## SPECTRAL MEASURE COMPUTATIONS FOR COMPOSITE MATERIALS \*

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Abstract. The analytic continuation method of homogenization theory provides Stieltjes integral representations for the effective parameters of composite media, involving spectral measures of self-adjoint random operators which depend only on the composite geometry. On finite bond lattices, these random operators are represented by random matrices and the spectral measures are given explicitly in terms of their eigenvalues and eigenvectors. Here provide the mathematical foundation for rigorous computation of spectral measures for such composite media. We also introduce a large family of random bond lattices and directly compute the associated spectral measures and effective parameters. The computed spectral measures agree with known theoretical results, and the behavior of the effective parameters is shown to be consistent with rigorous bounds.

**Key words.** composite materials, random resistor network, percolation, homogenization, spectral measure

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1. Introduction Over the years a broad range of mathematical techniques have been developed that reduce the analysis of complex composite materials, with rapidly varying structures in space, to solving averaged, or homogenized equations that do not have rapidly varying data, and involve an effective parameter. Homogenization for the effective parameter problem in composite media with rapidly varying coefficients of thermal conductivity, or equivalently [33] electrical conductivity and permittivity, and magnetic permeability, was developed by Papanicolaou and Varadhan [37] for the steady state, static case. This work was extended [22, 23] by Golden and Papanicolaou to the quasi-static frequency dependent case with complex parameters. Analysis of the effective dielectric problem for the fully frequency dependent case described by the Helmholtz equation is given in [42].

The effective parameter problem for two-component media in the quasi-static limit was developed by Bergman [3], Milton [30], and Golden and Papanicolaou [22], leading to Stieltjes integral representations for the effective parameters of composite media. The Golden-Papanicolaou formulation of this analytic continuation method (ACM) is based on the spectral theorem and resolvent formulas involving random self-adjoint operators. This formulation demonstrated that the measures underlying these integral representations are spectral measures associated with these random operators, which depend only on the composite geometry. These measures contain all the information about the mixture geometry, and provide a link between microgeometry and transport. Local geometry is encoded in "geometric" resonances in the measures [27], while global connectivity is encoded by spectral gaps in the measure at the spectral endpoints [34, 27]. A remarkable feature of the method is that once the spectral measures are found for a given composite geometry, by the symmetries in the governing equations [33],

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the effective electrical, magnetic, and thermal transport properties are *all* completely determined by these measures.

The integral representations yield rigorous forward bounds on the effective parameters of composites, given partial information on the microgeometry [3, 30, 22, 4]. One can also use the integral representations to obtain inverse bounds, where data on the electromagnetic response of a sample, for example, is used to bound its structural parameters, such as the volume fractions of the components [9, 7, 10, 44, 5, 8, 11, 21], and even the separation of the inclusions in matrix particle composites [36]. For classes of composites which undergo a percolation transition, the integral representations have been used to obtain detailed information regarding the critical behavior of the effective parameters in the scaling regime [19, 34]. Furthermore, the relationship between the effective parameters and the system energy has also led to a physically consistent statistical mechanics model for two-component dielectric media which is also mathematically tractable [35].

Despite the many applications which have stemmed from the ACM, explicit computations of the effective parameters have been obtained for only a handful of composite microstructures. To help overcome this limitation, here we develop a mathematical framework which provides a rigorous way to directly compute the spectral measures and effective parameters for finite lattice composite microstructures. We introduce a large class of locally isotropic, statistically isotropic, and anisotropic random bond networks and directly compute the associated spectral measures and effective parameters. (FINISH THIS PARAGRAPH WHEN THE MATHEMATICAL METHODS AND RESULTS SECTION IS FINISHED).

- 2. Mathematical Methods We now formulate the effective parameter problem for random two-phase conductive media in the lattice and continuum settings. In Section 2.1 we review the ACM for the continuum setting [22], while the *infinite* lattice setting [6, 18] is reviewed in Section 2.2.1. The mathematical framework underlying the infinite lattice case is directly analogous to that of the continuum case [6], and the integral representation for the effective parameters follow with minor modifications in the theory. In the *finite* lattice setting, the integral representations for the effective parameters are analogous to that of the continuum and infinite lattice cases. However, significant modifications must be made to the mathematical framework. A key theoretical contribution of this work is the formulation of the ACM for the finite lattice case, which is discussed in Section 2.2.2. In order to streamline the presentation of the ACM given in this section, we have placed many of the technical details in an appendix. The statement of Theorem A-1.1 of Section A-1.4.2 summarizes the formulation of the ACM for the finite lattice setting.
- **2.1. Continuum Setting** Consider a two-phase random conductive medium filling all of  $\mathbb{R}^d$ , which is determined by the probability space  $(\Omega, P)$ . Here  $\Omega$  is the set of all geometric realizations of our random medium, which is indexed by the parameter  $\omega \in \Omega$  representing one particular geometric realization, and P is the associated probability measure. Let  $\sigma(\vec{x},\omega)$  be the local complex conductivity tensor associated with the conductive medium, which has components  $\sigma_{jk}(\vec{x},\omega)$ ,  $j,k=1,\ldots,d$ , which are (spatially) stationary random fields (see Section A-1.1 for details).

Consider the Hilbert space  $\mathscr{H} = \bigotimes_{i=1}^d L^2(\Omega, P)$  with inner product  $\langle \cdot, \cdot \rangle$  defined by  $\langle \vec{\xi}, \vec{\zeta} \rangle = \langle \vec{\xi} \cdot \vec{\zeta} \rangle$ , where  $\vec{\xi} \cdot \vec{\zeta} = \vec{\xi}^T \vec{\zeta}$  denotes the dot-product on  $\mathbb{R}^d$  and  $\langle \cdot \rangle$  means ensemble average over  $\Omega$  or, by an ergodic theorem [22], spatial average over all of  $\mathbb{R}^d$ . Define the Hilbert spaces [22] of "curl free"  $\mathscr{H}_{\times} \subset \mathscr{H}$  and "divergence free"  $\mathscr{H}_{\bullet} \subset \mathscr{H}$  random

fields (see Section A-1.1 for details)

$$\mathcal{H}_{\times} = \left\{ \vec{Y} \in \mathcal{H} \mid \vec{\nabla} \times \vec{Y} = 0 \text{ weakly and } \langle \vec{Y} \rangle = 0 \right\},$$

$$\mathcal{H}_{\bullet} = \left\{ \vec{Y} \in \mathcal{H} \mid \vec{\nabla} \cdot \vec{Y} = 0 \text{ weakly and } \langle \vec{Y} \rangle = 0 \right\},$$
(2.1)

and consider the following variational problems [22]. Find  $\vec{E}_f \in \mathcal{H}_{\times}$  and  $\vec{J}_f \in \mathcal{H}_{\bullet}$  such that

$$\langle \boldsymbol{\sigma}(\vec{E}_0 + \vec{E}_f) \cdot \vec{Y} \rangle = 0 \quad \forall \ \vec{Y} \in \mathcal{H}_{\times} \quad \text{and} \quad \langle \boldsymbol{\rho}(\vec{J}_0 + \vec{J}_f) \cdot \vec{Y} \rangle = 0 \quad \forall \ \vec{Y} \in \mathcal{H}_{\bullet}, \quad (2.2)$$

respectively. When the bilinear forms  $\Psi(\vec{\xi}, \vec{\zeta}) = \sigma \vec{\xi} \cdot \vec{\zeta}$  and  $\Phi(\vec{\xi}, \vec{\zeta}) = \rho \vec{\xi} \cdot \vec{\zeta}$  are bounded and coercive, these problems have a unique solution [22, 37] satisfying the quasi-static limit of Maxwell's equations [26]

$$\vec{\nabla} \times \vec{E} = 0, \quad \vec{\nabla} \cdot \vec{J} = 0, \quad \vec{J} = \sigma \vec{E}, \quad \langle \vec{E} \rangle = \vec{E}_0,$$

$$\vec{\nabla} \times \vec{E} = 0, \quad \vec{\nabla} \cdot \vec{J} = 0, \quad \vec{E} = \rho \vec{J}, \quad \langle \vec{J} \rangle = \vec{J}_0.$$

$$(2.3)$$

Here,  $\vec{E}(\vec{x},\omega) = \vec{E}_0 + \vec{E}_f(\vec{x},\omega)$  is the random electric field, where  $\vec{E}_0 = \langle \vec{E} \rangle$  and  $\vec{E}_f$  is the fluctuating field of mean zero about the (constant) average  $\vec{E}_0$ . Similarly,  $\vec{J}(\vec{x},\omega) = \vec{J}_0 + \vec{J}_f(\vec{x},\omega)$  is the random current density. Moreover,  $\vec{E}_f$  and  $\vec{J}_f$  are stationary random fields [22].

As  $\vec{E}_f \in \mathscr{H}_{\times}$  and  $\vec{J}_f \in \mathscr{H}_{\bullet}$ , equation (2.2) yields the energy (power) [26] constraints  $\langle \vec{J} \cdot \vec{E}_f \rangle = 0$  and  $\langle \vec{E} \cdot \vec{J}_f \rangle = 0$ , respectively, which leads to the following reduced energy representations  $\langle \vec{J} \cdot \vec{E} \rangle = \langle \vec{J} \rangle \cdot \vec{E}_0$  and  $\langle \vec{E} \cdot \vec{J} \rangle = \langle \vec{E} \rangle \cdot \vec{J}_0$ . The effective complex conductivity and resistivity tensors,  $\sigma^*$  and  $\rho^*$ , are defined by

$$\langle \vec{J} \rangle = \boldsymbol{\sigma}^* \vec{E}_0 \quad \text{and} \quad \langle \vec{E} \rangle = \boldsymbol{\rho}^* \vec{J}_0.$$
 (2.4)

Consequently, we have the following energy representations involving the effective parameters  $\langle \vec{J} \cdot \vec{E} \rangle = \sigma^* \vec{E}_0 \cdot \vec{E}_0 = \rho^* \vec{J}_0 \cdot \vec{J}_0$ . We assume that the composite is a locally isotropic medium so that  $\sigma_{jk}(\vec{x},\omega) = \sigma(\vec{x},\omega)\delta_{jk}$  and  $\rho_{jk}(\vec{x},\omega) = \rho(\vec{x},\omega)\delta_{jk}$ , where  $\delta_{jk}$  is the Kronecker delta and  $j,k=1,\ldots,d$ . We further assume that the composite is a two-component medium, so that  $\sigma(\vec{x},\omega)$  takes the complex values  $\sigma_1$  and  $\sigma_2$ , and  $\rho(\vec{x},\omega)$  takes the values  $1/\sigma_1$  and  $1/\sigma_2$ , and satisfy [22]

$$\sigma(\vec{x},\omega) = \sigma_1 \chi_1(\vec{x},\omega) + \sigma_2 \chi_2(\vec{x},\omega), \qquad \rho(\vec{x},\omega) = \chi_1(\vec{x},\omega)/\sigma_1 + \chi_2(\vec{x},\omega)/\sigma_2. \tag{2.5}$$

Here  $\chi_i(\vec{x},\omega)$  is the characteristic function of medium i=1,2, which equals one for all  $\omega \in \Omega$  having medium i at  $\vec{x}$  and zero otherwise, with  $\chi_1 = 1 - \chi_2$ . For simplicity, we focus on one component of these tensors,  $\sigma_{ik}^*$  and  $\rho_{ik}^*$ , for some  $j, k=1,\ldots,d$ .

