \leq 1, \quad c_2 = 1 - c_1, \quad 0 < p_\mu \; \text{ for each } \mu, \qquad (3.50) \\
\xi_\mu R'_{\mu i} &= 0, \; p_\mu R'_{\mu i} = \bar{R}_i = 0 \; \text{ for } i \neq 1, \; Y^i_\mu R'_{\mu j} = \delta^i_j, \; \xi_\mu Y^i_\mu = 0, \\
\sum_\mu p_\mu R'_{\mu 2}R'_{\mu 2} &= \frac{3}{4}\left(\sum_\mu p_\mu R'_{\mu 1}R'_{\mu 1} - \frac{1}{c_1}p_\mu R'_{\mu 1}p_\nu R'_{\nu 1}\right), \\
\sum_\mu p_\mu R'_{\mu 2}R'_{\mu 2} &= \frac{3}{5}\frac{c_1}{c_2}\left(\frac{1}{c_1}p_\mu R'_{\mu 1} + \bar{R}_1\right)^2, \; \text{ and } \; 0 = \sum_\mu p_\mu R'_{\mu 1}R'_{\mu 2}
\end{aligned}$$

where non-negativity of probabilities for $0 \leq f_{12}\left(\vec{u}; \vec{\tau}, \vec{u}'\right)$ since $0 = F_\mu\left(0\right)$ are reduced to

(3.32)

$$0 \leq C_\mu = \frac{1}{3}\sqrt{\left(\alpha_1 Y_\mu^2\right)^2 + \left(\alpha_1 Y_\mu^1\right)^2} + \frac{1}{3}\alpha_1 Y_\mu^1 \leq 2\frac{p_\mu}{c_1} \text{ and} \tag{3.51}$$

$$0 \leq \frac{p_\mu p_\nu}{c_1 c_2} + \frac{p_{\mu\nu}\left(\vec{\tau}\right)}{c_1 c_2} \text{ for } 0 < \left|\vec{\tau}\right|.$$

CHAPTER 4 VARIATIONAL PRINCIPLE FOR HOMOGENIZATION IN PROBABILISTIC TERMS

With the variational principle for homogenization in probabilistic terms a composite with isotropic phases we can find the effective properties a_{eff} as well as field fluctuations and their probabilities through minimization under constraints (3.5) through (3.10) . For the two dimensional case

$$\begin{aligned} a_{\text{eff}} &= \text{Min}_{(3.11),(3.12),(3.22)} \ \frac{1}{|\vec{v}|^2} a_1 \sum_{\mu} p_\mu \left((v_1 + R_{\mu 1})^2 + (v_2 + R_{\mu 2})^2 \right) \\ &\quad + \frac{1}{|\vec{v}|^2} a_2 \sum_{\nu} q_\nu \left((v_1 + Q_{\nu 1})^2 + (v_2 + Q_{\nu 2})^2 \right) \end{aligned}$$

where $\vec{v}$ is the applied field and a_1, a_2 phase conductivities. It is clear that $\vec{R}_\mu$ and $\vec{Q}_1$ are proportional to $|\vec{v}|$ and without loss of generality we take $|\vec{v}|$ to be of unit intensity in the $1-$direction. After dividing through by a_1 and using introduced notations, we have

$$\frac{a_{\text{eff}}}{a_1} = \text{Min}_{(3.11),(3.12),(3.22)} \ \sum_{\mu} p_\mu \left(\left(1 + R'_{\mu 1} + \xi_\mu \bar{R}_1\right)^2 + \left(R'_{\mu 2}\right)^2 \right) + \frac{a_2}{a_1} c_2 \left(1 - \frac{1}{c_2} \left(p_\mu R'_{\mu 1} + c_1 \bar{R}_1 \right) \right)^2 \tag{4.1}$$

for two dimensional problems.

4.1 Three Field Fluctuations: an Approximate Solution

Iso contours of energy along with the regions where $0 \leq f_{12}(\vec{u}; \vec{\tau}, \vec{u}')$ and $0 \leq f_{11}(\vec{u}; 0, \vec{u}')$ are satisfied are shown as a function of R'_{11} and R'_{22} in Figure 4.1 for an example case of $p_1/c_1 = 3/7$ and $a_1/a_2 = 1/10$. Similar results are found for other p_1/c_1 and a_1/a_2We seek low values of energy, and it is observed that energy decays for larger values of α_1/R'_{11} up to the limit due to the upper limit due to the first constraint in (3.36) . With this fact, let us take α_1/R'_{11} as proportional to p_1/c_1 :

$$\frac{\alpha_1}{R'_{11}} = Z \frac{p_1}{c_1}.$$

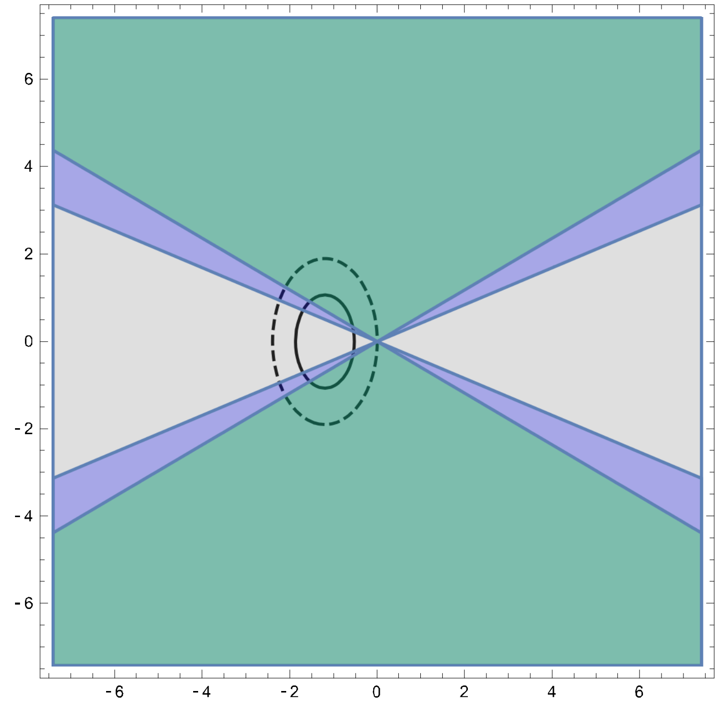

Figure 4.1: Typical topology of $0 \leq f_{12}(\vec{u};\vec{\tau},\vec{u}')$ and $0 \leq f_{11}(\vec{u};0,\vec{u}')$ in R_{11} R_{22} space. Regions $0 \leq f_{12}(\vec{u};\vec{\tau},\vec{u}')$ (blue) and $0 \leq f_{11}(\vec{u};0,\vec{u}')$ (green) shown with iso contours of energy (black).

where $Z \leq \frac{9}{2}$ from (3.36). This proportionality has the consequence of mean field fluctuation $\bar{R}_1$ being simply proportional to the first phase field fluctuation R'_{11}

$$X_1 = \left(1 + \frac{2}{9}Z\right)\frac{p_1}{c_1} - \frac{1}{3} \text{ and then } \frac{\bar{R}_1}{R'_{11}} = \frac{1}{2}(1 - 3\frac{p_1}{c_1}) - \frac{1}{3}Z\frac{p_1}{c_1}(1 - c_1).$$

Taking the largest admissible value, $Z = \frac{9}{2}$ then $\alpha_1 = \frac{9}{2}\frac{p_1}{c_1}R'_{11}$ and constraints $p_{\mu\nu}$ at $\tau_\mu = 0$ can be written compactly

$$\begin{aligned}
0 &\leq \frac{p_{11}(0) + p_1 p_1}{c_1 c_2} = 3\left(\frac{p_1}{c_1}\right)^2, \qquad (4.2)\\
0 &\leq \frac{p_{12}(0) + p_1 p_2}{c_1 c_2} = \frac{p_1}{c_1}\left(1 - \frac{7}{5}\frac{p_1}{c_1}\right),\\
0 &\leq \frac{p_{22}(0) + p_2 p_2}{c_1 c_2} = \frac{1}{10}\left(2 + \frac{27}{8}\left(\frac{R'_{11}}{R'_{22}}\right)^2\right)\left(\frac{p_1}{c_1}\right)^2 + \frac{1}{2}\left(\frac{p_1}{c_1} - 1\right)^2\\
\text{and } 0 &\leq \frac{p_{23}(0) + p_2 p_3}{c_1 c_2} = \frac{1}{10}\left(2 - \frac{27}{8}\left(\frac{R'_{11}}{R'_{22}}\right)^2\right)\left(\frac{p_1}{c_1}\right)^2 + \frac{1}{2}\left(\frac{p_1}{c_1} - 1\right)^2,
\end{aligned}$$

where

$$p_{12}(0) = p_{13}(0) = p_{21}(0) = p_{31}(0), \quad p_{22}(0) = p_{33}(0), \quad \text{and} \quad p_{23}(0) = p_{32}(0).$$

Since the constraints in Figure 4.1 form straight lines in R'_{11} and R'_{22} space and levels of constant energy are ellipses, the solution for a_{eff} will then inherently be found at the tangent point intersecting straight lines of constraints and ellipses of energy. To find this point, introduce a constant of proportionality Ω between components R'_{11} and R'_{22}

$$R'_{22} = \Omega R'_{11}$$

and then constraints $0 \leq F_2(|\vec{r}|)$ and $0 \leq F_3(|\vec{r}|)$ can be written compactly

$$\sqrt{\frac{9}{4}\left(\frac{1}{\Omega}\right)^2 + 1} \leq 2\frac{c_1}{p_1} - 1 \quad \text{alternatively} \quad |\Omega| \leq \frac{3}{4}\frac{1}{\sqrt{\frac{c_1}{p_1}\left(\frac{c_1}{p_1} - 1\right)}}. \tag{4.3}$$

The constraint line normals are in the $\{-\omega, \Omega\omega\}$ direction

$$\frac{\partial}{\partial R'_{22}} a_{\text{eff}} = -\omega \quad \text{and} \quad \frac{\partial}{\partial R'_{11}} a_{\text{eff}} = \Omega\omega,$$

where here, ω is an unknown constant of proportionality between the normal to the constraint line and the gradient of energy. With these two equations R'_{11} can be determined

$$R'_{11} = \frac{6(a_2 - a_1) - 6(a_2 - a_1)c_1}{4a_1\Omega^2\left(1 - \frac{c_1}{p_1}\right) + 9a_1(p_1 - 1) + 9(a_1 - a_2)p_1 - 9(a_1 - a_2)c_1 p_1}.$$

We wish to determine the values of p_1, R'_{11}, and R'_{22} which minimize energy for a given problem definition c_1, a_1, and a_2 under the constraints for positiveness of probabilities with the problem reduced to determination of Ω and p_1. The general topology of the problem is shown in Figure 4.2, and it is observed that energy is minimized for large p_1 and small Ω

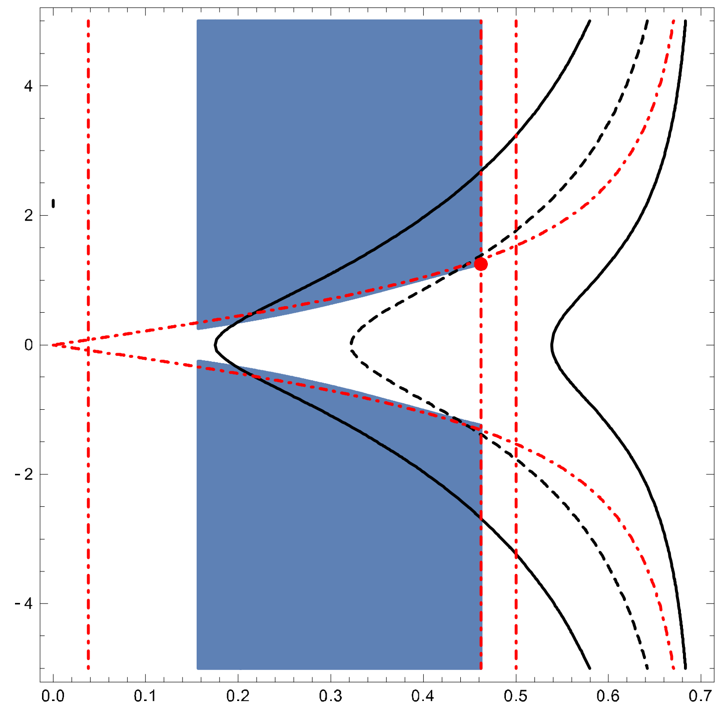

Figure 4.2: Typical topology of $0 \leq f_{11}(\vec{u}; 0, \vec{u}')$ and $0 \leq f_{12}(\vec{u}; \vec{\tau}, \vec{u}')$ in Ω p_1 space with isocontours of energy (black)