focus on one component of these tensors,  $\sigma_{jk}^*$  and  $\rho_{jk}^*$ , for some  $j,k=1,\ldots,d$ . Due to the homogeneity of these functions, e.g.  $\sigma_{jk}^*(a\sigma_1,a\sigma_2)=a\sigma_{jk}^*(\sigma_1,\sigma_2)$  for any complex number a, they depend only on the ratio  $h=\sigma_1/\sigma_2$ , and we define the tensor-valued functions  $\mathbf{m}(h)=\boldsymbol{\sigma}^*/\sigma_2$ ,  $\mathbf{w}(z)=\boldsymbol{\sigma}^*/\sigma_1$ ,  $\tilde{\mathbf{m}}(h)=\sigma_1\boldsymbol{\rho}^*$ , and  $\tilde{\mathbf{w}}(z)=\sigma_2\boldsymbol{\rho}^*$  with components

$$m_{jk}(h) = \sigma_{jk}^*/\sigma_2, \quad w_{jk}(z) = \sigma_{jk}^*/\sigma_1, \quad \tilde{m}_{jk}(h) = \sigma_1 \rho_{jk}^*, \quad \tilde{w}_{jk}(z) = \sigma_2 \rho_{jk}^*.$$
 (2.6)

where z = 1/h. The dimensionless functions  $m_{jk}(h)$  and  $\tilde{m}_{jk}(h)$  are analytic off the negative real axis in the h-plane, while  $w_{jk}(z)$  and  $\tilde{w}_{jk}(z)$  are analytic off the negative

real axis in the z-plane [22]. Each take the corresponding upper half plane to the upper half plane and are therefore examples of Herglotz functions [12, 22]. A key step in the ACM is obtaining Stieltjes integral representations for  $\sigma^*$  and  $\rho^*$ . These follow from resolvent representations for the electric field [22] and current density [34] (see Section A-1.2 for details)

$$\begin{split} \vec{E} &= s(sI - \Gamma \chi_1)^{-1} \vec{E}_0 = t(tI - \Gamma \chi_2)^{-1} \vec{E}_0, \quad s \in \mathbb{C} \setminus [0, 1], \\ \vec{J} &= t(tI - \Upsilon \chi_1)^{-1} \vec{J}_0 = s(sI - \Upsilon \chi_2)^{-1} \vec{J}_0, \quad t \in \mathbb{C} \setminus [0, 1], \end{split} \tag{2.7}$$

where I is the identity operator on  $\mathbb{R}^d$  and we have defined the complex variables s=1/(1-h) and t=1/(1-z)=1-s. The operator  $\Gamma=\vec{\nabla}(\Delta^{-1})\vec{\nabla}\cdot$  is an orthogonal projection from  $\mathscr{H}$  onto the Hilbert space  $\mathscr{H}_{\times}$  of curl-free random fields, and is based on convolution with the free-space Green's function for the Laplacian  $\Delta=\nabla\cdot\nabla=\nabla^2$  [22, 34]. Similarly, the operator  $\Upsilon=-\vec{\nabla}\times(\Delta^{-1})\vec{\nabla}\times$  is an orthogonal projection from  $\mathscr{H}$  onto the Hilbert space  $\mathscr{H}_{\bullet}$  of divergence-free random fields, involving the vector Laplacian  $\Delta=-\vec{\nabla}\times\vec{\nabla}\times+\vec{\nabla}\vec{\nabla}\cdot$  (see Section A-1.2 for details).

It is more convenient to consider the functions  $F_{jk}(s) = \delta_{jk} - m_{jk}(h)$  and  $E_{jk}(s) = \delta_{jk} - \tilde{m}_{jk}(h)$  which are analytic off [0,1] in the s-plane, and  $G_{jk}(t) = \delta_{jk} - w_{jk}(z)$  and  $H_{jk}(t) = \delta_{jk} - \tilde{w}_{jk}(z)$  which are analytic off [0,1] in the t-plane [22]. For the formulation of the effective parameter problem involving  $\mathscr{H}_{\times}$  and  $\sigma^*$ , define the coordinate system so that in (2.4) the constant vector  $\vec{E}_0$  is given by  $\vec{E}_0 = E_0 \vec{e}_j$ , where  $\vec{e}_j$  is a standard basis vector on  $\mathbb{R}^d$  in the  $j^{\text{th}}$  direction for some  $j=1,\ldots,d$ . In the other formulation involving  $\mathscr{H}_{\bullet}$  and  $\rho^*$  define  $\vec{J}_0 = J_0 \vec{e}_j$ . Also, in (2.5) write  $\sigma = \sigma_2(1-\chi_1/s) = \sigma_1(1-\chi_2/t)$  and  $\rho = (1-\chi_2/s)/\sigma_1 = (1-\chi_1/t)/\sigma_2$ . Equations (2.4) and (2.7), and the spectral theorem [38, 43] then yield the following integral representations [22, 2, 4, 34] for the effective parameters  $\sigma^*_{jk}$  and  $\rho^*_{jk}$ 

$$m_{jk}(h) = \delta_{jk} - F_{jk}(s), \qquad F_{jk}(s) = \langle \chi_1(sI - \Gamma\chi_1)^{-1}\vec{e}_j \cdot \vec{e}_k \rangle = \int_0^1 \frac{\mu_{jk}(d\lambda)}{s - \lambda}, \qquad (2.8)$$

$$w_{jk}(z) = \delta_{jk} - G_{jk}(t), \qquad G_{jk}(t) = \langle \chi_2(tI - \Gamma\chi_2)^{-1}\vec{e}_j \cdot \vec{e}_k \rangle = \int_0^1 \frac{\alpha_{jk}(d\lambda)}{t - \lambda},$$

$$\tilde{m}_{jk}(h) = \delta_{jk} - E_{jk}(s), \qquad E_{jk}(s) = \langle \chi_1(sI - \Upsilon\chi_1)^{-1}\vec{e}_j \cdot \vec{e}_k \rangle = \int_0^1 \frac{\eta_{jk}(d\lambda)}{s - \lambda},$$

$$\tilde{w}_{jk}(z) = \delta_{jk} - H_{jk}(t), \qquad H_{jk}(t) = \langle \chi_2(tI - \Upsilon\chi_2)^{-1}\vec{e}_j \cdot \vec{e}_k \rangle = \int_0^1 \frac{\kappa_{jk}(d\lambda)}{t - \lambda}.$$

Here  $\mu_{jk}$  and  $\alpha_{jk}$  are spectral measures associated with the random operators  $\chi_1\Gamma\chi_1$  and  $\chi_2\Gamma\chi_2$ , respectively, while  $\eta_{jk}$  and  $\kappa_{jk}$  are spectral measures associated with the random operators  $\chi_2\Upsilon\chi_2$  and  $\chi_1\Upsilon\chi_1$ , respectively (see Section A-1.4 for details).

It can be seen through the Stieltjes–Perron inversion theorem [25, 33] that the measures  $\mu_{jk}$  and  $\alpha_{jk}$ , and the measures  $\eta_{jk}$  and  $\kappa_{jk}$  are related by [34] (see Section A-1.3 for details)

$$\lambda \alpha_{jk}(\lambda) = (1 - \lambda)\mu_{jk}(1 - \lambda) + \lambda (m_{jk}(0)\delta_0(d\lambda) + w_{jk}(0)(\lambda - 1)\delta_1(d\lambda)),$$

$$\lambda \kappa_{jk}(\lambda) = (1 - \lambda)\eta_{jk}(1 - \lambda) + \lambda (\tilde{m}_{jk}(0)\delta_0(d\lambda) + \tilde{w}_{jk}(0)(\lambda - 1)\delta_1(d\lambda)),$$
(2.9)

where  $\delta_a(d\lambda)$  is the delta measure concentrated at  $\lambda = a$ . Equations (2.8) and (2.9) demonstrate the many symmetries between the functions  $m_{jk}(h)$ ,  $w_{jk}(z)$ ,  $\tilde{m}_{jk}(h)$ , and  $\tilde{w}_{jk}(z)$ , and the respective measures  $\mu_{jk}$ ,  $\alpha_{jk}$ ,  $\eta_{ij}$ , and  $\kappa_{jk}$ . Because of these

symmetries, for simplicity, we will focus on  $m_{jk}(h)$  and  $\mu_{jk}$ , and will reintroduce the other functions and measures where appropriate.

A key feature of equations (2.4), (2.6), and (2.8) is that the parameter information in h and  $E_0$  is separated from the geometry of the composite, which is encoded in the spectral measure  $\mu_{jk}$  via its moments  $\mu_{jk}^n = \int_0^1 \lambda^n \mu_{jk}(d\lambda)$ ,  $n = 0, 1, 2, \ldots$  For example, the mass  $\mu_{jk}^0$  of the measure  $\mu_{jk}$  is given by  $\mu_{jk}^0 = p_1 \delta_{jk}$  (see equation (A-6)), where  $p_1 = \langle \chi_1 \rangle$  is the volume fraction of material component 1. Moreover, if the medium is statistically isotropic then  $\mu_{jk}^1 = (p_1 p_2/d) \delta_{jk}$ , where  $p_2 = 1 - p_1 = \langle \chi_2 \rangle$  is the volume fraction of material component 2 [6, 20].

A principal application of the ACM is to derive forward bounds on the diagonal components  $\sigma_{kk}^*$  of the tensor  $\sigma^*$ , k=1,...,d, given partial information on the microgeometry [3, 30, 22, 4]. This information may be given in terms of the (n+1)-point correlation functions of the medium, or equivalently the moments  $\mu_{kk}^n$ , n=0,1,2,..., of the measure  $\mu_{kk}$  [32, 22]. Given this information, the bounds on  $\sigma_{kk}^*$  follow from the special structure of  $F_{kk}(s)$  in (2.8). Specifically, it is a linear functional of the positive measure  $\mu_{kk}$ . The bounds are obtained by fixing the contrast parameter s, varying over an admissible set of measures  $\mu_{kk}$  (or geometries) which is determined by the known information regarding the two-component composite. Knowledge of the moments  $\mu_{kk}^n$  for n=1,...,J confines  $\sigma_{kk}^*$  to a region of the complex plane which is bounded by arcs of circles, and the region becomes progressively smaller as more moments are known [32, 17]. When all the moments are known the measure  $\mu_{kk}$  is uniquely determined [1], hence  $\sigma_{kk}^*$  is explicitly known. For your convenience, the bounding procedure is reviewed in Section A-1.5.

- **2.2. Lattice Setting** In this section we formulate the effective parameter problem for the infinite and finite, two-component bond lattice on  $\mathbb{Z}^d$ . The infinite bond lattice, reviewed in Section 2.2.1, is a special case of the stationary random medium considered in Section 2.1. In Section 2.2.2 we develop the mathematical framework for the ACM in the finite lattice setting.
- **2.2.1.** Infinite Lattice Setting Consider a two-component bond lattice on all of  $\mathbb{Z}^d$  determined by the probability space  $(\Omega, P)$ , and let  $\sigma(\vec{x}, \omega)$  be the local complex conductivity tensor with components  $\sigma_{jk}(\vec{x}, \omega) = \sigma^j(\vec{x}, \omega)\delta_{jk}$ , j, k = 1, ..., d. Here  $\sigma^j(\vec{x}, \omega)$  is the conductivity of the bond emanating from  $\vec{x} \in \mathbb{Z}^d$  in the positive  $j^{\text{th}}$  direction, which is a stationary random field that takes the *complex* values  $\sigma_1$  and  $\sigma_2$  with probabilities  $p_1$  and  $p_2 = 1 p_1$ , respectively [18, 6]. The configuration space  $\Omega = {\sigma_1, \sigma_2}^{d\mathbb{Z}^d}$  represents the set of all realizations of the random medium and the probability measure P is compatible with stationarity, as defined in Section A-1.2. Analogous to equation (2.5), the local conductivity  $\sigma^j(\vec{x}, \omega)$  of the two-phase random medium takes the form [18]

$$\sigma^{j}(\vec{x},\omega) = \sigma_1 \chi_1^{j}(\vec{x},\omega) + \sigma_2 \chi_2^{j}(\vec{x},\omega), \quad j = 1, \dots, d.$$
 (2.10)

Here  $\chi_i^j(\vec{x},\omega)$  is the characteristic function of medium i=1,2, which equals one for all realizations  $\omega \in \Omega$  having medium i in the  $j^{\text{th}}$  positive bond at  $\vec{x}$ , and equals zero otherwise

In this lattice setting, the differential operators  $\vec{\nabla} \times$  and  $\vec{\nabla} \cdot$  in equation (2.3) are given [18, 6] in terms of forward and backward difference operators  $D_j^+$  and  $D_j^-$ , respectively, where

$$D_j^+ = T_j^+ - I, \quad D_j^- = I - T_j^-, \quad j = 1, ..., d.$$
 (2.11)

Here I is the identity operator on  $\mathbb{Z}^d$ , and  $T_j^+ = T_{+e_j}$  and  $T_j^- = T_{-e_j}$  are the generators (through composition) of the unitary group  $T_x$  acting on  $L^2(\Omega,P)$  defined by  $(T_x f)(0,\omega) = f(\vec{x},\omega)$  for any  $f \in L^2(\Omega,P)$ ,  $f: \mathbb{Z}^d \times \Omega \to \mathbb{R}$ , which is a stationary random field [18] (see Section A-1.1 for details). Define  $\mathscr{H} = \bigotimes_{i=1}^d L^2(\Omega,P)$  and let  $\vec{E}, \vec{J} \in \mathscr{H}$  be the random electric field and current density, respectively, where  $\vec{E}(\vec{x},\omega) = (E^1(\vec{x},\omega), \dots E^d(\vec{x},\omega))$  and  $E^j(\vec{x},\omega)$  is the electric field in the bond emanating from  $\vec{x}$  in the positive  $j^{\text{th}}$  direction, and similarly for  $\vec{J}(\vec{x},\omega)$ .