The constraints relating to correlations between the first and second phases ($0 \leq F_1(|\vec{\tau}|)$, $0 \leq F_2(|\vec{\tau}|)$, and $0 \leq F_3(|\vec{\tau}|)$) as well as $0 \leq f_{11}(\vec{u}; 0, \vec{u}')$ have been reduced to very simple conditions, only $0 \leq f_{11}(\vec{u}; \vec{\tau}, \vec{u}')$ for $0 < |\vec{\tau}|$ could not be simplified. First minimization (4.1) for all constraints except $0 \leq f_{11}(\vec{u}; \vec{\tau}, \vec{u}')$ for $0 < |\vec{\tau}|$ was conducted and yielded a very simple result

$$\frac{p_1}{c_1} = \frac{5}{7}, \;\; \Omega = \frac{5}{4}\sqrt{\frac{3}{2}}, a_{\text{eff}} = a_1 \frac{59 a_1 c_1 + 119 a_2 c_2}{60 a_2 c_1 c_2 + a_1 \left(59 + 60 \left(c_2\right)^2\right)};$$

this solution at $\vec{\tau} = 0$ does have the desired feature $p_{12}(0) + p_1 p_2 = p_{23}(0) + p_2 p_3 = 0$, however it leads to negative probabilities for nonzero $\vec{\tau}$.

Considering the case of considering all constraints directly to study the admissible space was found to be at the time not possible due to the lengthy minimization procedure required on each considered configuration of $p_{\mu\nu}(\vec{\tau})$ over $\vec{\tau}$. For this reason, instead the constraints on $f_{11}(\vec{u};\vec{\tau},\vec{u}')$ at $|\vec{\tau}|=0$ were strengthened by a small value δ

$$0 \leq \delta \leq \frac{p_{\mu\nu}(0)+p_\mu p_\nu}{c_1 c_2}$$

which was sufficiently large to ensure for all $\vec{\tau}$

$$0 \leq \frac{p_{\mu\nu}(\vec{\tau})+p_\mu p_\nu}{c_1 c_2}.$$

The case of $\delta = 0.0664$ was found to be suitable and led to the approximate solution which satisfies all constraints on probabilities but $p_{12}(0)+p_1p_2 \neq 0$ and $p_{23}(0)+p_2p_3 \neq 0$:

$$\frac{p_1}{c_1} = \frac{5}{\sqrt{61}},\ \Omega = \frac{5}{4},\quad \text{and} \quad a_{\text{eff}} = a_1 \frac{\left(125+11\sqrt{61}\right)a_1c_1 + \left(305+11\sqrt{61}\right)a_2c_2}{180a_2c_1c_2 + a_1\left(125+11\sqrt{61}+180\left(c_2\right)^2\right)}.$$

Thus far it has been assumed that there are three points of concentration the first phase and one point in the second phase. However this was for clarity, the entire procedure is identical if the situation was reversed. Therefore a_1 and c_1 can be swapped with a_2 and c_2, vice-versa, giving a second result for effective coefficient

$$a_{\text{eff}} = a_2 \frac{\left(125+11\sqrt{61}\right)a_2c_2 + \left(305+11\sqrt{61}\right)a_1c_1}{180a_1c_1c_2 + a_2\left(125+11\sqrt{61}+180\left(c_1\right)^2\right)}.$$

These new results are compared against the Voight, Reuss, and Hashin-Shtrikman bounds in Figure 4.3.The lower of the two predictions fall within the Hashin-Shtrikman bounds for nearly all combinations of conductivity and phase concentrations. The essential difference is that this new result the field is not homogenous in each phase. This is shown in Figure 4.4 and the fluctuations can be computed for any case giving an improved estimate of the orthogonal component of heat flux for a particulate composite.

While this approximate solution ensures non-negative probabilities, it has the very undesirable and unrealistic feature of the two point probability not collapsing to the one point probability. In the next section the case of a composite with statistically continuous material

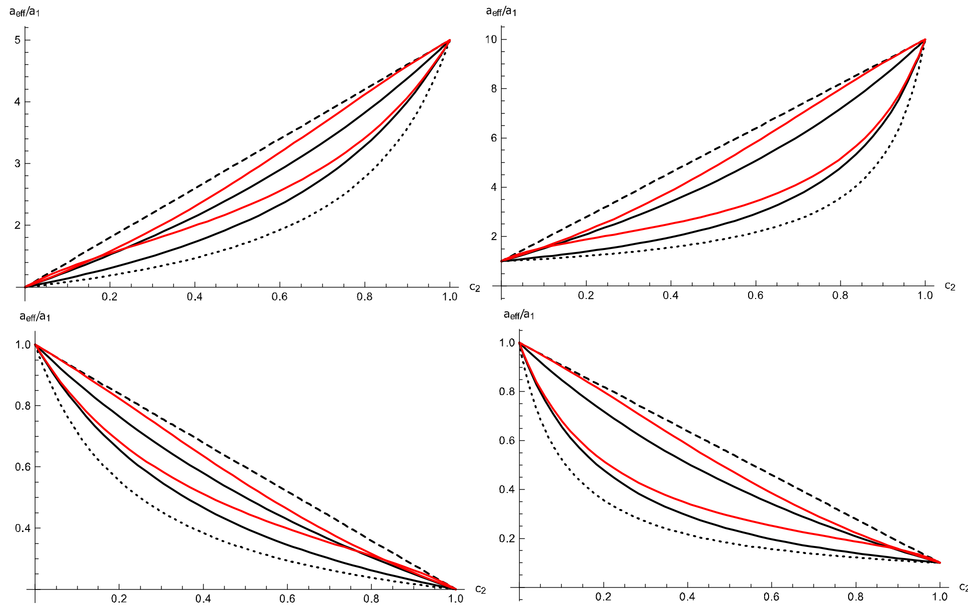

Figure 4.3: a_{eff} (solid, grey) with Reuss (dashed), Voight, (dotted), and Hashin-Shtrikman (solid, black) for four levels of contrast ($a_2/a_1 = 1/10$, top left), ($a_2/a_1 = 1/5$, top right), ($a_2/a_1 = 5$, bottom left), and ($a_2/a_1 = 10$, bottom right)

characteristics is considered.

4.2 Statistically Continuous Material Characteristics

Let us again consider the case when $N = 3$ and setting conditions of symmetry as previously shown

$$p_2 = p_3,\ R'_{12} = 0,\ R'_{31} = R'_{21},\ R'_{32} = -R'_{22}$$

but this time using the simplifications given in (3.50) and (3.51).

Solution of p_3 is found from $c_1 = \sum\limits_{\mu} p_\mu$ and R'_{21} from $\xi_\mu R'_{\mu 1} = 0$. Then from $\xi_\mu Y^i_\mu = 0$, Y^1_3 and Y^2_3 are found, and with $Y^i_\mu R'_{\mu j} = \delta^i_j$ solutions to Y^1_1, Y^1_2, Y^2_1 and Y^2_2 arise. With $\sum\limits_{\mu} p_\mu R'_{\mu 2} R'_{\mu 2} = \frac{3}{4}\left(\sum\limits_{\mu} p_\mu R'_{\mu 1} R'_{\mu 1} - \frac{1}{c_1} p_\mu R'_{\mu 1} p_\nu R'_{\nu 1}\right)$, the solution to p_3 is found.

We are strictly concerned with non degenerated cases, which are a homogenous material and the degeneracy of the number of fluctuations. These constraints made explicit, degeneracy of the solution is excluded by

$$c_1 \neq 1, \quad c_2 \neq 1, \quad R'_{11} \neq 0, \quad \text{and} \quad R'_{22} \neq 0. \tag{4.4}$$

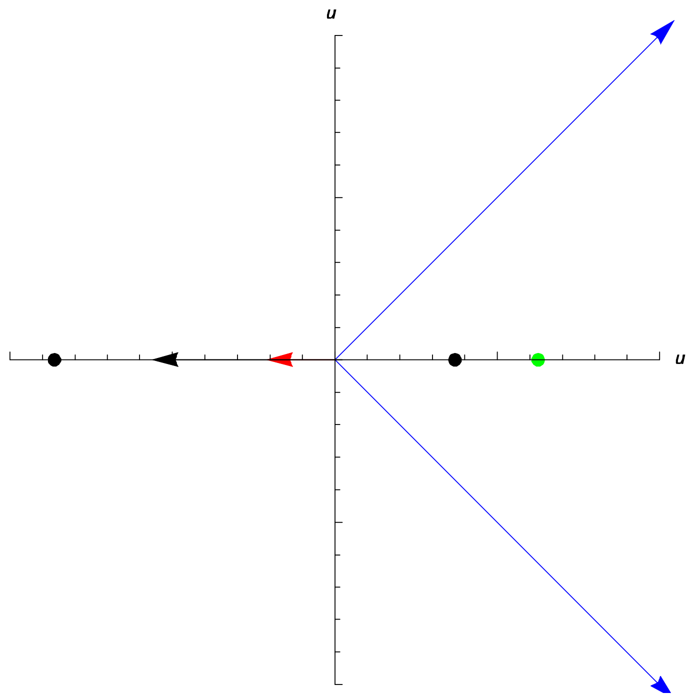

Figure 4.4: Points of concentration of fluctutations for the 3 vector approximation. The blue and red vectors are points of concentration in the first phase, and the black vector the point of concentration in the second phase. Black dots correspond to the Reuss solution.

Without a loss of generality use the notation previously introduced

$$R'_{22} = \Omega R'_{11}$$

and due to the symmetry of the problem, again without loss of generality, let

$$0 < R'_{22} = \Omega R'_{11}. \tag{4.5}$$

Then, the condition $c_1 = \sum_\mu p_\mu$ is satisfied only in the case

$$-\frac{3\sqrt{3}}{4} < \Omega < \frac{3\sqrt{3}}{4}. \tag{4.6}$$

Finally, $\bar{R}_1$ is found from $\sum_\mu p_\mu R'_{\mu 2} R'_{\mu 2} = \frac{3}{5} \frac{c_1}{c_2} \left(\frac{1}{c_1} p_\mu R'_{\mu 1} + \bar{R}_1 \right)^2$ and it has two possible values.

where $\bar{R}_1$ has two possible values. Denote these two cases as the larger $\bar{R}_1$

$$\begin{aligned} \bar{R}_1 &= \frac{1}{2}R'_{11}\left(1-\frac{16}{9}\Omega^2\right)+\sqrt{c_2}\sqrt{\frac{5}{3}\left(R'_{22}\right)^2\left(1-\frac{16}{27}\Omega^2\right)} \\ \text{where}\ \frac{\alpha_1}{R'_{11}} &= -\Omega\frac{3}{\sqrt{c_2}}\sqrt{\frac{5}{3}\left(1-\frac{16}{27}\Omega^2\right)} \end{aligned} \tag{4.7}$$

and the smaller $\bar{R}_1$

$$\begin{aligned} \bar{R}_1 &= \frac{1}{2}R'_{11}\left(1-\frac{16}{9}\Omega^2\right)-\sqrt{c_2}\sqrt{\frac{5}{3}\left(R'_{22}\right)^2\left(1-\frac{16}{27}\Omega^2\right)} \\ \text{where}\ \frac{\alpha_1}{R'_{11}} &= \Omega\frac{3}{\sqrt{c_2}}\sqrt{\frac{5}{3}\left(1-\frac{16}{27}\Omega^2\right)} \end{aligned} \tag{4.8}$$