As in Section 2.1 we write  $\vec{E} = \vec{E}_0 + \vec{E}_f$ , where  $\vec{E}_f$  is the fluctuating field of mean zero about the (constant) average  $\vec{E}_0$ . The variational problem in (2.2) for this lattice setting has a unique solution satisfying Kirchhoff's circuit laws [22, 6]

$$D_i^+ E^j - D_j^+ E^i = 0, \quad \sum_{k=1}^d D_k^- J^k = 0, \quad J^i = \sigma^i E^i, \quad \langle \vec{E} \rangle = \vec{E}_0,$$
 (2.12)

where i, j = 1, ..., d and the components  $E^i(\vec{x}, \omega)$  and  $J^i(\vec{x}, \omega)$  of  $\vec{E}(\vec{x}, \omega)$  and  $\vec{J}(\vec{x}, \omega)$ , respectively, are stationary random fields. Equation (2.12) is a direct analogue of equation (2.3) when written in component form [22]. Analogous to equation (2.4), the effective complex conductivity tensor  $\sigma^*$  is defined by  $\langle \vec{J} \rangle = \sigma^* \vec{E}_0$ , which has components  $\sigma^*_{jk} = \sigma_2 m_{jk}(h)$ , j,k = 1,...,d, where  $h = \sigma_1/\sigma_2$ . All of the results stated in Section 2.1, including the representation formulas for  $m_{jk}(h)$  and  $w_{jk}(z)$  in (2.8), still hold in this infinite lattice setting with  $\Gamma$  in (2.7) now given by

$$\Gamma = \nabla^{+}(\Delta^{-1})\nabla^{-}, \quad \nabla^{\pm} = (D_{1}^{\pm}, \dots, D_{d}^{\pm}),$$
 (2.13)

where  $\Delta^{-1}$  is based on discrete convolution with the lattice Green's function for the Laplacian  $\Delta = \nabla^2$  [6]. The formulation of the ACM for the effective resistivity tensor  $\rho^*$  in the infinite lattice setting is a direct analogue of that for  $\sigma^*$  given here (see Section 2.2.2 for more details).

**2.2.2. Finite Lattice Setting** Consider a finite, two-component bond lattice on  $\mathbb{Z}^d_L \subset \mathbb{Z}^d$  determined by the probability space  $(\Omega, P)$ , where

$$\mathbb{Z}_L^d = \{ \vec{x} \in \mathbb{Z}^d \mid 1 \le x_i \le L, \ i = 1, ..., d \},$$
(2.14)

 $L\in\mathbb{N},\,L\geq 2,\,$  and  $x_i=(\vec{x})_i$  is the  $i^{\,\mathrm{th}}$  component of the vector  $\vec{x}$ . Let  $\boldsymbol{\sigma}(\vec{x},\omega)$  be the local complex conductivity tensor with components  $\sigma_{jk}(\vec{x},\omega)=\sigma^j(\vec{x},\omega)\delta_{jk},\,\,j,k=1,\ldots,d,$  where  $\sigma^j(\vec{x},\omega)$  is defined in equation (2.10) for  $\vec{x}\in\mathbb{Z}_L^d$  and  $\omega\in\Omega$ . The configuration space  $\Omega=\{\sigma_1,\sigma_2\}^{d\mathbb{Z}_L^d}$  represents the set of all  $2^N$  realizations of the finite random lattice, where  $N=dL^d$ , and P is the associated probability measure. Define  $\mathscr{H}=\bigotimes_{i=1}^d L^2(\Omega,P)$  and let  $\vec{E},\vec{J}\in\mathscr{H}$  be the random electric field and current density, respectively, which satisfy Kirchhoff's circuit laws (2.12) with appropriate boundary conditions. Analogous to equation (2.4), the effective complex conductivity tensor  $\boldsymbol{\sigma}^*$  is defined by  $\langle \vec{J} \rangle = \boldsymbol{\sigma}^* \vec{E}_0$ , which has components  $\sigma_{jk}^* = \sigma_2 m_{jk}(h), \ \vec{E}_0 = \langle \vec{E} \rangle$ , and  $\langle \cdot \rangle$  denotes ensemble average over  $\Omega$ .

In this section we obtain a discrete version of the integral representation for  $m_{jk}(h)$  in (2.8) for this finite bond lattice setting, involving a spectral measure  $\mu_{jk}$  associated with a real-symmetric random matrix. Toward this goal, we define a bijective mapping  $\Theta$  from the two-dimensional set  $\mathbb{Z}_L^d$  onto the one dimensional set  $\mathbb{N}_L \subset \mathbb{N}$ 

given by

$$\mathbb{N}_{L} = \{ i \in \mathbb{N} \mid i \leq dL^{d} \}, \qquad \Theta(\vec{x}) = x_{1} + \sum_{k=2}^{d} (x_{k} - 1)L^{k-1}.$$
 (2.15)

Under the bijection  $\Theta$  the components  $E^j(\vec{x},\omega)$ ,  $j=1,\ldots,d$ , of the random electric field  $\vec{E}(\vec{x},\omega)=(E^1(\vec{x},\omega),\ldots,E^d(\vec{x},\omega))$  are mapped to vector valued functions  $E^j(\vec{x},\omega)\mapsto \vec{E}^j(\omega)=(E^j_1(\omega),\ldots,E^j_{L^d}(\omega))$  so that

$$\Theta(\vec{E}(\vec{x},\omega)) = (\vec{E}^1(\omega), \dots, \vec{E}^d(\omega)) \in \mathbb{R}^N$$
(2.16)

for each  $\omega \in \Omega$ , and similarly for  $\vec{J}(\vec{x},\omega)$ . Moreover, the bijection  $\Theta$  maps the standard basis vector  $\vec{e}_1 = (1,0,\dots,0) \in \mathbb{Z}^d$ , for example, to the vector  $(\vec{1},\vec{0},\dots,\vec{0}) \in \mathbb{Z}^N$ , where  $\vec{1}$  and  $\vec{0}$  are vectors of ones and zeros of length  $L^d$ , respectively, and similarly for the  $\vec{e}_j$  for  $j=2,\dots,d$ . Therefore, the vectors  $\hat{e}_i = \Theta(\vec{e}_i)/L^{d/2}, i=1,\dots,d$ , serve as the standard basis vectors on  $\mathbb{N}_L$ , with  $\hat{e}_i \cdot \hat{e}_j = \delta_{ij}$ .

On  $\mathbb{N}_L$  the difference operators  $D_j^{\pm}$ ,  $j=1,\ldots,d$ , in equation (2.11) are given in terms of finite difference matrices  $D_j$  [13]. Moreover, the Laplacian  $\Delta$  and the projection operator  $\Gamma$  in (2.13) are replaced by the real-symmetric matrices  $\Delta = \nabla^T \nabla$  and  $\Gamma = \nabla (\Delta^{-1}) \nabla^T$ , respectively, where  $\nabla = (D_1, \ldots, D_d)^T$ . The matrices  $\Delta$  and  $\Gamma$  depend only on the topology and the boundary conditions of the underlying finite bond lattice  $\mathbb{Z}_L^d$ , and  $\Gamma$  is a projection matrix satisfying  $\Gamma^2 = \Gamma$ . By writing the constitutive relation  $J^j(\vec{x},\omega) = \sigma^j(\vec{x},\omega)E^j(\vec{x},\omega)$  displayed in equation (2.12) as  $J^j(\vec{x},\omega) = \sigma_2(1-\chi_1^j(\vec{x},\omega)/s)E^j(\vec{x},\omega)$ , we see that the characteristic function  $\chi_1^j(\vec{x},\omega)$  in (2.10) operates, according to the probability measure P, on the electric field  $E^j(\vec{x},\omega)$  in each individual bond  $j=1,\ldots,d$  emanating from  $\vec{x}\in\mathbb{Z}_L^d$ . In view of this and equation (2.16), on  $\mathbb{N}_L$  the characteristic function  $\chi_1^j(\vec{x},\omega)$  is represented by a block diagonal matrix

$$\chi_1(\omega) = \operatorname{diag}(\chi_1^1(\omega), \dots, \chi_1^d(\omega)), \tag{2.17}$$

where  $\chi_1^j(\omega)$ ,  $j=1,\ldots,d$ , is a diagonal matrix of size  $L^d \times L^d$  with zeros and ones distributed according to P along the main diagonal. Moreover, the matrix  $\chi_1^j(\omega)$  acts on the vector  $\vec{E}^j(\omega) = \Theta(E^j(\vec{x},\omega))$  in (2.16) for each  $j=1,\ldots,d$ . Consequently,  $\chi_1(\omega)$  is also a real-symmetric projection matrix of size  $N \times N$ , which determines the geometry and component connectivity of the two-phase random medium. In summary, on  $\mathbb{N}_L$  the operator  $M_1 = \chi_1 \Gamma \chi_1$  is represented by a real-symmetric random matrix of size  $N \times N$  [21, 34].

In Section A-1.4.1 we discuss how the spectral theorem under the ACM leads to the Stieltjes integral representations for the effective parameter in equation (2.8), for the continuum and infinite lattice settings. In Section A-1.4.2 we discuss how these spectral methods must be modified for the finite lattice setting discussed in this section, to obtain the following Stieltjes integral representation for the effective conductivity  $\sigma^*$ , which has components  $\sigma_{jk}^* = \sigma_2 m_{jk}(h)$ , j,k = 1,...,d, that satisfy

$$m_{jk}(h) = \delta_{jk} - F_{jk}(s), \quad F_{jk}(s) = \int_0^1 \frac{\mu_{jk}(d\lambda)}{s - \lambda}, \quad \mu_{jk}(d\lambda) = \sum_{i=1}^N \langle \delta_{\lambda_i}(d\lambda) \chi_1 R_i \hat{e}_j \cdot \hat{e}_k \rangle,$$
(2.18)

and a similar formula for  $w_{jk}(z) = \delta_{jk} - G_{jk}(t)$  involving a spectral measure  $\alpha_{jk}$ . Here we have defined  $h = \sigma_1/\sigma_2$ , s = 1/(1-h),  $R_i = \vec{u}_i \vec{u}_i^T$  for i = 1, ..., N,  $\vec{u}_i$  is the  $i^{\text{th}}$  eigenvector of the matrix  $M_1$  with associated eigenvalue  $\lambda_i$ ,  $\delta_{\lambda_i}(d\lambda)$  is the delta measure concentrated at  $\lambda_i$ , and  $\langle \cdot \rangle$  denotes ensemble average over  $\Omega$ . Moreover, the mass  $\mu_{jk}^0$  of the spectral measure  $\mu_{jk}$  is given by

$$\mu_{jk}^{0} = \langle \chi_{1} \hat{e}_{k} \cdot \hat{e}_{k} \rangle \, \delta_{jk} = d \, \frac{\langle N_{1}^{k}(\omega) \rangle}{N} \, \delta_{jk}$$

where  $N_1^k(\omega) = \text{Trace}(\chi_1^k(\omega))$  is the total number of type-one bonds in the positive  $k^{\text{th}}$  direction for  $\omega \in \Omega$ . Furthermore, in the case of locally isotropic random media  $\mu_{jk}^0 = p_1 \delta_{jk}$ , where  $p_1$  is the number fraction of type-one bonds (see Section A-1.4.2 for details).

- 3. Numerical Results Eigenvalue densities, Spectral measures, forward bounds on effective parameters. What the new data and bounds tell us about the transitional behavior of the multiscale sea ice structures. In this section we discuss how the theoretical framework developed in Sections 2.2.2 and A-1.4.2 can be used to directly calculate the spectral measure  $\mu$ , hence the effective parameters for various composite microstructures. In Section ?? we calculate  $\mu$  and
- **4. Conclusion** What's new in this electromagnetic/thermal case and what can it tell us about sea ice? How could this potentially allow small scale processes to be up-scaled into coarsened climate models? How can the electromagnetic response of sea ice provide information regarding the fluid flow processes? Inverse problems. Spectral measures for the effective elasticity tensor of the ice pack....

## A-1. Appendix

In this section we provide many of the details to, and derivations of the formulas given in Section 2. Much of this appendix is devoted to reviewing well established results, in order to clarify the technical details underlying the mathematical framework. However, a key theoretical contribution of this work is given in Theorem A-1.1 of Section A-1.4.2 below.

**A-1.1. Stationary Random Fields** A stationary random field  $f \in L^2(\Omega, P)$ ,  $f: \mathbb{R}^d \times \Omega \to \mathbb{R}$ , is a field such that the joint distribution of  $f(\vec{x}_1, \omega), \ldots, f(\vec{x}_n, \omega)$  and that of  $f(\vec{x}_1 + \vec{\xi}, \omega), \ldots, f(\vec{x}_n + \vec{\xi}, \omega)$  is the same for all  $\vec{\xi} \in \mathbb{R}^d$  and  $n \in \mathbb{N}$  [22]. In particular, the ensemble average  $\langle \cdot \rangle$  of  $f(\vec{x}, \omega)$  over  $\Omega$  is invariant under the translation group  $\tau_y: \Omega \to \Omega$  defined by  $f(\tau_{-y}\vec{x}, \omega) = f(\vec{x} + \vec{y}, \omega)$  for all  $\vec{x}, \vec{y} \in \mathbb{R}^d$ , with  $\tau_x \tau_y = \tau_{x+y}$ . Consequently,  $\langle f(\vec{x}, \omega) \rangle = \langle f(0, \omega) \rangle$  and we can focus on the origin and drop the  $\vec{x}$  notation by writing  $f(0, \omega) = f(\omega)$ , with  $f(\tau_{-x}\omega) = f(\vec{x}, \omega)$ . We shall assume that there is such a group of transformations that is one-to-one and preserves the measure P, i.e.  $P(\tau_x A) = P(A)$  for all P-measurable sets A [22, 37].

The group of transformations  $\tau_x$  acting on  $\Omega$  induces a group of operators  $T_x$  on the Hilbert space  $L^2(\Omega, P)$  defined by  $(T_x f)(\omega) = f(\tau_{-x} \omega)$  for all  $f \in L^2(\Omega, P)$ . Since  $\tau_x$  is measure preserving, the operators  $T_x$  form a unitary group and therefore have closed densely defined infinitesimal generators  $L_i$  in each direction i = 1, ..., d with domain  $\mathcal{J}_i \subset L^2(\Omega, P)$  [18]. Thus,

$$L_i = \frac{\partial}{\partial x_i} T_x \bigg|_{x=0}, \quad i = 1, \dots, d,$$

where  $x_i$  is the  $i^{\text{th}}$  component of the vector  $\vec{x}$  and differentiation is defined in the sense of convergence in  $L^2(\Omega,P)$  for elements of  $\mathscr{J}_i$  [18]. The closed subset  $\mathscr{J}=\bigcap_{i=1}^d\mathscr{J}_i$  of  $L^2(\Omega,P)$  is a Hilbert space [18] with inner product  $\langle\cdot,\cdot\rangle_D$  given by  $\langle f,g\rangle_D=\langle f,g\rangle+\sum_{i=1}^d\langle L_if,L_ig\rangle$ , where  $\langle\cdot,\cdot\rangle$  is the  $L^2(\Omega,P)$  inner product. (IS THERE A LIMIT THEOREM FROM LATTICE TO CONTINUUM THAT WE COULD CITE HERE?) In the case of a two-phase locally isotropic random medium, where the local fields can be non-differentiable at the phase boundaries, the Hilbert space  $\mathscr{H}_\times$  of "curl free" random fields given in equation (2.1), for example, is more precisely defined by [18]

$$\mathscr{H}_{\times} = \{Y_i(\omega) \in L^2(\Omega,P), \ i=1,\ldots,d \ | \ L_iY_j = L_jY_i \text{ weakly and } \langle Y_i \rangle = 0\}, \tag{A-1}$$

where  $j = 1, \dots, d$ .

**A-1.2. Resolvent Representations of Physical Fields** When the current density  $\vec{J}(\vec{x},\omega)$  and the electric field  $\vec{E}(\vec{x},\omega)$  are sufficiently smooth in a neighborhood of  $\vec{x} \in \mathbb{R}^d$  for  $\omega \in \Omega$ , equation (2.7) is obtained as follows. On the Hilbert space  $L^2(\Omega,P)$ , the operator  $\Delta^{-1}$  is well defined in terms of convolution with respect to the free space Green's function of the Laplacian  $\Delta = \vec{\nabla} \cdot \vec{\nabla}$  [22, 15]. Similarly, on the Hilbert space  $\mathscr{H} = \bigotimes_{i=1}^d L^2(\Omega,P)$ , the inverse  $\Delta^{-1}$  of the vector Laplacian  $\Delta$  is defined in terms of component-wise convolution with respect to the free space Green's function of the Laplacian.

Applying the integro-differential operator  $\vec{\nabla}(\Delta^{-1})$  to the formula  $\vec{\nabla} \cdot \vec{J} = 0$  in equation (2.3) yields  $\Gamma_{\bullet} \vec{J} = 0$ , where  $\Gamma_{\bullet} = \Gamma = \vec{\nabla}(\Delta^{-1})\vec{\nabla} \cdot$  is an orthogonal projection [22] from  $\mathscr{H}$  onto the Hilbert space  $\mathscr{H}_{\times}$  of curl-free random fields,  $\Gamma : \mathscr{H} \mapsto \mathscr{H}_{\times}$ . More

specifically, for every sufficiently smooth  $\vec{\zeta} \in \mathscr{H}_{\times}$  there exists [26] a scalar potential  $\varphi$  which is unique up to a constant such that  $\vec{\zeta} = \vec{\nabla} \varphi$ , so that  $\Gamma \vec{\zeta} = \vec{\zeta}$ .

Similarly we have  $\Upsilon \vec{E} = 0$ , where  $\Upsilon = \vec{\nabla} \times (\Delta^{-1}) \vec{\nabla} \times$  is an orthogonal projection from  $\mathscr{H}$  onto the Hilbert space [22]  $\mathscr{H}_{\bullet}$  of divergence-free random fields (of Coulomb or transverse gauge) [34]. This can be seen as follows. For every sufficiently smooth  $\vec{\zeta} \in \mathscr{H}_{\bullet}$  we have  $\vec{\zeta} = \vec{\nabla} \times (\vec{A} + \vec{C})$ , where  $\vec{A}$  is a vector potential associated with  $\vec{\zeta}$  and the vector  $\vec{C}$  satisfies  $\vec{\nabla} \times \vec{C} = 0$  [26]. The arbitrary vector  $\vec{C}$  can be chosen so that  $\vec{A}$  satisfies  $\vec{\nabla} \cdot \vec{A} = 0$  [26]. Hence,  $\vec{\nabla} \times \vec{\zeta} = \vec{\nabla} \times \vec{\nabla} \times \vec{A} = \vec{\nabla} (\vec{\nabla} \cdot \vec{A}) - \Delta \vec{A} = -\Delta \vec{A}$ . The vector  $\vec{C}$  chosen in this manner gives the Coulomb (or transverse) gauge of  $\vec{\zeta}$  [26]. Choosing the members of  $\mathscr{H}$  to have transverse gauge, the action of  $\Upsilon$  is given by  $\Upsilon = \vec{\nabla} \times (\vec{\nabla} \times \vec{\nabla} \times)^{-1} \vec{\nabla} \times = \vec{\nabla} \times (\Delta^{-1}) \vec{\nabla} \times$ , and it is clear that  $\Upsilon \vec{\zeta} = \vec{\zeta}$ .

When considering a two-phase locally isotropic random media, the local fields can be non-differentiable at the phase boundaries. In this case, the differential operators  $\nabla \times$ ,  $\nabla \cdot$ , and  $\nabla \cdot$  are given [22] in terms of the infinitesimal generators  $L_i$  defined in Section A-1.1, and the Hilbert space  $\mathscr{H}_{\times}$ , for example, is given by equation (A-1). The Hilbert space  $\mathscr{H}_{\bullet}$  is analogously defined.

We now derive the formulas in equation (2.7). Write  $\sigma$  and  $\rho$  in equation (2.5) as  $\sigma = \sigma_2(1 - \chi_1/s) = \sigma_1(1 - \chi_2/t)$  and  $\rho = (1 - \chi_2/s)/\sigma_1 = (1 - \chi_1/t)/\sigma_2$ . Note that  $\vec{E} = \vec{E}_0 + \vec{E}_f$ , where  $\vec{E}_0$  is a constant field and  $\vec{E}_f \in \mathscr{H}_{\times}$  so that  $\Gamma \vec{E} = \vec{E}_f$ , and similarly  $\Upsilon \vec{J} = \vec{J}_f$ . Consequently, from  $\Gamma \vec{J} = 0$  and  $\Upsilon \vec{E} = 0$  we have the following formulas which are equivalent to that in (2.7)

$$\vec{E}_f = \frac{1}{s} \Gamma \chi_1 \vec{E} = \frac{1}{t} \Gamma \chi_2 \vec{E}, \qquad \vec{J}_f = \frac{1}{t} \Upsilon \chi_1 \vec{J} = \frac{1}{s} \Upsilon \chi_2 \vec{J}. \tag{A-2}$$

It is important to note that the formulas  $\Gamma \vec{E} = \vec{E}_f$  and  $\Upsilon \vec{J} = \vec{J}_f$  are sufficient conditions for the energy constraints  $\langle \vec{J} \cdot \vec{E}_f \rangle = 0$  and  $\langle \vec{E} \cdot \vec{J}_f \rangle = 0$ , respectively, which follow from equation (2.2). These energy constraints are at the heart of the existence and uniqueness of solutions to equations (2.3) and (2.12) for the continuum and lattice settings, respectively. The sufficiency of these conditions can be seen by writing  $\sigma = \sigma_2(1 - \chi_1/s)$  in  $\vec{J} = \sigma \vec{E}$ , for example, to obtain

$$\langle \vec{J} \cdot \vec{E}_f \rangle = \sigma_2(\langle \vec{E} \cdot \vec{E}_f \rangle - \langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle / s), \tag{A-3}$$

for  $s \neq 0$   $(h \neq \pm \infty)$ . Now, if we have  $\Gamma \vec{E} = \vec{E}_f$  then  $\vec{\nabla} \cdot \vec{J} = 0$  yields equation (A-2)  $(\vec{E}_f = \Gamma \chi_1 \vec{E}/s)$ . Therefore, as  $\Gamma$  is a self-adjoint operator on  $\mathscr{H}$  [15], we have

$$\langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle = \langle \chi_1 \vec{E} \cdot \Gamma \vec{E} \rangle = \langle \Gamma \chi_1 \vec{E} \cdot \vec{E} \rangle = s \langle \vec{E}_f \cdot \vec{E} \rangle. \tag{A-4}$$

Consequently, from equation (A-3) we have  $\langle \vec{J} \cdot \vec{E}_f \rangle = 0$  for  $s \neq 0$ . The argument involving the operator  $\Upsilon$  and the vector field  $\vec{J}_f$  is analogous.