For a given problem definition with $0 < c_1 < 1$ and $c_2 = 1 - c_1$, the problem of three fluctuations is reduced to unknowns Ω and R'_{11}

$$\begin{aligned} p_1 &= c_1\frac{16}{27}\Omega^2,\ \ p_3 = \frac{1}{2}\left(c_1 - p_1\right),\ \ R'_{21} = -\frac{1}{2}R'_{11}, \\ Y_1^1 &= \frac{2}{3}\frac{1}{R'_{11}},\ \ Y_1^2 = 0,\ \ Y_2^1 = Y_3^1 = -\frac{1}{2}Y_1^1,\ \ \text{and}\ \ Y_2^2 = -Y_3^2 = \frac{1}{2}\frac{1}{R'_{22}} \end{aligned} \tag{4.9}$$

where $\bar{R}_1$ can take one of two possible values, (4.7) or (4.8), some simple constraints remain (4.4), (4.5), and (4.6) and let us now make simplifications to non negativity of probabilities for positive $|\vec{\tau}|$

$$0 \le \frac{p_\mu}{c_1} + \frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2}\ \text{and}\ \ 0 \le \frac{p_\mu p_\nu}{c_1 c_2} + \frac{p_{\mu\nu}(\vec{\tau})}{c_1 c_2}\ \ \text{for}\ \ 0 < |\vec{\tau}|\,.$$

As previously shown, the constraints for $0 \le f_{12}(\vec{u};\vec{\tau},\vec{u}')$ are

$$0 \le \frac{p_\mu}{c_1} + \frac{s_{\mu 1}(\vec{\tau})}{c_1 c_2}$$

and since they are collapsed at $|\vec{\tau}| = 0$ ($0 = f_{12}(\vec{u};0,\vec{u}')$) the derivative of probabilities with respect to $|\vec{\tau}|$ must be non-negative requiring $0 \le \partial_{|\vec{\tau}|} f_{12}(\vec{u};|\vec{\tau}|,\vec{u}')$ as $|\vec{\tau}| \to 0$. This condition was previously shown to reduce to the simple conditions

$$\begin{aligned} 0 &\leq \frac{2}{9}\left|\frac{\alpha_1}{R'_{11}}\right| + \frac{2}{9}\frac{\alpha_1}{R'_{11}} \leq 2\frac{p_1}{c_1} \\ \text{and } 0 &\leq \frac{1}{9}\left|\frac{\alpha_1}{R'_{11}}\right|\sqrt{\frac{9}{4}\frac{1}{\Omega^2}+1} - \frac{1}{9}\frac{\alpha_1}{R'_{11}} \leq 2\frac{p_2}{c_1}. \end{aligned} \tag{4.10}$$

Similarly for $0 \leq f_{11}(\vec{u}; 0, \vec{u}')$ if $\vec{u} \neq \vec{u}'$ the constraints are again collapsed

$$\frac{p_{12}(0) + p_1 p_2}{c_1 c_2} = \frac{p_{13}(0) + p_1 p_3}{c_1 c_2} = 0 \text{ and } \frac{p_{23}(0) + p_2 p_3}{c_1 c_2} = \frac{p_{32}(0) + p_3 p_2}{c_1 c_2} = 0$$

requiring $0 \leq \partial_{|\vec{\tau}|} f_{11}(\vec{u}; |\vec{\tau}|, \vec{u}')$ if $\vec{u} \neq \vec{u}'$ as $|\vec{\tau}| \to 0$ which are

$$0 \leq \frac{\partial}{\partial |\vec{\tau}|} p_{12}(|\vec{\tau}|) \text{ and } 0 \leq \frac{\partial}{\partial |\vec{\tau}|} p_{23}(|\vec{\tau}|) \text{ as } |\vec{\tau}| \to 0. \tag{4.11}$$

Both (4.10) and (4.11) lack dependence on the magnitude of R'_{11}, they only depend on the sign. Therefore we can study admissible space easily.

4.3 Admissible values of c_2 and Ω from $0 \leq f_{12}(\vec{u}; |\vec{\tau}|, \vec{u}')$

Let us first consider the admissible space of (4.10). Since we have two admissible values of $\bar{R}_1$, and R'_{11} can be either positive or negative, there are four cases to consider. These are shown graphically in Figure 4.5 with the top row corresponding to the larger value of $\bar{R}_1$, lower the smaller value of $\bar{R}_1$, the left set the case of positive R'_{11} and the right negative R'_{11}.

Negative α_1/R'_{11}

As evident from Figure 4.5, the case of large $\bar{R}_1$ with positive R'_{11} has the same admissible space for $0 \leq f_{12}(\vec{u}; \vec{\tau}, \vec{u}')$ as small $\bar{R}_1$ with negative R'_{11} and vice versa on the sign of R'_{11}. Reviewing both cases of $\bar{R}_1$ (4.7) and (4.8) with the constraint $0 < \Omega R'_{11}$ (4.5) both of these cases have the same result

$$\frac{\alpha_1}{R'_{11}} = -\frac{3}{\sqrt{c_2}}|\Omega|\sqrt{\frac{5}{3}\left(1 - \frac{16}{27}\Omega^2\right)}.$$

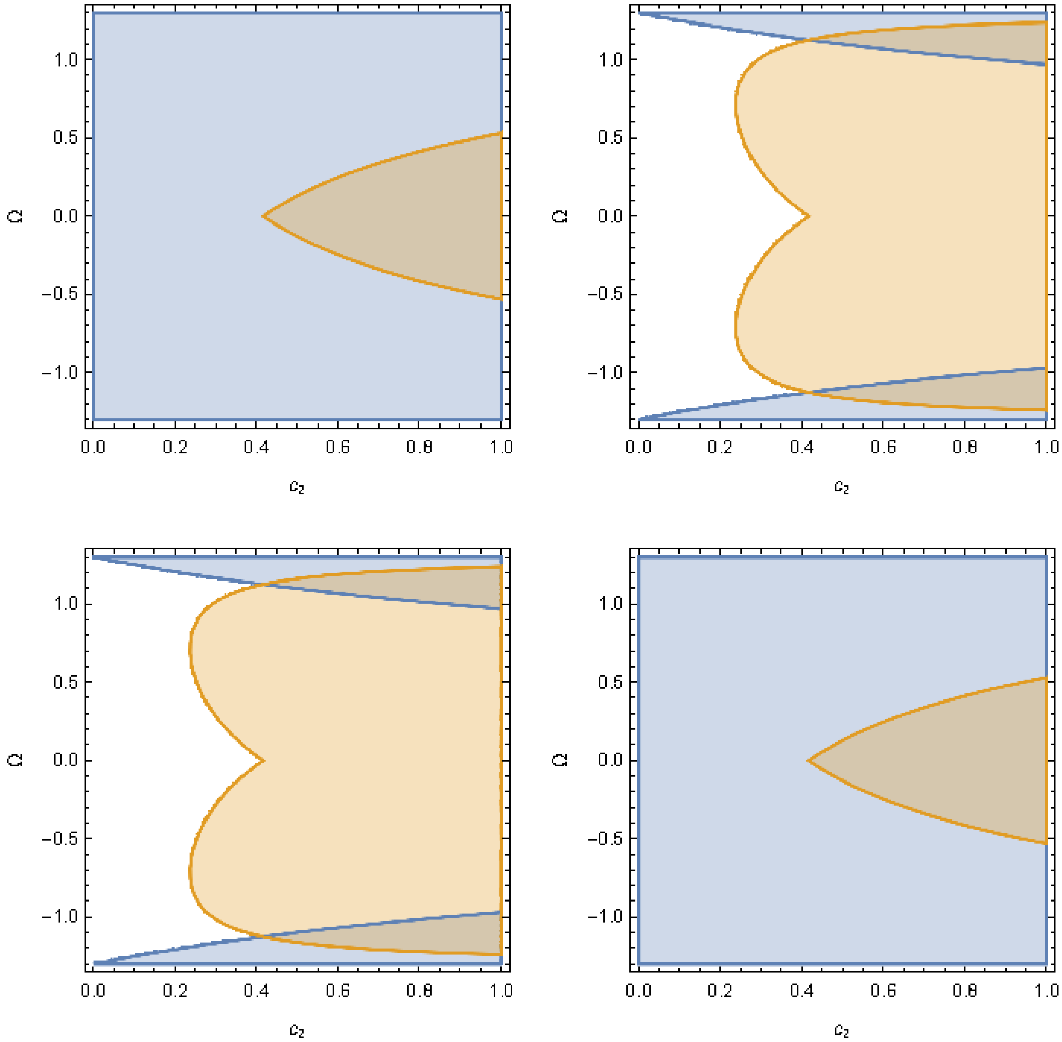

Figure 4.5: Admissible $0 \leq f_{12}(\vec{u};\vec{\tau},\vec{u}')$ in c_2 Ω with $0 \leq F_1(0)$ (blue) and $0 \leq F_2(0) = F_3(0)$ (orange) . Upper row is the larger value of $\bar{R}_1$ and lower the smaller. Left is the case of positive R'_{11} and the right negative R'_{11}.

The first constraint in (4.10) is satisfied upon inspection since α_1/R'_{11} is negative.

The second constraint in (4.10) provides a minimum value of c_2 of $5/12$, however this case occurs at $\Omega = 0$, therefore the minimum admissible range of c_2 is reduced to

$$\frac{5}{12} < c_2 < 1.$$

The second constraint further reduces the admissible space to

$$\frac{45}{108-64\Omega^2}\left(1+\frac{8}{9}+\frac{4}{3}\left|\Omega\right|\sqrt{1-\frac{4}{9}\Omega^2}\right)<c_2<1.$$

The lower bound is unchanged $c_2 = 5/12$ for the degenerate case $\Omega = 0$.

Positive α_1/R'_{11}

Similarly, as evident from Figure 4.5, the case of large $\bar{R}_1$ with negative R'_{11} has the same admissible space for $0 \leq f_{12}\left(\vec{u};\vec{r},\vec{u}'\right)$ as small $\bar{R}_1$ with positive R'_{11} and vice versa on the sign of R'_{11}, here

$$\frac{\alpha_1}{R'_{11}}=\frac{3}{\sqrt{c_2}}\left|\Omega\right|\sqrt{\frac{5}{3}\left(1-\frac{16}{27}\Omega^2\right)}.$$

The first constraint in (4.10) provides a lower limit of the magnitude of Ω

$$\frac{\sqrt{15}}{4}<\left|\Omega\right|<\frac{3\sqrt{3}}{4}$$

and the second constraint reduces the maximum magnitude of Ω, leaving

$$\frac{\sqrt{15}}{4}<\left|\Omega\right|<\frac{7}{4\sqrt{2}}.$$

As seen in Figure 4.5 the constraint which is limiting depends on the value of $\left|\Omega\right|$. The first constraint in (4.10) shown in blue limits the lower value of $\left|\Omega\right|$ and the second the upper value of $\left|\Omega\right|$, with the intersection at $\left|\Omega\right| = 9/8$. The solution of the admissible space of $0 \leq f_{12}\left(\vec{u};\vec{r},\vec{u}'\right)$ for positive α_1/R'_{11} is

$$\begin{aligned}
\frac{135}{64\Omega^2}-\frac{5}{4} &\leq c_2 \text{ for } \frac{9}{8}\leq\left|\Omega\right|<\frac{7}{4\sqrt{2}} \text{ and}\\
5\frac{9+8\Omega^2}{108-64\Omega^2}-5\frac{\sqrt{\Omega^2\left(9+4\Omega^2\right)}}{27-16\Omega^2} &\leq c_2 \text{ for } \frac{\sqrt{15}}{4}<\left|\Omega\right|\leq\frac{9}{8}.
\end{aligned}$$

Note at at $\left|\Omega\right| = 9/8$ provides the same minimum value of c_2, $5/12$, therefore values of c_2 less than $5/12$ can be excluded from any further consideration.

4.4 Admissible values of c_2 and Ω from $0 \leq f_{11}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$

The constraints $0 \leq \partial_{|\vec{\tau}|} f_{11}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$ at $\left|\vec{\tau}\right| = 0$ if $\vec{u} \neq \vec{u}'$ also have a dependence on the sign of α_1/R'_{11} only. However, unlike the previous case, no analytical simplifications could be found. For this reason, the admissible space was studied by computing contour plots of the value of $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ and $\partial_{|\vec{\tau}|} p_{23}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$, both of which must be non-negative.

For the case of negative α_1/R'_{11}, fitted contours of the values of $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ and $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ are shown in Figure 4.6 with the outline of $0 \leq f_{12}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$ shown in black.

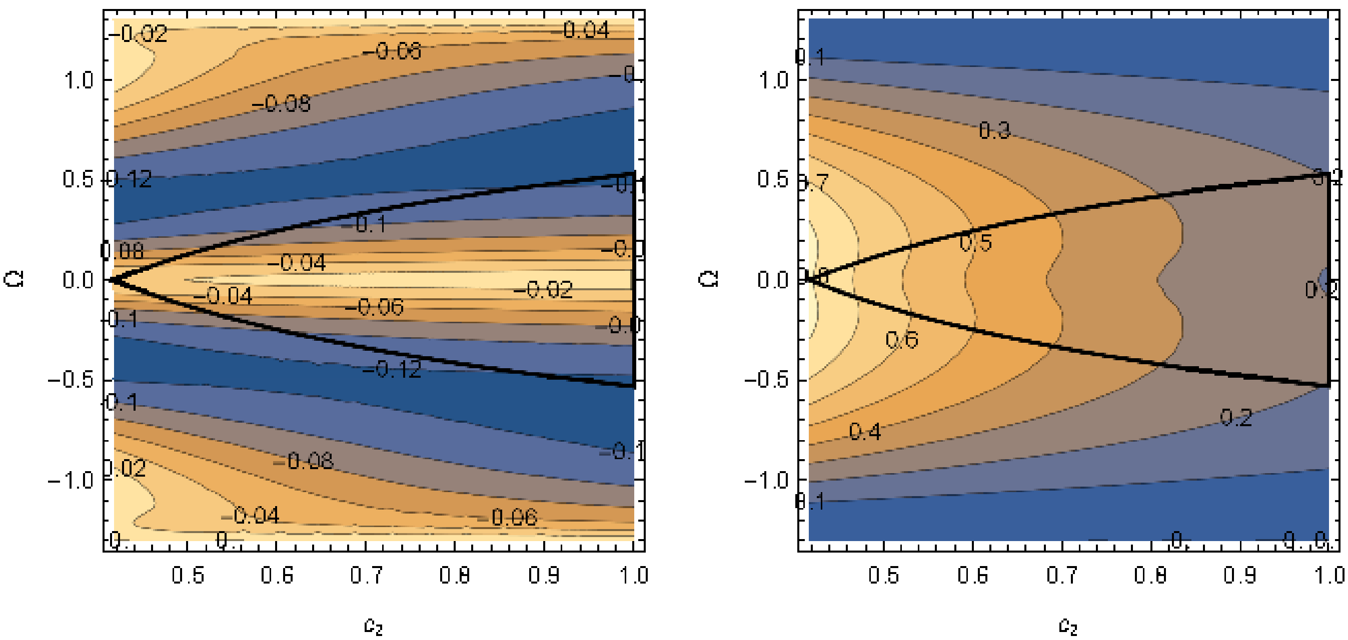

Figure 4.6: Contours of $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ (left) and $\partial_{|\vec{\tau}|} p_{23}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ (right) for the case of negative α_1/R'_{11}. The extent of the admissible space due to $0 \leq f_{12}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$ is outlined in black.

Since both $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ and $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ must be non-negative, but this does not hold for any combination of c_2 and $\Omega = 0$ within the admissible space of $0 \leq f_{12}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$, it has bene shown that negative α_1/R'_{11} will always result in non physical negative probabilities. It is excluded from further consideration.

Next, for the case of positive α_1/R'_{11}, fitted contours of the values of $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ and $\partial_{|\vec{\tau}|} p_{12}\left(\left|\vec{\tau}\right|\right)$ at $\left|\vec{\tau}\right| = 0$ are shown in Figure 4.7 with the outline of $0 \leq f_{12}\left(\vec{u};\left|\vec{\tau}\right|,\vec{u}'\right)$ shown in black.

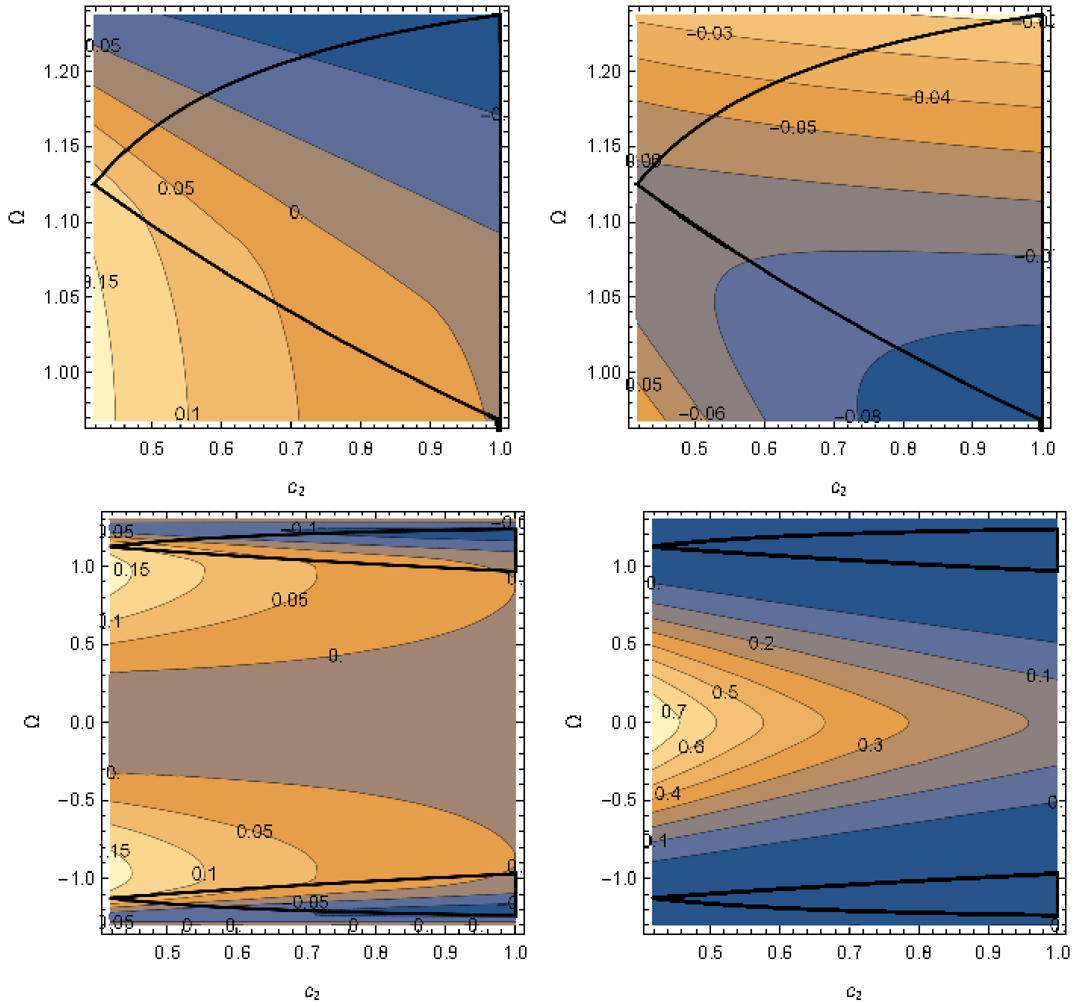

Figure 4.7: Contours of $\partial_{|\vec{\tau}|}p_{12}\left(|\vec{\tau}|\right)$ at $|\vec{\tau}|=0$ (left) and $\partial_{|\vec{\tau}|}p_{23}\left(|\vec{\tau}|\right)$ at $|\vec{\tau}|=0$ (Right) for the case of postive α_1/R'_{11}. The extent of the admissible space due to $0\leq f_{12}\left(\vec{u};|\vec{\tau}|,\vec{u}'\right)$ is outlined in black. Bottom shows the full remaining admissible space of c_2 Ω and the top is reduced to $\frac{\sqrt{15}}{4}<|\Omega|<\frac{7}{4\sqrt{2}}$

With Figure 4.7 we can now see that there is no admissible space which has the desired feature of a statistically continuous material without causing negative probabilities.

CHAPTER 5 FUTURE WORK

5.1 Three Dimensional Case

This section outlines future work required to apply the results derived in this thesis to the problem of three dimensions. As with the two dimensional case, for a statistically isotropic material the field fluctuation statistics should be invariant to rotations about the applied field direction.

For two dimensions this was satisfied by allowing field fluctuations to enter as independent field fluctuations directed along or in equiprobable pairs symmetric to the applied field direction. In the case of three dimensions, the first case can remain. However for any case where the field fluctuations are not collinear with the applied field, this requires an infinite set of pairs to satisfy symmetry for all directions orthogonal to the applied field. This is the fundamental difference which must be addressed to enable a solution for three dimensions.

In the case of three dimensions, field fluctuations can occur in the applied field direction or with field values equiprobable concentrated on a circle which exists on a plane orthogonal and having the average value aligned with, the applied field direction.

Building upon the previous results, again take one field fluctuation in the second phase and assume the applied field is in the 1 direction. Then the generalization of the previous results to three dimensions for many sets of vectors is

$$\begin{aligned} f_1(\vec{u}) &= \sum_{\mu} p_\mu \delta\left(\vec{u} - \vec{R}_\mu\right) \\ &\quad + \sum_{\gamma} \frac{1}{2\pi r_\gamma} p_{\gamma o} \delta(u_1 - x_\gamma)\, \delta\left(\sqrt{(u_2)^2 + (u_3)^2} - r_\gamma\right) \\ \text{and} \quad f_2(\vec{u}) &= \sum_{\alpha} q_\alpha \delta\left(\vec{u} - \vec{Q}_\alpha\right), \end{aligned}$$

where p_μ, $p_{\mu o}/2\pi r_\mu$, and q_α are probabilities of the corresponding field fluctuation, μ runs values $1...N$, γ runs values $1...L$, and α runs values $1...M$.

If we limit to only one vector collinear with the applied field and one vector set which has some orthogonal component, this is the generalization of the three vector case to three dimensions

$$\begin{aligned} f_1(\vec{u}) &= p_1\delta(u_1-R)\,\delta(u_2)\,\delta(u_3) \\ &\quad+\frac{1}{2\pi r}p_o\delta(u_1-x)\,\delta\left(\sqrt{(u_2)^2+(u_3)^2}-r\right) \\ \text{and}\quad f_2(\vec{u}) &= c_2\delta(u_1-Q)\,\delta(u_2)\,\delta(u_3)\,. \end{aligned}$$

Similar constraints on probabilities arise

$$0\le p_1+p_o\equiv c_1\le 1,\ 0\le p_1\le 1,\ 0\le p_o\le 1,$$
$$0\le c_2\le 1,\quad \text{and}\quad 1=c_1+c_2$$

and as before field fluctuations by definition vanish

$$p_1R+p_ox+c_2Q=0.$$

It is the two point probabilities which will introduce a challenge for the three dimensional case. Since realistic composites must have the feature of two points of observation at the limit of coinciding resulting in the one point statistics

$$\begin{aligned} f_{11}(\vec{u},0;\vec{u}') &= f_1(\vec{u})\,\delta(\vec{u}-\vec{u}')\,, \\ f_{12}(\vec{u},0;\vec{u}') &= 0, \\ \text{and}\quad f_{22}(\vec{u},0;\vec{u}') &= f_2(\vec{u})\,\delta(\vec{u}-\vec{u}')\quad\text{as } |\vec{\tau}|\to 0 \end{aligned}$$

the field fluctuation correlations are not independent like they were in the two dimensional case. The correlation is denoted below by $p_{oo}(\vec{\tau},u_2,u_3,u_2',u_3')$ and lacks an equivalent for the two dimensional case.