We conclude this section with some final remarks regarding the energy constraint  $\langle \vec{J} \cdot \vec{E}_f \rangle = 0$ , for example. We see from equation (A-3) that the energy constraint is equivalent to the following "field representation" for the contrast parameter  $s = \langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle / \langle \vec{E} \cdot \vec{E}_f \rangle$ , when  $\langle \vec{E} \cdot \vec{E}_f \rangle \neq 0$  (if and only if  $\langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle \neq 0$ ). Since h = 1 - 1/s and  $h \geq 0$  for  $h \in \mathbb{R}$ , this implies that  $|\langle \vec{E} \cdot \vec{E}_f \rangle| \leq |\langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle|$  for  $h \in \mathbb{R}$ . Moreover, the energy constraint also provides the limiting behavior of the ratio  $\mathcal{R}(h) = \langle \vec{E} \cdot \vec{E}_f \rangle / \langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle = 1/s$ 

$$\lim_{h \to 0} \mathcal{R}(h) = 1, \quad \lim_{h \to 1} \mathcal{R}(h) = 0, \quad \lim_{h \to +\infty} \mathcal{R}(h) = -\infty.$$

Analogous formulas involving the vector field  $\vec{J}_f$  also hold.

THE FORMULA  $s = \langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle / \langle \vec{E} \cdot \vec{E}_f \rangle$  AND THE ANALOGOUS FORMULA INVOLVING THE CURRENT DENSITY PROVIDES A NON-LINEAR RELATIONSHIP BETWEEN  $\sigma^*$  and  $\rho^*$ .

- A-1.3. The Stieltjes–Perron Inversion Theorem TAKE THIS FROM GRAEME'S BOOK  $m_{jk}(h) = h w_{jk}(z)$ , and  $\tilde{m}_{jk}(h) = h \tilde{w}_{jk}(z)$
- **A-1.4.** The Spectral Theorem Under the ACM In this section we discuss the spectral theorem [38, 43] as it pertains to the ACM. In Section A-1.4.1 we review the spectral theorem for the operators  $M_i = \chi_i \Gamma \chi_i$ , i = 1, 2, in the continuum and infinite lattice settings discussed in Sections 2.1 and 2.2.1, respectively. In Section A-1.4.2 we discuss the spectral theorem in the finite lattice setting, where the operators  $M_i$ , i = 1, 2, are real-symmetric random matrices. In each case we obtain the Stieltjes integral representation for  $\sigma^*$  and  $\sigma^*$  given in equations (2.8) and (2.18), respectively, involving a matrix valued spectral measure  $\mu$ .
- **A-1.4.1. Continuum and Infinite Lattice Settings** Due to the inherent symmetries in the ACM between the continuum and infinite lattice settings discussed in Sections 2.1 and 2.2.1, respectively, here we focus on the continuum setting. The associated discussion regarding the infinite lattice setting is a direct parallel to that given here. Let the random operators  $\chi_i$ , i=1,2, and the non-random integro-differential operator  $\Gamma$  be defined as in equations (2.5) and (2.7), respectively. On the Hilbert space  $\mathscr{H}_{\times} \subset \mathscr{H}$ ,  $\Gamma$  and  $\chi_i$ , i=1,2, are projection operators [22]. Therefore  $M_i = \chi_i \Gamma \chi_i$ , i=1,2, are compositions of projection operators on  $\mathscr{H}_{\times}$ , and are consequently positive definite and bounded by 1 in the underlying operator norm [39]. They are self-adjoint with respect to the  $\mathscr{H}$ -inner-product  $\langle \cdot, \cdot \rangle$  [22]. Therefore, on the Hilbert space  $\mathscr{H}_{\times}$  with weight  $\chi_1$  in the inner-product,  $\langle \cdot, \cdot \rangle_1 = \langle \chi_1 \cdot, \cdot \rangle$  for example,  $\Gamma \chi_1$  is a bounded linear self-adjoint operator with spectrum in the interval [0,1] [22]. Hence the resolvent operator  $(sI \Gamma \chi_1)^{-1}$  is also self-adjoint with respect to the same inner-product, and is bounded for  $s \in \mathbb{C} \setminus [0,1]$  [43].

By the spectral theorem [38, 43] for the operator  $\Gamma \chi_1$  on the Hilbert space  $\mathscr{H}_{\times}$  with inner-product  $\langle \cdot, \cdot \rangle_1$ , there exists an increasing family of self-adjoint projection operators  $\{R(\lambda)\}$  - the resolution of the identity - that satisfy R(0) = 0 and R(1) = I such that

$$f(M_1) = \int f(\lambda)R(d\lambda), \quad \langle f(M_1)\vec{e}_j \cdot \vec{e}_k \rangle_1 = \int_0^1 f(\lambda)\mu_{jk}(d\lambda), \tag{A-5}$$

for all bounded continuous functions  $f: \mathbb{C} \mapsto \mathbb{C}$ . Here 0 and I are the null and identity operators on  $\mathbb{R}^d$ , respectively,  $R(d\lambda)$  is the projection valued measure associated with the operator  $R(\lambda)$  [38], and  $\mu_{jk}(d\lambda) = \langle R(d\lambda)\vec{e_j}\cdot\vec{e_k}\rangle_1$ ,  $j,k=1,\ldots,d$ , are the components of the matrix valued spectral measure  $\mu(d\lambda)$  in the  $(\vec{e_j},\vec{e_k})$  state [22, 38, 43]. Setting  $f(\lambda) = (s-\lambda)^{-1}$  in (A-5) yields the the integral formula for  $F_{jk}(s)$  and  $\sigma_{jk}^* = \sigma_2 m_{jk}(h)$  in equation (2.8). An analogous discussion involving the operator  $\chi_2 \Gamma_{\times} \chi_2$  on the Hilbert space  $\mathscr{H}_{\bullet}$  and the function  $E_{jk}(s)$  introduced in Section A-1.2 leads to the integral representation for  $E_{jk}(s)$  displayed in equation (A-30) below.

As the spectrum of the operator  $M_1$  is contained in the interval [0,1], the support  $\Sigma_{jk}$  of the measure  $\mu_{jk}$  satisfies  $\Sigma_{jk} \subseteq [0,1]$  [38]. The mass  $\mu_{jk}^0$  of the measure  $\mu_{jk}$  satisfies  $\mu_{jk}^0 = p_1 \delta_{jk}$  for all  $p_1 \in [0,1]$ , where  $p_1 = \langle \chi_1 \rangle$  is the volume fraction of material component one. To see this, note that setting  $f(M_1) = I$  ( $f(\lambda) = 1$ ) in equation (A-5)

implies that  $\int_0^1 R(d\lambda) = I$ . Moreover, recall that the projection operator  $R(\lambda)$  is self-adjoint on  $\mathscr{H}_{\times}$  for all  $\lambda \in \Sigma_{jk}$  [38, 43]. Consequently, we have

$$\mu_{jk}^{0} = \int_{0}^{1} \mu_{jk}(d\lambda) = \int_{0}^{1} \langle R(d\lambda)\vec{e}_{j} \cdot \vec{e}_{k} \rangle_{1} = \langle 1 \rangle_{1} \, \vec{e}_{j} \cdot \vec{e}_{k} = \langle \chi_{1} \rangle \, \delta_{jk}, \tag{A-6}$$

$$\mu_{kk}(d\lambda) = \langle R(d\lambda)\vec{e}_{k} \cdot \vec{e}_{k} \rangle_{1} = \langle R(d\lambda)\vec{e}_{k} \cdot R(d\lambda)\vec{e}_{k} \rangle_{1} = \|R(d\lambda)\vec{e}_{k}\|_{1}^{2} > 0,$$

where we have used a Fubini theorem [16] and  $\|\cdot\|_1$  denotes the norm induced by the inner-product  $\langle\cdot,\cdot\rangle_1$ . From equation (A-6) we see, generically, that the diagonal components  $\mu_{kk}$  of  $\mu$  are positive measures of mass  $p_1$ , while the off-diagonal components  $\mu_{jk}$ ,  $j \neq k = 1,...,d$ , have zero mass and are consequently signed measures [16, 39].

The higher order moments  $\mu_{jk}^n$ , n=1,2,3,..., in principle, may be found using a perturbation expansion of  $F_{jk}(s)$  about a homogeneous medium  $(\sigma_1 = \sigma_2, s = \infty)$  [22]. In particular  $\mu_{jk}^0 = p_1 \delta_{jk}$ , generically, and  $\mu_{jk}^1 = (p_1 p_2/d) \delta_{jk}$  for a statistically isotropic random medium [22, 6]. In the case of a square bond lattice, which is an example of an infinitely interchangeable random medium,  $\mu_{kk}^2 = p_1 p_2 (1 + (d-2)p_2)/d^2$  for any dimension d and  $\mu_{kk}^3 = p_1 p_2 (p_2^2 - p_2 - 1)/8$  for d=2. In general, the moments  $\mu_{jk}^n$  depend on the (n+1)-point correlation functions of the random medium [22, 6].

**A-1.4.2. Finite Lattice Setting** In this section we derive a discrete version of the spectral theorem given in equation (A-5). This leads to the discrete integral representation for the effective conductivity tensor  $\sigma^*$  displayed in equation (2.18). Toward this goal, we defined in Section 2.2.2 a bijective mapping  $\Theta: \mathbb{Z}_L^d \to \mathbb{N}_L$  from the finite d-dimensional bond lattice  $\mathbb{Z}_L^d$  defined in (2.14) onto the one dimensional set  $\mathbb{N}_L$  defined in equation (2.15). Moreover we showed that, under the mapping  $\Theta$ , the random operator  $M_1 = \chi_1 \Gamma \chi_1$  can be represented as a real-symmetric random matrix of size  $N \times N$ , where  $N = dL^d$  [21, 34]. More specifically,  $\Gamma$  is a non-random real-symmetric projection matrix ( $\Gamma^2 = \Gamma$ ) and  $\chi_1$  is a random diagonal projection matrix with zeros and ones along the main diagonal, and has the block diagonal form displayed in equation (2.17). Since  $M_1(\omega)$  is a composition of projection matrices, it is positive definite,  $M_1(\omega)\vec{\xi}\cdot\vec{\xi}=(\Gamma\chi_1(\omega)\vec{\xi})\cdot(\Gamma\chi_1(\omega)\vec{\xi})\geq 0$  for every  $\omega\in\Omega$  and  $\vec{\xi}\in\mathbb{R}^N$ , and consequently has spectra  $\Sigma^\lambda(\omega)\subseteq[0,1]$  [24].

It is well known [24, 28] that the eigenvectors  $\vec{u}_i(\omega)$ ,  $i=1,\ldots,N$ , of the symmetric matrix  $M_1(\omega)$  form an orthonormal basis for  $\mathbb{R}^N$ , for each  $\omega \in \Omega$ , i.e.,  $\vec{u}_j^T \vec{u}_k = \delta_{jk}$  and for every  $\vec{\xi} \in \mathbb{R}^N$  we have  $\vec{\xi} = \sum_{i=1}^N (\vec{u}_i^T \vec{\xi}) \vec{u}_i = \left(\sum_{i=1}^N \vec{u}_i \vec{u}_i^T\right) \vec{\xi}$ . Consequently,

$$\sum_{i=1}^{N} R_i(\omega) = I, \quad R_i(\omega) = \vec{u}_i(\omega) \vec{u}_i^T(\omega), \quad \forall \ \omega \in \Omega,$$
(A-7)

where I is the identity operator on  $\mathbb{R}^N$  and the matrix  $R_i$  is the orthogonal projector  $(R_iR_j = R_i\delta_{ij})$  onto the eigenspace spanned by  $\vec{u}_i$ , which is associated with the *real* eigenvalue  $\lambda_i(\omega) \in \Sigma^{\lambda}(\omega)$ .