The functions which relate each of the field fluctuations spatially over $\vec{\tau}$ can be expressed in a general form as

$$
\begin{aligned}
f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right) = {} & p_{11}\left(\vec{\tau}\right)\delta\left(u_1-R\right)\delta\left(u_2\right)\delta\left(u_3\right)\delta\left(u_1'-R\right)\delta\left(u_2'\right)\delta\left(u_3'\right)\\
& + p_{o1}\left(\vec{\tau}\right)\frac{1}{2\pi r}\delta\left(u_1-x\right)\delta\left(\sqrt{\left(u_2\right)^2+\left(u_3\right)^2}-r\right)\delta\left(u_1'-R\right)\delta\left(u_2'\right)\delta\left(u_3'\right)\\
& + p_{1o}\left(\vec{\tau}\right)\delta\left(u_1-R\right)\delta\left(u_2\right)\delta\left(u_3\right)\frac{1}{2\pi r}\delta\left(u_1'-x\right)\delta\left(\sqrt{\left(u_2'\right)^2+\left(u_3'\right)^2}-r\right)\\
& + p_{oo}\left(\vec{\tau},u_2,u_3,u_2',u_3'\right)\frac{1}{2\pi r}\delta\left(u_1-x\right)\delta\left(\sqrt{\left(u_2\right)^2+\left(u_3\right)^2}-r\right)\\
& \mathrm{x}\frac{1}{2\pi r}\delta\left(u_1'-x\right)\delta\left(\sqrt{\left(u_2'\right)^2+\left(u_3'\right)^2}-r\right)\\
& + f_1\left(\vec{u}\right)f_1\left(\vec{u}'\right)
\end{aligned}
$$

$$
\begin{aligned}
f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right) = {} & s_{11}\left(\vec{\tau}\right)\delta\left(u_1-R\right)\delta\left(u_2\right)\delta\left(u_3\right)\delta\left(u_1'-Q\right)\delta\left(u_2'\right)\delta\left(u_3'\right)\\
& + s_{o1}\left(\vec{\tau}\right)\frac{1}{2\pi r}\delta\left(u_1-x\right)\delta\left(\sqrt{\left(u_2\right)^2+\left(u_3\right)^2}-r\right)\delta\left(u_1'-Q\right)\delta\left(u_2'\right)\delta\left(u_3'\right)\\
& + f_1\left(\vec{u}\right)f_2\left(\vec{u}'\right)
\end{aligned}
$$

$$
f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right) = q_{11}\left(\vec{\tau}\right)\delta\left(u_1-Q\right)\delta\left(u_2\right)\delta\left(u_3\right)\delta\left(u_1'-Q\right)\delta\left(u_2'\right)\delta\left(u_3'\right)+f_2\left(\vec{u}\right)f_2\left(\vec{u}'\right)
$$

where due to symmetry

$$
p_{o1}\left(\vec{\tau}\right)=p_{1o}\left(\vec{\tau}\right)\quad\text{and}\quad f_{21}\left(\vec{u};\vec{\tau},\vec{u}'\right)=f_{12}\left(\vec{u}';\vec{\tau},\vec{u}\right).
$$

Compatibility conditions of joint probabilities with microstructural characteristics

$$
\begin{aligned}
& \int f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right)d\vec{u}d\vec{u}' = f_{11}\left(\vec{\tau}\right)\\
& \int f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right)d\vec{u}d\vec{u}' = f_{12}\left(\vec{\tau}\right)\\
\text{and}\quad & \int f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right)d\vec{u}d\vec{u}' = f_{22}\left(\vec{\tau}\right),
\end{aligned}
$$

compatibility condition of one and two point joint distributions

$$\int \left(f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right)+f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right)\right)d\vec{u}'=f_1\left(\vec{u}\right)\quad\text{and}\quad\int \left(f_{21}\left(\vec{u};\vec{\tau},\vec{u}'\right)+f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right)\right)d\vec{u}'=f_2\left(\vec{u}\right)$$

and the remaining conditions for non-negativeness of probability remain

$$0\leq f_{11}\left(\vec{u};\vec{\tau},\vec{u}'\right),\ 0\leq f_{12}\left(\vec{u};\vec{\tau},\vec{u}'\right),\ \text{and } 0\leq f_{22}\left(\vec{u};\vec{\tau},\vec{u}'\right)$$

As before the positive definiteness of joint 2 point probability must be satisfied and the solution of any conductivity problem must be potential (i.e. for any realization $\Delta u=0$) which in statistical terms requires the existence of the correlation function of field potential

$$\begin{aligned}B\left(\vec{k}\right)k_ik_j &= \int u_iu_j'(f_{11}\left(\vec{u};\vec{k},\vec{u}'\right)-f_1\left(\vec{u}\right)f_1\left(\vec{u}'\right)+f_{12}\left(\vec{u};\vec{k},\vec{u}'\right)-f_1\left(\vec{u}\right)f_2\left(\vec{u}'\right)\\ &\quad+f_{21}\left(\vec{u};\vec{k},\vec{u}'\right)-f_2\left(\vec{u}\right)f_1\left(\vec{u}'\right)+f_{22}\left(\vec{u};\vec{k},\vec{u}'\right)-f_2\left(\vec{u}\right)f_2\left(\vec{u}'\right))d\vec{u}d\vec{u}'\end{aligned}$$

where

$$0\leq B\left(\vec{k}\right)\quad\text{for all }\vec{k}.$$

For three dimensions, a large amount of the methods utilized apply directly, however the addition of the correlation function $p_{oo}\left(\vec{\tau},u_2,u_3,u_2',u_3'\right)$ introduces additional complexities.

5.2 Hashin-Shtrikman Variational Principal

An additional opportunity for future work, is using the simplified expressions developed in this thesis is to develop new bounds for problems of conductivity using the Hashin-Shtrikman variational principle for probabilistic measure [see e.g. 3]. The statistically anisotropic case can also be considered.

This variational principle enables determination of the effective conductivity from the true probability densities $f\left(a\right)$, $f\left(a,p\right)$, and $f\left(a;\vec{\tau},a\prime\right)$ limited to only the constraints of satisfying the non-negativity of probabilities and compatibility conditions $(3.1 - 3.9)$

$$\frac{1}{2}a_{ij}^{\text{eff}}v_iv_j = \max_{f,a_o\in(3.1-3.9)} \mathbb{I}^-(f) = \int \left(v_ip_i - \frac{1}{2}b_{ij}^-(a,a_o)\,p_ip_j\right) f(a,p)\,dadp + \frac{1}{2}a_ov_iv_i \quad (5.1)$$
$$- \int \frac{k_ik_j}{|k|^2}(p_i - \bar{p}_i)\left(p_j' - \bar{p}_j'\right)\left(f(a;\vec{\tau},a\prime) - f(a)\,f(a')\right) da\ dp\ da'dp'dV_k$$

where p is known as the polarization field

$$p_i = \left(a_{ij} - a_o\delta_{ij}'\right)u_j$$

$\bar{p}_i$ average polarization

$$\bar{p}_i = \int p_if(a,p)\,dadp$$

b_{ij}^- is inverse to conductivities

$$(a_{ij} - a_o\delta_{ij})\,b_{jk}^- = \delta_{ik}$$

$f(a,p)$ is the joint probability of conductivities and polarizations, and a_o and unknown parameter.

In the classical bounds it was taken that the correlation function of fluctuations are isotropic which stands in contrast to both the analytically determined correlations for Debye materials (3.26) as well as the results for particulate composites (Chapter 2).

For this reason, using the simplified relationships developed in 3.11 and the probability densities determined for Debye type materials which can be used a trial functions, it seems improvements upon the classical Hashin-Shtrikman bounds for isotopic materials can now be developed using the Hashin-Shtrikman variational principle for probabilistic measure.

APPENDIX A PROBABILTIY DENSITY OF ELECTRIC POTENTIAL

To find probability density of electric potential (2.18) for particles of equal radii, one has to find the limit

$$\lim_{N/V\to n, V\to\infty}\left(\int\limits_V e^{-iy\varphi}\frac{d^3r}{|V|}\right)^N. \tag{A.1}$$

This limit can be rewritten as

$$\lim_{N/V\to n, V\to\infty}\left(1-\int\limits_V\left(1-e^{-iy\varphi}\right)\frac{d^3r}{|V|}\right)^N = \lim_{N/V\to n, V\to\infty} e^{N\ln\left[1-\frac{1}{|V|}\frac{4\pi R^3}{3}A(y,R)\right]} \tag{A.2}$$

$$A(y,R)\equiv\frac{3}{4\pi R^3}\lim_{V\to\infty}\int\limits_V\left(1-e^{-iy\varphi}\right)d^3r, \tag{A.3}$$

where $\varphi(r)$ is given by (2.16). In $(A.2)$ the value within the logarithm tends to 1 as the integration volume $|V|$ tends to infinity; approximating the logarithm by the first nonzero term of the Taylor series expansion of the logarithm results in (2.20).

Due to spherical symmetry of the integral in $(A.3)$, without loss of generality vector v_i can be directed along x_3-axis, using (2.16)

$$A(y,R)=\frac{3}{4\pi R^3}\left(\int\limits_{0\le|r|\le R}\left(1-e^{iyr_3}\right)d^3r+\int\limits_{R\le|r|\le\infty}\left(1-e^{iyr_3\left(\frac{R}{|r|}\right)^3}\right)d^3r\right).$$

Scaling of coordinates $r_i\to\rho_i$, shows that $A(y,R)$ is, in fact, a function of one argument $t=yR$, $\rho_i=r_i/R$,

$$A(t)=\frac{3}{4\pi}\left(\int\limits_{0\le|\rho|\le 1}\left(1-e^{it|\rho|\cos\theta}\right)d^3\rho+\int\limits_{1\le|\rho|\le\infty}\left(1-e^{it\left(\frac{1}{|\rho|}\right)^2\cos\theta}\right)d^3\rho\right).$$

Integration over spherical coordinates θ and ϕ, can be done explicitly. We get for the first integral

$$
\begin{aligned}
&\frac{3}{4\pi}\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}\int\limits_{0\le|\rho|\le1}\left(1-e^{it|\rho|\cos\theta}\right)|\rho|^2\,d\,|\rho|\sin\theta d\theta d\phi \\
=\;& 3\int\limits_0^1\left(1-\frac{1}{t\,|\rho|}\sin\left[t\,|\rho|\right]\right)|\rho|^2\,d\,|\rho| = 3\left(\frac{1}{t}\right)^3\int\limits_0^t\left(1-\frac{1}{m}\sin\left[m\right]\right)m^2dm,
\end{aligned}
$$

and for the second integral

$$
\begin{aligned}
&\frac{3}{4\pi}\int\limits_{0\le\theta\le\pi}\int\limits_{0\le\phi\le2\pi}\int\limits_{1\le|\rho|\le\infty}\left(1-e^{it\left(\frac{1}{|\rho|}\right)^2\cos\theta}\right)|\rho|^2\,d\,|\rho|\sin\theta d\theta d\phi \\
=\;& 3\int\limits_1^\infty\left(1-\frac{|\rho|^2}{t}\sin\left[\frac{t}{|\rho|^2}\right]\right)|\rho|^2\,d\,|\rho| = 3\,|t|^{3/2}\int\limits_{1/\sqrt{|t|}}^\infty\left(1-m^2\sin\left[\frac{1}{m^2}\right]\right)m^2dm.
\end{aligned}
$$

We have arrived at (2.20).

APPENDIX B ELECTRIC FIELD FOR RANDOM DISTRIBUTION OF PARTICLE RADII

Consider a distribution of particle radii, with k_1 particles of radius R_1 number density n_1, k_2 particles of radius R_2 number density n_2, and so on, such that $k_1 + k_2 + ...k_m = N$ and $n_1 + n_2 + ...n_m = n$. Then the following limit is to be found

$$\lim_{N/V\to n, V\to\infty} \left(\frac{1}{|V|} \int\limits_V e^{-iy\psi_1} d^3r \right)^{k_1} \left(\frac{1}{|V|} \int\limits_V e^{-iy\psi_2} d^3r \right)^{k_2} ... \left(\frac{1}{|V|} \int\limits_V e^{-iy\psi_m} d^3r \right)^{k_m} . \quad \text{(B.1)}$$

The first member of the product $(B.1)$ can be written as

$$\lim_{k_1/V\to n_1, V\to\infty} \left(\frac{1}{|V|} \int\limits_V e^{-iy\psi_1} d^3r \right)^{k_1} = \lim_{k_1/V\to n_1, V\to\infty} e^{k_1 \ln\left(1 - \frac{1}{|V|} \frac{4\pi R_1{}^3}{3} B(y)\right)} \quad \text{(B.2)}$$

where

$$B(y) \equiv \frac{3}{4\pi (R_1)^3} \lim_{V\to\infty} \int\limits_V \left(1 - e^{-iy\psi_1}\right) d^3r, \quad \text{(B.3)}$$

and ψ is given by (2.16).

Due to spherical symmetry of the integral in $(B.3)$, without loss of generality vector v_i again can be directed along x_3-axis. Using (2.16) we have

$$B(y) = \frac{3}{4\pi (R_1)^3} \left(\int\limits_{0\le|r|\le R_1} \left(1 - e^{-iy\zeta_3}\right) d^3r + \int\limits_{R_1\le|r|\le\infty} \left(1 - e^{-iy\left(\frac{R_1}{|r|}\right)^3 \left(\delta_{i3} - 3\frac{r_i r_3}{|r|^2}\right)\zeta_i}\right) d^3r \right).$$

After scaling of coordinates $\rho_i = r_i/R_1$, $B(y)$ takes the form

$$B(y) = \frac{3}{4\pi} \left(\int\limits_{0\le|\rho|\le 1} \left(1 - e^{-iy\zeta_3}\right) d^3\rho + \int\limits_{1\le|\rho|\le\infty} \left(1 - e^{-iy\left(\frac{1}{|\rho|}\right)^3 \left(\delta_{i3} - 3\frac{\rho_i \rho_3}{|\rho|^2}\right)\zeta_i}\right) d^3\rho \right), \quad \text{(B.4)}$$

which shows that $B(y)$ does not depend on the particle size.

Therefore the limit $(B.1)$ simplifies to

$$\lim_{k_1/V\to n_1, V\to\infty} e^{k_1 \ln\left(1-\frac{1}{|V|}\frac{4\pi R_1{}^3}{3}B(y)\right)} \lim_{k_2/V\to n_2, V\to\infty} e^{k_2 \ln\left(1-\frac{1}{|V|}\frac{4\pi R_2{}^3}{3}B(y)\right)}$$
$$\cdots \lim_{k_m/V\to n_m, V\to\infty} e^{k_m \ln\left(1-\frac{1}{|V|}\frac{4\pi R_m{}^3}{3}B(y)\right)}$$

where $B(y)$ is the function $(B.4)$. Replacing the logarithms by the first nonzero terms of the Taylor expansion gives the value of the limit

$$e^{-\bar{c}B(y)},$$

where $\bar{c}$, for any size distribution of particle radii $f(R)$, is

$$\bar{c} = n\frac{4\pi}{3}\int_0^\infty R^3 f(R)\, dR.$$

To obtain (2.20) we note that due to spherical symmetry in $(B.4)$ one can choose ζ as

$$\zeta = \left\{ \sin\alpha, \quad 0, \quad \cos\alpha \right\}. \tag{B.5}$$

Then

$$B(y) = \left(1 - e^{-iy\cos\alpha}\right) + \frac{3}{4\pi}\int_{1\le|\rho|\le\infty} \left(1 - e^{-iy/|\rho|^3 C}\right)|\rho|^2\, d|\rho| \sin\theta d\theta d\phi, \tag{B.6}$$

where $C = \cos\alpha - 3\cos\theta\left(\sin\alpha\cos\phi\sin\theta + \cos\alpha\cos\theta\right)$. The second integral in $(B.6)$, is simplified by the substitution $|\rho|^3 = y/s$, which gives the expression

$$\frac{1}{4\pi}y\int_{0\le\theta\le\pi}\int_{0\le\phi\le 2\pi}\int_{0\le s\le y}\left(1 - e^{isC}\right)\frac{1}{s^2}ds\sin\theta d\theta d\phi.$$

Let us break the integrand into even and odd parts:

$$\frac{1}{4\pi}\int\limits_{0\leq\theta\leq\pi}\int\limits_{0\leq\phi\leq 2\pi}\int\limits_{0\leq s\leq|y|}\left(|y|\,\frac{1-\cos\left[s\,|C|\right]}{s^2}-iy\frac{\sin\left[sC\right]}{s^2}\right)ds\sin\theta d\theta d\phi. \tag{B.7}$$

The second term in $(B.7)$ causes issues in numerical integration for small values of y. We modify the integrand by adding the term sC, integral of which is zero over θ and ϕ for the two cases considered ($\alpha = 0$ and $\alpha = \pi/2$):

$$\frac{1}{4\pi}\int\limits_{0\leq\theta\leq\pi}\int\limits_{0\leq\phi\leq 2\pi}\int\limits_{0\leq s\leq|y|}\left(|y|\,\frac{1-\cos\left[s\,|C|\right]}{s^2}-iy\left(\frac{\sin\left[sC\right]}{s^2}-\frac{C}{s}\right)\right)ds\sin\theta d\theta d\phi. \tag{B.8}$$

Integrand in $(B.8)$ is not singular at $s = 0$ and $(B.8)$ is easily evaluated numerically.

APPENDIX C TWO VECTORS IN PHASE 1

Two vectors in the first phase is insuffi cient to satisfy the condition of potentiality.

$$\hat{B}\left(\vec{k}\right)k_ik_j = \left(\hat{p}_{\mu\nu}\left(\vec{k}\right) - \frac{1}{c_2}\left(p_\mu\hat{s}_{\nu 1}\left(\vec{k}\right) + p_\nu\hat{s}_{\mu 1}\left(\vec{k}\right)\right) + \frac{1}{\left(c_2\right)^2}p_\mu p_\nu c_1 c_2\hat{h}_o\left(\vec{k}\right)\right)R_{\mu i}R_{\nu j}.$$

To find $\hat{B}\left(\vec{k}\right)$ introduce inverse to R_μ [1] (i.e. $R_{\mu i}R^{\nu i} = \delta^\nu_\mu$) then

$$\mathbb{R}^{\mu\nu}\left(\vec{k}\right) = \hat{B}\left(\vec{k}\right)k_ik_jR^{\mu i}R^{\nu j},$$

and solve for $\hat{p}_{\mu\nu}\left(\vec{k}\right)$

$$\hat{p}_{\mu\nu}\left(\vec{k}\right) = \hat{B}\left(\vec{k}\right)k_ik_jR^{\mu i}R^{\nu j} + \frac{1}{c_2}\left(p_\mu\hat{s}_{\nu 1}\left(\vec{k}\right) + p_\nu\hat{s}_{\mu 1}\left(\vec{k}\right)\right) - \frac{1}{\left(c_2\right)^2}p_\mu p_\nu\hat{q}_{11}\left(\vec{k}\right).$$

Then make sum over μ, ν

$$\sum_{\mu,\nu}\hat{p}_{\mu\nu}\left(\vec{k}\right) = \hat{B}\left(\vec{k}\right)\sum_{\mu,\nu}k_ik_jR^{\mu i}R^{\nu j} - \left(2\frac{c_1}{c_2} + \left(\frac{c_1}{c_2}\right)^2\right)c_1c_2\hat{h}_o\left(\vec{k}\right) = c_1c_2\hat{h}_o\left(\vec{k}\right),$$

and

$$\hat{B}\left(\vec{k}\right) = \left(c_2\sum_\mu k_iR^{\mu i}\right)^{-2}c_1c_2\hat{h}_o\left(\vec{k}\right). \tag{C.1}$$

Next, to solve for $\hat{s}_{\mu 1}\left(\vec{k}\right)$ insert solution of $\hat{B}\left(\vec{k}\right)$, then

$$\sum_\nu\hat{p}_{\mu\nu}\left(\vec{k}\right) = \left(k_iR^{\mu i}\left(\sum_\nu k_jR^{\nu j}\right)^{-1}\left(\frac{1}{c_2}\right)^2 - \frac{p_\mu}{c_2}\left(\frac{1}{c_2}\right)\right)c_1c_2\hat{h}_o\left(\vec{k}\right) + \frac{c_1}{c_2}\hat{s}_{\mu 1}\left(\vec{k}\right) = -\hat{s}_{\mu 1}\left(\vec{k}\right).$$

After simplifications

$$\hat{s}_{\mu 1}\left(\vec{k}\right) = \frac{1}{c_2}\left(p_\mu - k_iR^{\mu i}\left(\sum_\nu k_jR^{\nu j}\right)^{-1}\right)c_1c_2\hat{h}_o\left(\vec{k}\right). \tag{C.2}$$

---

[1] this is possible only for $N = 2$ in the 2-dimensional case when $\vec{R}_1 \neq const\ \vec{R}_2$. If $\vec{R}_1$ is proportional to $\vec{R}_2$ then this constraint can only hold for the trivial case of a uniform field.

Also, with this relation $\hat{p}_{\mu\nu}\left(\vec{k}\right)$ can be further simplified to

$$\hat{p}_{\mu\nu} = \left(\frac{1}{c_2}\right)^2 \left(p_\mu - k_i R^{\mu i} \left(\sum_a k_i R^{ai}\right)^{-1}\right) \left(p_\nu - k_j R^{\nu j} \left(\sum_a k_i R^{ai}\right)^{-1}\right) c_1 c_2 \hat{h}_o\left(\vec{k}\right). \quad \text{(C.3)}$$

Using $(C.2, C.3)$ positive definiteness always holds.

We will find $s_{\mu 1}(\vec{\tau})$ from $\hat{s}_{\mu 1}\left(\vec{k}\right)$ directly

$$s_{\mu 1}(\vec{\tau}) = c_1 p_\mu h_o(\vec{\tau}) - c_1 \int \mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right) \vec{R}^\mu \cdot \vec{k} \left(\sum_\nu \vec{R}^\nu \cdot \vec{k}\right)^{-1} \hat{h}_o\left(\vec{k}\right) dV_k. \quad \text{(C.4)}$$

To find the second term, define

$$\sum_\nu \vec{R}^\nu = \vec{\boldsymbol{R}}$$

and consider 2 dimensional space with $\mathbf{R}^2 = 0$ and $\mathbf{R}^1 \neq 0$, then the integral in (3.16) is

$$\frac{R^{\mu 1}}{\boldsymbol{R}^1} h_o(\vec{\tau}) + \frac{R^{\mu 2}}{\boldsymbol{R}^1} \int \mathrm{Exp}\left(-i\ \tau_1 k_1\right) \mathrm{Exp}\left(-i\ \tau_2 k_2\right) \frac{k_2}{k_1} \hat{h}_o\left(\vec{k}\right) \frac{dk_1 dk_2}{(2\pi)^2}. \quad \text{(C.5)}$$

Consider the integral in (2.27) and denote this integral by Φ; break the integrand into even and odd parts, and since $\hat{h}_o\left(\vec{k}\right)$ is even over k_1 and k_2 and integration is over all space the cosine terms evaluate to zero simplifying to

$$\Phi(\vec{\tau}) = \int \frac{k_2}{k_1} \mathrm{Sin}\left(\tau_1 k_1\right) \mathrm{Sin}\left(\tau_2 k_2\right) \hat{h}_o\left(\vec{k}\right) \frac{dk_1 dk_2}{(2\pi)^2}.$$

Thus

$$p_\mu c_2 + \left(p_\mu - \frac{R^{\mu 1}}{\boldsymbol{R}^1}\right) c_1 h_o(\vec{\tau}) - \frac{R^{\mu 2}}{\boldsymbol{R}^1} c_1 \Phi(\tau_1, \tau_2) \geq 0.$$