Since  $M_1\vec{u}_i = \lambda_i\vec{u}_i$ , for each i = 1, ..., N, equation (A-7) implies that we also have  $M_1R_i = \lambda_iR_i$  which, in turn, implies that the matrix  $M_1$  has the spectral decomposition  $M_1 = \sum_{i=1}^N \lambda_i R_i$ . By the orthogonality of the projection matrices  $R_i$  and by induction we have  $M_1^n = \sum_{i=1}^N \lambda_i^n R_i$  for all  $n \in \mathbb{N}$ , which implies that  $f(M_1) = \sum_{i=1}^N f(\lambda_i)R_i$  for any polynomial  $f: \mathbb{C} \mapsto \mathbb{C}$ . This formula is a discrete version of the first formula in equation (A-5) for polynomial  $f(\lambda)$ , and leads to a discrete

version of the functional representation of  $f(M_1)$  in (A-5) involving a matrix valued spectral measure  $\mu(d\lambda)$  with components  $\mu_{jk}(d\lambda)$ 

$$\langle f(M_1)\hat{e}_j \cdot \hat{e}_k \rangle = \int_0^1 f(\lambda)\mu_{jk}(d\lambda), \quad \mu_{jk}(d\lambda) = \sum_{i=1}^N \langle \delta_{\lambda_i}(d\lambda)R_i \,\hat{e}_j \cdot \hat{e}_k \rangle. \tag{A-8}$$

Here  $R(d\lambda) = \sum_i \delta_{\lambda_i}(d\lambda) R_i$  is a discrete version of the projection valued measure introduced in equation (A-5),  $\delta_{\lambda_i}(d\lambda)$  is the Dirac measure concentrated at  $\lambda_i$ , the orthonormal vectors  $\hat{e}_i = \Theta(\vec{e}_i)/L^{d/2}$ , for i = 1, ..., d, represent the standard basis vectors on  $\mathbb{N}_L$ , and  $\langle \cdot \rangle$  denotes ensemble average over  $\Omega$ . The spectral end points  $\lambda_{jk}^0$  and  $\lambda_{jk}^1$  of the support  $\Sigma_{jk} \subseteq [\lambda_{jk}^0, \lambda_{jk}^1] \subseteq [0,1]$  of the measure  $\mu_{jk}$  in (A-8) are given by  $\lambda_{jk}^0 = \inf A_{jk}$  and  $\lambda_{jk}^1 = \sup A_{jk}$ , where

$$A_{jk} = \bigcup_{\omega \in \Omega} \{ \lambda_i(\omega) \in \Sigma^{\lambda}(\omega), i = 1, ..., N \mid R_i(\omega) \hat{e}_j \cdot \hat{e}_k \neq 0 \}$$

and  $\Sigma_1^{\lambda}(\omega) \subseteq [0,1]$  is the support of the eigenvalues of the matrix  $M_1(\omega)$  for  $\omega \in \Omega$ .

We now show that equation (A-8) also holds for the function  $f(\lambda) = (s - \lambda)^{-1}$  when  $s \notin [0,1]$ . For each  $\omega \in \Omega$ , let  $U(\omega)$  denote the matrix with columns consisting of the eigenvectors  $\vec{u}_i(\omega)$  of  $M_1(\omega)$  and let  $\Lambda(\omega) = \operatorname{diag}(\lambda_1(\omega), \dots, \lambda_N(\omega))$  denote the diagonal matrix of the corresponding eigenvalues  $\lambda_i(\omega)$ ,  $i = 1, \dots, N$ , so that  $M_1 = U\Lambda U^T$  [24]. By the orthogonality,  $U^TU = UU^T = I$ , of the matrix U we have

$$\langle f(M_1)\hat{e}_j \cdot \hat{e}_k \rangle = \langle (sI - U\Lambda U^T)^{-1}\hat{e}_j \cdot \hat{e}_k \rangle = \langle U(sI - \Lambda)^{-1}U^T\hat{e}_j \cdot \hat{e}_k \rangle$$

$$= \langle (sI - \Lambda)^{-1}U^T\hat{e}_j \cdot U^T\hat{e}_k \rangle = \sum_{i=1}^N \left\langle \frac{R_i\hat{e}_j}{s - \lambda_i} \cdot \hat{e}_k \right\rangle.$$
(A-9)

Equation (A-9) is equivalent to equation (A-8) when the function  $f(\lambda) = (s - \lambda)^{-1}$ ,  $s \in \mathbb{C} \setminus [0,1]$ .

We now discuss the fundamental difference in the mathematical framework between the *infinite* settings formulated in Sections 2.1, 2.2.1, and A-1.4.1, and the finite lattice setting formulated here. In the infinite settings, the (infinite-dimensional) operator  $\Gamma \chi_1$  appears in the integral representation (2.8) for the effective parameter, involving the  $\mathcal{H}$ -inner-product weighted by  $\chi_1(\vec{x},\omega)$ . In this abstract (infinitedimensional) Hilbert space formulation of the effective parameter problem, the resolvent  $(sI - \Gamma \chi_1)^{-1}$  is a linear self-adjoint operator which is bounded for  $s \in \mathbb{C} \setminus [0,1]$  [43]. In contrast, the finite lattice formulation of the effective parameter problem involves a finite dimensional Hilbert space, and the operators  $\Gamma$  and  $\chi_1$  are matrices. In this case, the matrix  $\Gamma \chi_1$  is not symmetric, it typically has complex spectrum, and it may not even have a full set of eigenvectors which span  $\mathbb{R}^N$ . Consequently, the integral formulas in equations (A-8) and (A-9), which were derived for the symmetric matrix  $M_1 = \chi_1 \Gamma \chi_1$ , fail to hold for the matrix  $\Gamma \chi_1$  in general. Due to this fundamental difference in the theory for the finite lattice setting, the mathematical framework must be significantly modified from that for the infinite settings. The ACM for the finite lattice case is summarized by the following theorem.

THEOREM A-1.1. For each  $\omega \in \Omega$ , let the real-symmetric projection matrices  $\Gamma$  and  $\chi_1(\omega)$  of size  $N \times N$  be defined as in Section 2.2.2, and let  $N_1(\omega) = \operatorname{Trace}(\chi_1(\omega))$  be the number of ones along the main diagonal of  $\chi_1(\omega)$ , with  $N_0(\omega) = N - N_1(\omega)$ . Moreover, let the matrix  $M_1(\omega) = \chi_1(\omega)\Gamma\chi_1(\omega)$  and let  $M_1(\omega) = U(\omega)\Lambda(\omega)U(\omega)$  be its spectral decomposition. Here, the columns of the matrix  $U(\omega)$  consist of the orthonormal eigenvectors  $\vec{u}_i(\omega)$ , i = 1, ..., N, of  $M_1(\omega)$  and the diagonal matrix  $\Lambda(\omega) = 1$ 

diag $(\lambda_1(\omega),...,\lambda_N(\omega))$  involves the eigenvalues  $\lambda_i(\omega) \in [0,1]$  of  $M_1(\omega)$ . Then, there exists a permutation matrix  $\Pi(\omega)$  of size  $N \times N$ , an orthogonal matrix  $U_1(\omega)$  of size  $N_1(\omega) \times N_1(\omega)$ , and a diagonal matrix  $\Lambda_1(\omega)$  of size  $N_1(\omega) \times N_1(\omega)$  such that

$$U = \Pi^{T} \begin{bmatrix} I_{0} & 0_{01} \\ 0_{10} & U_{1} \end{bmatrix}, \qquad \Lambda = \begin{bmatrix} O_{00} & O_{01} \\ O_{10} & \Lambda_{1} \end{bmatrix}, \tag{A-10}$$

where  $I_0$  is the identity matrix of size  $N_0(\omega) \times N_0(\omega)$  and  $O_{ab}$  is a matrix of zeros of size  $N_a(\omega) \times N_b(\omega)$ , for a,b=0,1. Furthermore, let  $R_i(\omega)$  be the projection matrices defined in equation (A-7). If the electric field  $\vec{E}(\omega)$  satisfies  $\vec{E}(\omega) = \vec{E}_0 + \vec{E}_f(\omega)$ , with  $\vec{E}_0 = \langle \vec{E}(\omega) \rangle$  and  $\Gamma \vec{E}(\omega) = \vec{E}_f(\omega)$ , then the effective complex conductivity tensor  $\sigma^*$  has components  $\sigma^*_{jk} = \sigma_2 m_{jk}(h)$ ,  $j,k=1,\ldots,d$ , which satisfy

$$m_{jk}(h) = \delta_{jk} - F_{jk}(s), \quad F_{jk}(s) = \int_0^1 \frac{\mu_{jk}(d\lambda)}{s - \lambda}, \quad \mu_{jk}(d\lambda) = \sum_{i=1}^N \langle \delta_{\lambda_i}(d\lambda) \chi_1 R_i \hat{e}_j \cdot \hat{e}_k \rangle. \tag{A-11}$$

Moreover, the mass  $\mu_{ik}^0$  of the measure  $\mu_{jk}$  satisfies

$$\mu_{jk}^{0} = \langle \chi_1 \hat{e}_k \cdot \hat{e}_k \rangle \, \delta_{jk} = d \frac{\langle N_1^k(\omega) \rangle}{N} \, \delta_{jk}, \tag{A-12}$$

where  $N_1^k(\omega) = \operatorname{Trace}(\chi_1^k(\omega))$  is the total number of type-one bonds in the positive  $k^{th}$  direction for  $\omega \in \Omega$  and the matrix  $\chi_1^k(\omega)$  is defined in equation (2.17).

Before we prove Theorem A-1.1, we first discuss some consequences of equation (A-12). In particular, we use this formula to explicitly determine  $\mu_{kk}^0$  for some important examples of a large class of composite micro-geometries. Namely, the class of geometries such that  $N_1^k(\omega)$  is a non-random constant  $N_1^k$  for all  $k=1,\ldots,d$ , i.e.  $N_1^k(\omega)=N_1^k$  for all  $\omega\in\Omega$ . Consequently,  $N_1(\omega)=N_1$  for all  $\omega\in\Omega$  and  $N_1=\sum_k N_1^k$ . Let  $p_1^k=N_1^k/N$  be the number fraction of type-one bonds in the positive  $k^{\text{th}}$  direction, so that  $p_1=\sum_k p_1^k$ . By equation (A-12), for this class of composites we have

$$\mu_{jk}^0 = dp_1^k \delta_{jk}. \tag{A-13}$$

Given a fixed number fraction  $p_1=N_1/N$  of type-one bonds, one can define a class of highly anisotropic composite geometries by fixing  $p_1^k$  close to  $p_1$  for some  $k=1,\ldots,d$ , i.e.  $p_1-p_1^k\ll 1$ . A class of locally isotropic random media is obtained by requiring that every bond emanating from  $\vec{x}\in\mathbb{Z}_L^d$  in the positive direction is of the same type, i.e.  $\chi_1^j(\omega)=\chi_1^k(\omega)$  hence  $N_1^j(\omega)=N_1^k(\omega)$  for all  $j,k=1,\ldots,d$  and  $\omega\in\Omega$ . In this case  $N_1^k(\omega)=N_1/d$ , thus  $p_1^k=p_1/d$  for all  $k=1,\ldots,d$  and  $\omega\in\Omega$ . Consequently, equations (A-12) and (A-13) yield

$$\mu_{jk}^0 = p_1 \delta_{jk}. \tag{A-14}$$

In this case, equations (A-12) and (A-14) provide a direct analogue of equation (A-6) for the continuum and infinite lattice settings (WE MUST RESOLVE WHETHER EQUATION (A-13) HOLDS IN SOME SENSE FOR THE INFINITE LATTICE SETTING. IF SO, WE NEED A SEPARATE DISCUSSION FOR THE INFINITE LATTICE SETTING WHICH LEADS TO THE ANALOGUE OF EQUATION (A-6)). Equation (A-14) also holds when each of the N bonds are chosen (independently) to

be type-one with probability  $p_1 = N_1/N$  and type-two with probability  $1-p_1$ . In this case the  $N_1^k(\omega)$ ,  $k=1,\ldots,d$ , are independent, identically distributed random variables with mean  $\langle N_1^k(\omega) \rangle = p_1$  (IS THIS STATEMENT CORRECT?).