Express the two equations explicitly

$$p_1 c_2 + \left(p_1 - \frac{R^{11}}{\boldsymbol{R}^1}\right) c_1 h_o(\vec{\tau}) - \frac{R^{12}}{\boldsymbol{R}^1} c_1 \Phi(\tau_1, \tau_2) \geq 0$$
$$p_2 c_2 + \left(p_2 - \frac{R^{21}}{\boldsymbol{R}^1}\right) c_1 h_o(\vec{\tau}) - \frac{R^{22}}{\boldsymbol{R}^1} c_1 \Phi(\tau_1, \tau_2) \geq 0$$

and add the first equation to the second giving an upper bound to the first relation

$$c_2 c_1 (1 - h_o(\vec{\tau})) \geq p_1 c_2 + \left(p_1 - \frac{R^{11}}{\boldsymbol{R}^1} \right) c_1 h_o(\vec{\tau}) - \frac{R^{12}}{\boldsymbol{R}^1} c_1 \Phi(\tau_1, \tau_2) \geq 0$$

here it was used that

$$\frac{R^{12}}{\boldsymbol{R}^1} + \frac{R^{22}}{\boldsymbol{R}^1} = 0 \text{ and } \frac{R^{11}}{\boldsymbol{R}^1} + \frac{R^{21}}{\boldsymbol{R}^1} = 1.$$

Since $h_o(\vec{\tau}) = 1$ and $\Phi(\tau_1, \tau_2) = 0$ at $|\vec{\tau}| = 0$, the upper and lower bounds collapse and the equality follows

$$p_\mu = \frac{R^{\mu 1}}{\boldsymbol{R}^1} c_1,$$

and after rearrangement

$$1 \geq \frac{p_\mu}{c_1} - \frac{R^{\mu 2}}{\boldsymbol{R}^1} \frac{1}{c_2} \frac{\Phi(\tau_1, \tau_2)}{(1 - h_o(\vec{\tau}))} \geq 0.$$

Numerically it was found

$$\frac{2}{3} > \frac{\Phi(\tau_1, \tau_2)}{(1 - h_o(\vec{\tau}))} > -\frac{2}{3}$$

then

$$1 \geq \frac{p_\mu}{c_1} \pm \frac{2}{3} \frac{1}{c_2} \frac{R^{\mu 2}}{\boldsymbol{R}^1} \geq 0.$$

We also must find $p_{\mu\nu}(\vec{\tau})$ from $\hat{p}_{\mu\nu}\left(\vec{k}\right)$. It is clear that

$$\hat{p}_{\mu\nu}\left(\vec{k}\right) = \left(p_\mu p_\nu - \left(p_\mu k_i R^{\nu i} + p_\nu k_j R^{\mu j}\right) \left(\sum_a k_i R^{ai} \right)^{-1} + k_i R^{\mu i} k_j R^{\nu j} \left(\sum_a k_i R^{ai} \right)^{-2} \right) \frac{c_1}{c_2} \hat{h}_o\left(\vec{k}\right)$$

using the previous results, assumption that $\mathbf{R}^2 = 0$, and notation used for $s_{\mu 1}(\vec{\tau})$

$$p_{\mu\nu}(\vec{\tau}) = \left(p_\mu p_\nu - p_\mu \frac{R^{\nu 1}}{\boldsymbol{R}^1} - p_\nu \frac{R^{\mu 1}}{\boldsymbol{R}^1} \right) \frac{c_1}{c_2} h_o(\vec{\tau}) - \left(p_\mu \frac{R^{\nu 2}}{\boldsymbol{R}^1} + p_\nu \frac{R^{\mu 2}}{\boldsymbol{R}^1} \right) \frac{c_1}{c_2} \Phi(\tau_1, \tau_2)$$
$$+ \frac{c_1}{c_2} \int \mathrm{Exp}\left(-i\, \vec{\tau} \cdot \vec{k} \right) \frac{\vec{R}^\mu \cdot \vec{k}}{\vec{\boldsymbol{R}} \cdot \vec{k}} \frac{\vec{R}^\nu \cdot \vec{k}}{\vec{\boldsymbol{R}} \cdot \vec{k}} \hat{h}_o\left(\vec{k}\right) dV_k.$$

The integral is

$$\frac{R^{\mu 1}R^{\mu 1}}{\boldsymbol{R}^1\boldsymbol{R}^1}h_o\left(\vec{\tau}\right)+\left(\frac{R^{\mu 1}R^{\nu 2}}{\boldsymbol{R}^1\boldsymbol{R}^1}+\frac{R^{\mu 2}R^{\nu 1}}{\boldsymbol{R}^1\boldsymbol{R}^1}\right)\Phi\left(\tau_1,\tau_2\right)+\frac{R^{\mu 2}R^{\nu 2}}{\boldsymbol{R}^1\boldsymbol{R}^1}\Psi\left(\tau_1,\tau_2\right)$$

where

$$\Psi\left(\tau_1,\tau_2\right)=\int\left(\frac{k_2}{k_1}\right)^2\mathrm{Exp}\left(-i\ \vec{\tau}\cdot\vec{k}\right)\hat{h}_o\left(\vec{k}\right)dV_k,$$

simplifying to

$$p_{\mu\nu}\left(\vec{\tau}\right)=\frac{R^{\mu 1}}{\boldsymbol{R}^1}\frac{R^{\nu 1}}{\boldsymbol{R}^1}c_1c_2h_o\left(\vec{\tau}\right)-\left(\frac{R^{\mu 1}}{\boldsymbol{R}^1}\frac{R^{\nu 2}}{\boldsymbol{R}^1}+\frac{R^{\mu 2}}{\boldsymbol{R}^1}\frac{R^{\nu 1}}{\boldsymbol{R}^1}\right)c_1\Phi\left(\tau_1,\tau_2\right)+\frac{R^{\mu 2}R^{\nu 2}}{\boldsymbol{R}^1\boldsymbol{R}^1}\frac{c_1}{c_2}\Psi\left(\tau_1,\tau_2\right).$$

here we used

$$p_\mu=\frac{R^{\mu 1}}{\boldsymbol{R}^1}c_1.$$

Break into even odd parts and make scaling of coordinate $k_2=\left(1+k_1^2\right)^{1/2}y$

$$\begin{aligned}\Psi\left(\tau_1,\tau_2\right)&=\int\left(\frac{k_2}{k_1}\right)^2\frac{\mathrm{Cos}\left(\tau_2k_2\right)\mathrm{Cos}\left(\tau_1k_1\right)}{\left(1+k_1^2+k_2^2\right)^{3/2}}\frac{dk_1dk_2}{2\pi}\\&=\int\left(\frac{k_2}{k_1}\right)^2\frac{\mathrm{Cos}\left(\tau_2k_2\right)\mathrm{Cos}\left(\tau_1k_1\right)}{\left(1+k_1^2\right)^{3/2}\left(1+\frac{k_2^2}{1+k_1^2}\right)^{3/2}}\frac{dk_1dk_2}{2\pi}\\&=\int\frac{\mathrm{Cos}\left(\tau_1k_1\right)}{k_1^2}\frac{y^2\mathrm{Cos}\left(\tau_2\left(1+k_1^2\right)^{1/2}y\right)}{\left(1+y^2\right)^{3/2}}\frac{dk_1dy}{2\pi}\end{aligned}$$

and introduce function

$$\varphi=\frac{\mathrm{Cos}\left(ty\right)}{\left(1+y^2\right)^{3/2}}$$

where

$$t=\tau_2\left(1+k_1^2\right)^{1/2}.$$

Then

$$\Psi\left(\tau_1,\tau_2\right)=-\int\frac{\mathrm{Cos}\left(\tau_1k_1\right)}{k_1^2}\left(\frac{d^2}{dt^2}\varphi\right)\frac{dk_1dy}{2\pi}$$

Function Ψ can take values from negative to positive infinity. Considering the constraint must hold for any $\vec{\tau}$

$$p_{\mu\nu}\left(\vec{\tau}\right)+p_{\mu}p_{\nu}\geq 0$$

$$\left(\frac{c_1}{c_2}+h_o\left(\vec{\tau}\right)\right)\frac{R^{\mu 1}}{\boldsymbol{R}^1}\frac{R^{\nu 1}}{\boldsymbol{R}^1}c_1c_2-\left(\frac{R^{\mu 1}}{\boldsymbol{R}^1}\frac{R^{\nu 2}}{\boldsymbol{R}^1}+\frac{R^{\mu 2}}{\boldsymbol{R}^1}\frac{R^{\nu 1}}{\boldsymbol{R}^1}\right)c_1\Phi\left(\tau_1,\tau_2\right)+\frac{R^{\mu 2}R^{\nu 2}}{\boldsymbol{R}^1\boldsymbol{R}^1}\frac{c_1}{c_2}\Psi\left(\tau_1,\tau_2\right)\geq 0$$

then it follows that $R^{\mu 2}=0$; the vectors R^{μ} must be collinear.

As noted previously, the case of collinear vectors cannot satisfy potentiality for $N=2$. Therefore this result cannot satisfy all required constraints.

APPENDIX D GENERAL SOLUTION OF CORRELATIONS

In this section the positive definiteness and potentiality conditions will be shown to be constraints between intra phase correlations $p'_{\mu\nu}\left(\vec{k}\right)$ and inter phase correlations $s'_{\mu}\left(\vec{k}\right)$.

Beginning from the relationship

$$\hat{B}\left(\vec{k}\right)k_i k_j = \left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_{\mu}\left(\vec{k}\right)s'_{\nu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j} \tag{D.1}$$

$$+\left(\frac{s'_{\mu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}R'_{\mu i} - \frac{1}{c_2}\left(p_{\mu}R'_{\mu i} + \bar{R}_i\right)\right)$$

$$\text{x}\left(\frac{s'_{\nu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}R'_{\nu j} - \frac{1}{c_2}\left(p_{\nu}R'_{\nu j} + \bar{R}_j\right)\right)c_1 c_2 \hat{h}_o\left(\vec{k}\right), \tag{D.2}$$

define a vector $\vec{k}^*$ orthogonal to $\vec{k}$ (i.e. $k_i k_i^* = 0$) with the magnitude of $k_l^*\left(\vec{k}\right)$ such that $k_l^* R'_{\gamma l} = \varepsilon_{lm} k_l R'_{\gamma m}$. Here, ε_{lm} is the Levi-Chivita symbol. In the case of two dimensions a single vector orthogonal to $\vec{k}$ exists and for the case of three dimensions this vector lies in a plane orthogonal to $\vec{k}$.

Then, after contracting $(D.1)$ with $\vec{k}^*\vec{k}^*$ we have

$$\begin{aligned}0 \;=\;& k_i^* k_j^*\left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_{\mu}\left(\vec{k}\right)s'_{\nu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j}\\ &+\left(k_i^*\left(\frac{s'_{\mu}\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}R'_{\mu i} - \frac{1}{c_2}\left(p_{\mu}R'_{\mu i} + \bar{R}_i\right)\right)\right)^2 c_1 c_2 \hat{h}_o\left(\vec{k}\right).\end{aligned}$$

Since the positive definiteness condition (3.18) requires the first term to be non-negative and by definition $c_1c_2\hat{h}_o\left(\vec{k}\right)$ is non-negative, then it follows that

$$\begin{aligned} k_i^*k_j^*\left(p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j} &= 0 \\ \text{and}\quad k_i^*\left(\frac{s'_\mu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}R'_{\mu i}-\frac{1}{c_2}\left(p_\mu R'_{\mu i}+\bar{R}_i\right)\right) &= 0. \end{aligned} \tag{D.3}$$

After contracting $(D.1)$ with $\vec{k}\vec{k}^*$ we have

$$\begin{aligned} 0 = {} & k_i^*k_j\left(p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j} \\ & + k_i^*k_j\left(\frac{s'_\mu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}R'_{\mu i}-\frac{1}{c_2}\left(p_\mu R'_{\mu i}+\bar{R}_i\right)\right)\left(\frac{s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}R'_{\nu j}-\frac{1}{c_2}\left(p_\nu R'_{\nu j}+\bar{R}_j\right)\right)c_1c_2\hat{h}_o\left(\vec{k}\right) \end{aligned}$$

and the additional constraint follows

$$k_i^*k_j\left(p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}\right)R'_{\mu i}R'_{\nu j}=0.$$