**Proof of Theorem A-1.1.** Taking  $\vec{E} = \vec{E}_0 + \vec{E}_f$  with the condition  $\Gamma \vec{E} = \vec{E}_f$  as a definition greatly simplifies the proof of Theorem A-1.1, by avoiding the formulation and proof of some technical lemmas regarding the commutativity of the matrices  $D_j$ ,  $D_j^T$ , and  $(\Delta^{-1})$  for j = 1, ..., d. This is a natural assumption to make, since in equation (A-4) we showed that the condition  $\Gamma \vec{E} = \vec{E}_f$  is sufficient for the energy constraint  $\langle \vec{J} \cdot \vec{E}_f \rangle = 0$ , which is at the heart of the existence of solutions to equations (2.3) and (2.12) in the (infinite) continuum and lattice settings, respectively. In the finite lattice setting where  $\Gamma$  and  $\chi_1$  are matrices, this condition leads to equation (A-2) exactly as in Section (A-1.2), which is equivalent to the formula  $(sI - \Gamma \chi_1)\vec{E} = s\vec{E}_0$  and the resolvent representation of the electric field in (2.7). As discussed above in this section, the matrix  $\Gamma \chi_1$  is not symmetric and we therefore multiply this formula by the matrix  $\chi_1$ , yielding the following equation involving the real-symmetric random matrix  $M_1 = \chi_1 \Gamma \chi_1$ 

$$(s\chi_1 - \chi_1 \Gamma \chi_1) \vec{E} = s\chi_1 \vec{E}_0. \tag{A-15}$$

Define the sets  $\mathbb{N}_L^1(\omega)$  and  $\mathbb{N}_L^0(\omega)$  by

$$\mathbb{N}_L^1(\omega) = \{ i \in \mathbb{N}_L \mid (\chi_1(\omega))_{ii} = 1 \}, \qquad \mathbb{N}_L^0(\omega) = \mathbb{N}_L \setminus \mathbb{N}_L^1(\omega). \tag{A-16}$$

Also, define elementary permutation matrices [13]  $\Pi_{i,j}, i, j = 1, ..., N$ , such that  $\Pi_{i,j} = \Pi_{i,j}^{-1} = \Pi_{i,j}^{T}$  and  $\Pi_{i,j}\vec{\xi}$  is the vector  $\vec{\xi}$  with the  $i^{\text{th}}$  and  $j^{\text{th}}$  entries interchanged. Since  $\chi_1(\omega)$  is a diagonal matrix with  $N_1(\omega)$  ones and  $N_0(\omega)$  zeros along it's main diagonal, it is clear that there exists a permutation matrix  $\Pi(\omega)$  such that

$$\Pi(\omega)\chi_1(\omega)\Pi^T(\omega) = \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & I_1 \end{bmatrix}, \quad \Pi(\omega) = \prod_{i,j \in \mathbb{N}_L} \Pi_{i,j}(\omega), \tag{A-17}$$

for each  $\omega \in \Omega$ , where  $i \in \mathbb{N}_L^1(\omega)$ ,  $j \in \mathbb{N}_L^0(\omega)$ , and  $I_1$  is the identity matrix of size  $N_1(\omega) \times N_1(\omega)$ . Therefore, as  $\Pi^T = \Pi^{-1}$  we have

$$\begin{split} \chi_{1}\Gamma\chi_{1} &= \Pi^{T} \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & I_{1} \end{bmatrix} \Gamma_{\Pi} \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & I_{1} \end{bmatrix} \Pi = \Pi^{T} \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & \Gamma_{1} \end{bmatrix} \Pi = \Pi^{T} \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & \Gamma_{1} \end{bmatrix} \Pi \\ &= \Pi^{T} \begin{bmatrix} I_{0} & 0_{01} \\ 0_{10} & U_{1} \end{bmatrix} \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & \Lambda_{1} \end{bmatrix} \begin{bmatrix} I_{0} & 0_{01} \\ 0_{10} & U_{1}^{T} \end{bmatrix} \Pi, \end{split} \tag{A-18}$$

where  $\Gamma_{\Pi} = \Pi \Gamma \Pi^{T}$ ,  $\Gamma_{1}$  is the (real-symmetric) lower right principal sub-matrix of  $\Gamma_{\Pi}$  of size  $N_{1}(\omega) \times N_{1}(\omega)$ , and  $\Gamma_{1} = U_{1}\Lambda_{1}U_{1}^{T}$  is its eigenvalue decomposition. As  $\Gamma_{1}$  is a real-symmetric matrix,  $U_{1}$  is an orthogonal matrix. Moreover, since  $\Gamma_{\Pi} = \Pi \Gamma \Pi^{T}$  is a similarity transformation of a projection matrix and  $\Pi \chi_{1}\Pi^{T}$  is a projection matrix,  $\Lambda_{1}$  is a diagonal matrix with entries  $\lambda_{j}^{\Pi_{1}} \in [0,1], j=1,\ldots,N_{1}(\omega)$ , along the main diagonal [13]. Equation (A-18) and  $\chi_{1}\Gamma\chi_{1} = U\Lambda U^{T}$  imply equation (A-10) in the statement of the theorem, where U is an orthogonal matrix and  $\Lambda$  is a diagonal matrix with entries  $\lambda_{j} \in [0,1], j=1,\ldots,N$ , along the main diagonal.

From  $\Pi^{\mathbf{T}} = \Pi^{-1}$  and equations (A-15), (A-17), and (A-18) we have

$$\begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & (sI_1 - U_1\Lambda_1U_1^T) \end{bmatrix} \Pi \vec{E} = \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & sI_1 \end{bmatrix} \Pi \vec{E}_0. \tag{A-19}$$

Define the coordinate system such that  $\vec{E_0} = E_0 \hat{e}_j$ , for some j = 1, ..., d, and write

$$\Pi \vec{E} = \begin{bmatrix} \vec{E}^{\Pi_0} \\ \vec{E}^{\Pi_1} \end{bmatrix}, \quad \Pi \vec{E}_0 = \begin{bmatrix} \vec{E}_0^{\Pi_0} \\ \vec{E}_0^{\Pi_1} \end{bmatrix} = E_0 \begin{bmatrix} \hat{e}_i^{\Pi_0} \\ \hat{e}_j^{\Pi_1} \end{bmatrix}, \quad \Pi \vec{u}_i = \begin{bmatrix} \vec{u}_i^{\Pi_0} \\ \vec{u}_i^{\Pi_1} \end{bmatrix}, \tag{A-20}$$

where  $\vec{E}^{\Pi_0} \in \mathbb{R}^{N_0}$ ,  $\vec{E}^{\Pi_1} \in \mathbb{R}^{N_1}$ , and similarly for the vectors  $\Pi \vec{E}_0$  and  $\Pi \vec{u}_i$ . From equation (A-19) we have  $(sI_1 - U_1\Lambda_1U_1^T)\vec{E}^{\Pi_1} = s\vec{E}_0^{\Pi_1}$ , which yields the following resolvent representation for  $\vec{E}^{\Pi_1}$  that is a direct analogue of equation (2.7)

$$\vec{E}^{\Pi_1} = s(sI_1 - U_1\Lambda_1 U_1^T)^{-1} \vec{E}_0^{\Pi_1}, \quad s \in \mathbb{C} \setminus [0, 1]. \tag{A-21}$$

Equation (A-21) leads to a Stieltjes integral representation for  $\sigma^*$  with components  $\sigma_{jk}^* = \sigma^* \hat{e}_j \cdot \hat{e}_k$  as follows. Recall the definition of the effective complex conductivity tensor  $\sigma^*$ :

$$\boldsymbol{\sigma}^* \vec{E}_0 = \langle \vec{J} \rangle = \sigma_2 \langle (1 - \chi_1/s) \vec{E} \rangle = \sigma_2 (\vec{E}_0 - \langle \chi_1 \vec{E} \rangle / s). \tag{A-22}$$

Since the symmetric matrix  $\chi_1$  satisfies  $\chi_1^2 = \chi_1$  and  $\Pi^T = \Pi^{-1}$ , equations (A-17) and (A-20) yield the following projection identity

$$\chi_1 \vec{E} \cdot \hat{e}_k = \vec{E}^{\Pi_1} \cdot \hat{e}_k^{\Pi_1}. \tag{A-23}$$

Therefore, equations (A-9), (A-16), and (A-20)–(A-23) imply that

$$\delta_{ij} - \sigma_{jk}^* / \sigma_2 = \langle \chi_1 \vec{E} \cdot \hat{e}_k \rangle / (sE_0) = \langle \vec{E}^{\Pi_1} \cdot \hat{e}_k^{\Pi_1} \rangle / (sE_0)$$

$$= \langle (sI_1 - U_1 \Lambda_1 U_1^T)^{-1} \hat{e}_j^{\Pi_1} \cdot \hat{e}_k^{\Pi_1} \rangle = \sum_{i \in \mathbb{N}_1^1} \left\langle \frac{R_i^{\Pi_1} \hat{e}_j^{\Pi_1}}{s - \lambda_i^{\Pi_1}} \cdot \hat{e}_k^{\Pi_1} \right\rangle, \quad (A-24)$$

where  $R_i^{\Pi_1}$ ,  $i \in N_L^1(\omega)$ , is the projection matrix of size  $N_1(\omega) \times N_1(\omega)$  associated with the columns  $\vec{u}_i^{\Pi_1}$  of the orthogonal matrix  $U_1$ .

We now show that equation (A-24) is equivalent to equation (A-11). Since  $\Pi^T = \Pi^{-1}$ , equations (A-10) and (A-17) imply that

$$\chi_1 U = \Pi^T \begin{bmatrix} 0_{00} & 0_{01} \\ 0_{10} & U_1 \end{bmatrix}. \tag{A-25}$$

Recalling the definitions of  $\vec{u}_i$  and  $R_i$  from the statement of the theorem, equation (A-25) implies that

$$\chi_1 \vec{u}_i = \begin{cases} \vec{u}_i, & \text{for } i \in \mathbb{N}_L^1 \\ 0, & \text{for } i \in \mathbb{N}_L^0 \end{cases} \Rightarrow \chi_1 R_i = \begin{cases} R_i, & \text{for } i \in \mathbb{N}_L^1 \\ 0, & \text{for } i \in \mathbb{N}_L^0 \end{cases}.$$
 (A-26)

Similar to equation (A-23) we have the projection identity  $\vec{u}_i^{\Pi_1} \cdot \hat{e}_j^{\Pi_1} = \chi_1 \vec{u}_i \cdot \hat{e}_j$ . This and equation (A-26) yield another important projection identity

$$R_i^{\Pi_1} \hat{e}_j^{\Pi_1} \cdot \hat{e}_k^{\Pi_1} = (\vec{u}_i^{\Pi_1} \cdot \hat{e}_j^{\Pi_1}) (\vec{u}_i^{\Pi_1} \cdot \hat{e}_k^{\Pi_1}) = (\chi_1 \vec{u}_i \cdot \hat{e}_j) (\chi_1 \vec{u}_i \cdot \hat{e}_k) = \chi_1 R_i \hat{e}_j \cdot \hat{e}_k.$$

Therefore, equation (A-26) implies that

$$\sum_{i \in \mathbb{N}_{t}^{1}} \left\langle \frac{R_{i}^{\Pi_{1}} \hat{e}_{j}^{\Pi_{1}}}{s - \lambda_{i}^{\Pi_{1}}} \cdot \hat{e}_{k}^{\Pi_{1}} \right\rangle = \sum_{i \in \mathbb{N}_{t}^{1}} \left\langle \frac{\chi_{1} R_{i} \hat{e}_{j}}{s - \lambda_{i}^{\Pi_{1}}} \cdot \hat{e}_{k} \right\rangle = \sum_{i=1}^{N} \left\langle \frac{\chi_{1} R_{i} \hat{e}_{j}}{s - \lambda_{i}} \cdot \hat{e}_{k} \right\rangle,$$

which proves our claim that equation (A-24) is equivalent to equation (A-11). Note that the equivalence of equations (A-24) and (A-11) implies that the spectral measure  $\mu_{jk}$  in (A-11) satisfies

$$\mu_{jk}(d\lambda) = \sum_{i=1}^{N} \langle \delta_{\lambda_i}(d\lambda) \chi_1 R_i \hat{e}_j \cdot \hat{e}_k \rangle \equiv \sum_{i \in \mathbb{N}_I} \langle \delta_{\lambda_i^{\Pi_1}}(d\lambda) R_i^{\Pi_1} \hat{e}_j^{\Pi_1} \cdot \hat{e}_k^{\Pi_1} \rangle. \tag{A-27}$$