Introducing notations

$$p'_{\mu\nu}\left(\vec{k}\right)-\frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1c_2\hat{h}_o\left(\vec{k}\right)}=\mathbb{P}'_{\mu\nu},\quad k_i^*R'_{\mu i}=\mathbb{R}^*_\mu,\quad \text{and}\quad k_jR'_{\nu j}=\mathbb{R}_\nu \tag{D.4}$$

we can then write the unknown tensor $\mathbb{P}'_{\mu\nu}$ in the $\xi_\mu,\mathbb{R}_\mu,\mathbb{R}^*_\mu$ basis, assuming that $\xi_\mu,\mathbb{R}_\mu,\mathbb{R}^*_\mu$ are linearly independent and we have

$$\mathbb{P}'_{\mu\nu}\mathbb{R}^*_\mu\mathbb{R}^*_\nu=\mathbb{P}'_{\mu\nu}\mathbb{R}_\mu\mathbb{R}^*_\nu=0. \tag{D.5}$$

Alternatively if we introduce a new vector $\mathbb{S}$ which is orthogonal to ξ_μ and $\mathbb{R}^*_\mu$ with components

$$\mathbb{S}_\mu = \varepsilon_{\mu\nu\lambda}\xi_\nu\mathbb{R}^*_\lambda \tag{D.6}$$

we can then express $\mathbb{P}'_{\mu\nu}$ in the orthogonal basis of $\xi_\mu, \mathbb{S}_\mu, \mathbb{R}^*_\mu$, which in the general bilinear form is

$$\mathbb{P}'_{\mu\nu} = \mathbb{P}_1\xi_\mu\xi_\nu + \mathbb{P}_2\mathbb{S}_\mu\mathbb{S}_\nu + \mathbb{P}_3\mathbb{R}^*_\mu\mathbb{R}^*_\nu + \mathbb{P}_4\left(\mathbb{S}_\mu\xi_\nu + \xi_\mu\mathbb{S}_\nu\right) + \mathbb{P}_5\left(\mathbb{R}^*_\mu\xi_\nu + \xi_\mu\mathbb{R}^*_\nu\right) + \mathbb{P}_6\left(\mathbb{R}^*_\mu\mathbb{S}_\nu + \mathbb{S}_\mu\mathbb{R}^*_\nu\right).$$

Since $\mathbb{P}'_{\mu\nu}\xi_\mu\xi_\nu = 0,\ \xi_\mu\mathbb{S}_\mu = 0$, and $\xi_\mu\mathbb{R}^*_\mu = 0$ we find $\mathbb{P}_1 = 0$ since the number of vectors N is not zero. Next, due to the constraints $\mathbb{P}'_{\mu\nu}\xi_\nu = 0$ and $\mathbb{P}'_{\mu\nu}\xi_\mu = 0$

$$\mathbb{P}_4\mathbb{S}_\mu + \mathbb{P}_5\mathbb{R}^*_\mu = 0.$$

By definition $\mathbb{S}_\mu$ and $\mathbb{R}^*_\mu$ are orthogonal and non zero and we find

$$\mathbb{P}_4 = \mathbb{P}_5 = 0.$$

Since $\mathbb{P}'_{\mu\nu}\mathbb{R}^*_\mu\mathbb{R}^*_\nu = 0$ $(D.5)$ we have

$$\mathbb{P}'_{\mu\nu}\mathbb{R}^*_\mu\mathbb{R}^*_\nu = \mathbb{P}_3\mathbb{R}^*_\mu\mathbb{R}^*_\mu\mathbb{R}^*_\nu\mathbb{R}^*_\nu = 0 \quad \text{thus} \quad \mathbb{P}_3 = 0$$

and similarly with the second condition in $(D.4)$

$$\mathbb{P}'_{\mu\nu}\mathbb{R}_\mu\mathbb{R}^*_\nu = \mathbb{P}_6\mathbb{S}_\mu\mathbb{R}_\mu\mathbb{R}^*_\nu\mathbb{R}^*_\nu = 0.$$

Here we have the result that at least $\mathbb{P}_6$ or $\mathbb{S}_\mu\mathbb{R}_\mu$ must be zero. Since both $\mathbb{S}_\mu\mathbb{R}_\mu = \mathbb{S}_\mu k_i R'_{\mu i} = 0$ and $\mathbb{S}_\mu\mathbb{R}^*_\mu = \mathbb{S}_\mu k^*_i R'_{\mu i} = 0$ can only be satisfied in the case of a zero length $\vec{k}$ vector due to the orthogonality of k_i and k^*_i, it follows $\mathbb{P}_6$ must be zero. We now have the result that the general solution of $\mathbb{P}'_{\mu\nu}$ with $(D.4)$ and $(D.6)$ is

$$\mathbb{P}'_{\mu\nu} = \mathbb{P}_2 \mathbb{S}_\mu \mathbb{S}_\nu = \mathbb{P}_2 \left(\varepsilon_{\mu\lambda\gamma}\xi_\lambda k^*_l R'_{\gamma l}\right)\left(\varepsilon_{\nu\sigma\tau}\xi_\sigma k^*_n R'_{\tau n}\right) = \mathbb{P}_2 \left(\varepsilon_{\mu\lambda\gamma}\xi_\lambda \varepsilon_{lm} k_l R'_{\gamma m}\right)\left(\varepsilon_{\nu\sigma\tau}\xi_\sigma \varepsilon_{pn} k_p R'_{\tau n}\right).$$

If we introduce notation

$$\Delta\ \varepsilon_{im} = R'_{\mu i}\varepsilon_{\mu\lambda\gamma}\xi_\lambda R'_{\gamma m}$$

where Δ is an unknown scalar function of $R'_{\mu i}$. Upon contraction of $\mathbb{P}'_{\mu\nu}$ with $R'_{\mu i}R'_{\nu j}$ we find the relation

$$\mathbb{P}'_{\mu\nu} R'_{\mu i} R'_{\nu j} = \mathbb{P}_2 \left(\Delta \varepsilon_{im}\varepsilon_{lm} k_l\right)\left(\Delta \varepsilon_{jn}\varepsilon_{pn} k_p\right) = \mathbb{P}_2 \Delta^2 k_i k_j = \Psi\left(\vec{k}\right) k_i k_j \tag{D.7}$$

where the unknown function $\mathbb{P}_2\Delta^2$ is now denoted by $\Psi\left(\vec{k}\right)$. Here the relationship $\varepsilon_{im}\varepsilon_{lm} = \delta_{il}$ for the two dimensional case was used.

The positive definiteness condition (3.18) requires that

$$0 \le \mathbb{P}_2 \left(\mathbb{S}_\mu \varphi'_\mu\right)^2 \quad \text{therefore} \quad 0 \le \Psi\left(\vec{k}\right). \tag{D.8}$$

The second term in $(D.3)$ shows the value within the brackets is orthogonal to k^*_i, thus it must be collinear to k_i up to a constant.

Solutions of $(D.1)$ are then

$$\begin{aligned}
\left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right) s'_\nu\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}\right) R'_{\mu i} R'_{\nu j} &= \Psi\left(\vec{k}\right) k_i k_j \quad \text{and} \\
\frac{R'_{\mu i} s'_\mu\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)} - \frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right) &= \Phi\left(\vec{k}\right) k_i.
\end{aligned} \tag{D.9}$$

Contracting $(D.1)$ with $\vec{k}\vec{k}$ we have

$$\hat{B}\left(\vec{k}\right)\left(\vec{k}\cdot\vec{k}\right)^2 = k_i k_j \left(p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)}\right) R'_{\mu i} R'_{\nu j}$$
$$+ \left(k_i\left(\frac{s'_\mu\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)} R'_{\mu i} - \frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right)\right)\right)^2 c_1 c_2 \hat{h}_o\left(\vec{k}\right)$$

and similarly contracting the terms in $(D.9)$ with $\vec{k}\vec{k}$, yields the value of $\hat{B}\left(\vec{k}\right)$ in terms of $\Psi\left(\vec{k}\right)$ and $\Phi\left(\vec{k}\right)$:

$$\hat{B}\left(\vec{k}\right) = \Psi\left(\vec{k}\right) + c_1 c_2 \hat{h}_o\left(\vec{k}\right)\left(\Phi\left(\vec{k}\right)\right)^2. \tag{D.10}$$

The constraint on potentiality $(D.10)$ is always satisfied due to the non negativity of $\Psi\left(\vec{k}\right)$ from the positive definiteness condition and definitions of $c_1 c_2 \hat{h}_o\left(\vec{k}\right)$.

The general solution $(D.9)$ can be written

$$p'_{\mu\nu}\left(\vec{k}\right) - \frac{s'_\mu\left(\vec{k}\right)s'_\nu\left(\vec{k}\right)}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)} = \Psi\left(\vec{k}\right) Y^i_\mu k_i Y^j_\nu k_j + \tilde{\mathbb{P}}_{\mu\nu}\left(\vec{k}\right) \tag{D.11}$$

where

$$Y^i_\mu R'_{\mu j} = \delta^i_j \quad \text{and} \quad \tilde{\mathbb{P}}_{\mu\nu}\left(\vec{k}\right) R'_{\mu i} R'_{\nu j} = 0 \quad \text{for each } i \quad \text{and} \quad j$$

$\tilde{\mathbb{P}}_{\mu\nu} = 0.$

After contraction of $(D.11)$ with $\xi_\mu \xi_\nu$ we have

$$0 = \Psi\left(\vec{k}\right)\left(\xi_\mu Y^i_\mu k_i\right)^2. \tag{D.12}$$

Consider the decomposition of Y^i_μ

$$Y^i_\mu = Y'^i_\mu + \bar{Y}^i_\mu \xi_\mu;$$

since $Y^i_\mu R'_{\mu j} = \delta^i_j$ the term $\bar{Y}^i_\mu$ must be zero due to the definition $\xi_\mu R'_{\mu j} = 0$. We then have the condition

$$Y_\mu^i \xi_\mu = 0,$$

and $(D.12)$ is satisfied for any $\Psi\left(\vec{k}\right)$ and k_i.

The solution of $s'_\mu\left(\vec{k}\right)$ in $(D.9)$ can also be written in terms of the particular and homogenous solutions

$$s'_\mu\left(\vec{k}\right) = c_1 c_2 \hat{h}_o \left(\Phi\left(\vec{k}\right) Y_\mu^i k_i - X_\mu\right) + \tilde{s}_\mu \tag{D.13}$$

where

$$X_\mu R'_{\mu i} = -\frac{1}{c_2}\left(p_\mu R'_{\mu i} + \bar{R}_i\right), \quad \text{and} \quad \tilde{s}_\mu\left(\vec{k}\right) R'_{\mu i} = 0 \quad \text{for each } i. \tag{D.14}$$

In this solution we have the freedom to select an additional constraint on X_μ since $\tilde{s}_\mu$ is also an unknown. Let us take the case that $\xi_\mu X_\mu = 0$. Since $\xi_\mu s'_\mu\left(\vec{k}\right) = 0$ we then have the constraint

$$\frac{1}{c_1 c_2 \hat{h}_o\left(\vec{k}\right)} \xi_\mu \tilde{s}_\mu\left(\vec{k}\right) = \xi_\mu X_\mu = 0. \tag{D.15}$$

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ABSTRACT

STUDY OF PROBABILISTIC CHARACTERISTICS OF LOCAL FIELD FLUCTUATIONS IN ISOTROPIC TWO PHASE COMPOSITES: CONDUCTIVITY TYPE PROBLEMS

by

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Major: Mechanical Engineering

Degree: Doctor of Philosophy

Probability distributions of electric field and electric potential in two-phase particulate composite materials with spherical inclusions are found in the limit of small particle concentration. Additionally, a method for the approximation of local fields within random statistically isotropic composites with a finite number of parameters is presented and an approximate solution is found using the variational principle for probabilistic measure.

AUTOBIOGRAPHICAL STATEMENT

David Ostberg received a BS in Mechanical and a BS in Electrical Engineering from Lawrence Technological University in Southfield MI and an MS in Mechanical Engineering from Wayne State University in Detroit MI. He is currently employed as an Research Mechanical Engineer at the US Army Tank Automotive Research Development and Engineering Center in Warren, MI where he has been employed since 2001.