We now discuss the analogue of equation (A-6) for the finite lattice setting and prove equation (A-12) in the statement of the theorem. Recall that the projection matrices  $R_i^{\Pi_1}$ ,  $i \in \mathbb{N}_L$ , are symmetric. Therefore, by equations (A-7) and (A-27) we have

$$\mu_{jk}^{0} = \int_{0}^{1} \mu_{jk}(d\lambda) = \int_{0}^{1} \sum_{i=1}^{N} \langle \delta_{\lambda_{i}}(d\lambda) \chi_{1} R_{i} \hat{e}_{j} \cdot \hat{e}_{k} \rangle = \langle \chi_{1} \hat{e}_{j} \cdot \hat{e}_{k} \rangle = \langle \chi_{1} \hat{e}_{k} \cdot \hat{e}_{k} \rangle \delta_{jk},$$

$$\mu_{kk}(d\lambda) = \sum_{i \in \mathbb{N}_{L}} \langle \delta_{\lambda_{i}^{\Pi_{1}}}(d\lambda) R_{i}^{\Pi_{1}} \hat{e}_{k}^{\Pi_{1}} \cdot \hat{e}_{k}^{\Pi_{1}} \rangle = \sum_{i \in \mathbb{N}_{L}} \langle \delta_{\lambda_{i}^{\Pi_{1}}}(d\lambda) | R_{i}^{\Pi_{1}} \hat{e}_{k}^{\Pi_{1}} |^{2} \rangle \geq 0, \quad (A-28)$$

where  $|\cdot|$  denotes the  $l^2$  norm on  $\mathbb{R}^{N_1(\omega)}$ . Therefore, as in the continuum and infinite lattice settings, the diagonal components  $\mu_{kk}$  of the matrix valued measure  $\mu$  are positive measures of mass  $\langle \chi_1 \hat{e}_k \cdot \hat{e}_k \rangle$  while the off-diagonal components  $\mu_{jk}$ , for  $j \neq k$ , have zero mass and are consequently signed measures. Using equation (2.17) we may write  $\mu^0_{jk}$  in (A-28) in a more suggestive form. Recall that  $\hat{e}_1 = (\vec{1}, \vec{0}, ..., \vec{0})/L^{d/2}$ , where  $\vec{l}$  and  $\vec{0}$  are vectors of ones and zeros of length  $L^d$ , respectively, and similarly for the  $\vec{e}_j$  for j = 2, ..., d (see Section 2.2.2 for details). Since  $\chi_1$  is a symmetric projection matrix, equations (2.17) and (A-28) imply that

$$\mu_{jk}^{0} = \langle \chi_{1} \hat{e}_{k} \cdot \hat{e}_{k} \rangle \, \delta_{jk} = \langle \chi_{1} \hat{e}_{k} \cdot \chi_{1} \hat{e}_{k} \rangle \, \delta_{jk} = \frac{1}{L^{d}} \langle \chi_{1}^{k} \vec{1} \cdot \chi_{1}^{k} \vec{1} \rangle \, \delta_{jk} = \frac{1}{L^{d}} \langle \operatorname{Trace}(\chi_{1}^{k}) \rangle \, \delta_{jk} = d \frac{\langle N_{1}^{k}(\omega) \rangle}{N} \, \delta_{jk},$$
(A-29)

where  $N_1^k(\omega) = \operatorname{Trace}(\chi_1^k(\omega))$  is the total number of type-one bonds in the positive  $k^{\text{th}}$  direction for  $\omega \in \Omega$  and  $N = dL^d$ . This proves equation (A-12) and concludes our proof of Theorem A-1.1  $\square$ .

**A-1.5.** Bounding Procedure A key feature of the integral representation for  $F_{jk}(s)$ , j,k=1,...,d, displayed in equations (2.8) and (2.18), is that parameter information in s and  $E_0$  is separated from the geometry of the composite, which is encoded in the spectral measure  $\mu_{jk}$  via its moments  $\mu_{jk}^n$ , n=0,1,2,... [6, 22]. Another important feature of the representation for  $F_{jk}(s)$  is that it is a linear functional of the measure  $\mu_{jk}$ . Moreover, the diagonal components  $\mu_{kk}$  are positive measures. These important properties are also shared by the function  $E_{jk}(s)$  which was introduced in Section A-1.2. It also has a Stieltjes integral representation which involves a spectral measure  $\eta_{jk}$  associated with the self-adjoint random operator  $\chi_2\Gamma_{\times}\chi_2$  on the Hilbert space  $\mathscr{H}_{\bullet}$  (see Sections A-1.2 and A-1.4 for details)

$$E_{jk}(s) = \langle \chi_2(s - \Gamma_{\times} \chi_2)^{-1} \vec{e_j} \cdot \vec{e_k} \rangle = \int_0^1 \frac{\eta_{jk}(d\lambda)}{s - \lambda}, \quad s \in \mathbb{C} \setminus [0, 1]. \tag{A-30}$$

These important properties of the functions  $F_{jk}(s)$  and  $E_{jk}(s)$  may be exploited to obtain rigorous bounds for the diagonal components of the effective parameters.

In this section we review a bounding procedure which was introduced in [22]. The bounds incorporate the moments  $\mu_{kk}^n$  and  $\eta_{kk}^n$  of the measures  $\mu_{kk}$  and  $\eta_{kk}$ . In the infinite continuum and lattice settings, by equation (A-6) and the symmetries between  $F_{kk}(s)$  and  $E_{kk}(s)$ , the masses of these measures are generically given by  $\mu_{kk}^0 = p_1$  and  $\eta_{kk}^0 = p_2$ . However, in the finite lattice setting, these symmetries and equation (A-29) yield  $\mu_{kk}^0 = d\langle N_1^k(\omega) \rangle/N$  and  $\eta_{kk}^0 = d\langle N_2^k(\omega) \rangle/N$ . Here  $N_1^k(\omega) = \operatorname{Trace}(\chi_1^k(\omega))$  is the total number of type-one bonds in the positive  $k^{\text{th}}$  direction for  $\omega \in \Omega$ , and similarly for  $N_2^k(\omega)$ . This is a fundamental difference between the infinite and finite lattice settings. We discuss the bounding procedure for the infinite continuum and lattice settings in Section A-1.5.1 and that for the finite lattice setting in Section A-1.5.2.

**A-1.5.1.** Continuum and infinite lattice settings In this section, we will discuss the bounding procedure in terms of the diagonal components  $\sigma_{kk}^*$ ,  $k=1,\ldots,d$ , of the effective complex conductivity tensor  $\sigma^*$ . For simplicity, we will focus on one such component and set  $\sigma^* = \sigma_{kk}^*$ ,  $F(s) = F_{kk}(s)$ ,  $m(h) = m_{kk}(h)$ ,  $\mu = \mu_{kk}$ ,  $E(s) = E_{kk}(s)$ ,  $\tilde{m}(h) = \tilde{m}_{kk}(h)$ , and  $\eta = \eta_{kk}$ . Here  $\sigma^* = \sigma_2 m(h) = \sigma_1/\tilde{m}(h)$ , F(s) = 1 - m(h), and  $E(s) = 1 - \tilde{m}(h)$ . We will also exploit the symmetries between equations (2.8) and (A-30), and initially focus on the function F(s) and the measure  $\mu$ , introducing the function E(s) and the measure  $\eta$  when appropriate.

Bounds on  $\sigma^*$  are obtained as follows. By equation (A-6) the support of the measure  $\mu$  is contained in the interval [0,1] and its mass is given by  $\mu^0 = p_1$ , where  $0 \le p_1 \le 1$ . Consider the set  $\mathscr{M}$  of positive Borel measures on [0,1] with mass  $\le 1$ . By equation (2.8), for fixed  $s \in \mathbb{C} \setminus [0,1]$ , F(s) is a linear functional of the measure  $\mu$ ,  $F: \mathscr{M} \mapsto \mathbb{C}$ , and we write  $F(s) = F(s,\mu)$  and  $m(h) = m(h,\mu)$ . Suppose that we know the moments  $\mu^n$  of the measure  $\mu$  for n = 1, ..., J and let

$$\mathcal{M}_{J}^{\mu} = \left\{ \nu \in \mathcal{M} \mid \int_{0}^{1} \lambda^{n} \nu(d\lambda) = \mu^{n}, \ n = 0, \dots, J \right\}. \tag{A-31}$$

The set  $A_J^{\mu} \subset \mathbb{C}$  which represents the possible values of the function  $m(h,\mu) = 1 - F(s,\mu)$  that is compatible with the known information about the random medium is given by

$$A_I^{\mu} = \{ m(h, \mu) \in \mathbb{C} \mid h \notin (-\infty, 0], \ \mu \in \mathcal{M}_I^{\mu} \}. \tag{A-32}$$

The set of measures  $\mathscr{M}_J^{\mu}$  is a compact, convex subset of  $\mathscr{M}$  with the topology of weak convergence [22]. Since the mapping  $F(s,\mu)$  in (2.8) is linear in  $\mu$  it follows that  $A_J^{\mu}$  is a compact convex subset of the complex plane  $\mathbb{C}$ . The extreme points of  $\mathscr{M}_0^{\mu}$  are the one point measures  $\alpha \delta_a$ ,  $0 \le \alpha, a \le 1$  [14], while the extreme points of  $\mathscr{M}_J^{\mu}$  for J > 0 are weak limits of convex combinations of measures of the form [22]

$$\mu_J(d\lambda) = \sum_{i=1}^{J+1} \alpha_i \delta_{\lambda_i}(d\lambda), \quad \alpha_i \ge 0, \quad 0 \le \lambda_1 < \dots < \lambda_{J+1} < 1, \quad \sum_{i=1}^{J+1} \alpha_i \lambda_i^n = \mu^n, \quad (A-33)$$

for n=0,1,...,J. It is important to note that, except for the case of multi-rank laminates [33], (ELENA, IS THIS WORDING AND REFERENCE CORRECT?) not every measure  $\mu \in \mathscr{M}_J^{\mu}$  gives rise to a function  $m(h,\mu)$  that is the effective (relative) conductivity a random medium [22]. Therefore, in general, the set  $A_J^{\mu}$  will contain the exact range of values of the effective conductivity [22]. This is sufficient for the bounding procedure discussed in this section.

By the symmetries between equations (2.8) and (A-30), and equation (A-6), the support of the measure  $\eta$  is contained in the interval [0,1] and its mass is given

by  $\eta^0 = p_2 = 1 - p_1$ , where  $0 \le p_2 \le 1$ . We can therefore define compact, convex sets  $\mathcal{M}_J^{\eta} \subset \mathcal{M}$  and  $A_J^{\eta} \subset \mathbb{C}$  which are analogous to those defined in equations (A-31) and (A-32), respectively, involving the functions  $\tilde{m}(h,\eta) = 1 - E(s,\eta) = \sigma_1/\sigma^*$ . Moreover, the extreme points of  $\mathcal{M}_0^{\eta}$  are the one point measures  $\alpha \delta_a$ ,  $0 \le \alpha, a \le 1$  while the extreme points of  $\mathcal{M}_J^{\eta}$  are weak limits of convex combinations of measures of the form given in equation (A-33).

Consequently, in order to determine the extreme points of the sets  $A_J^{\mu}$  and  $A_J^{\eta}$  it suffices to determine the range of values in  $\mathbb C$  of the functions  $m(h,\mu_J) = 1 - F(s,\mu_J)$  and  $\tilde{m}(h,\eta_J) = 1 - E(s,\eta_J)$ , respectively, where

$$F(s,\mu_J) = \sum_{i=1}^{J+1} \frac{\alpha_i}{s - \lambda_i}, \qquad E(s,\eta_J) = \sum_{i=1}^{J+1} \frac{\alpha_i}{s - \lambda_i},$$
 (A-34)

as the  $\alpha_i$  and  $\lambda_i$  vary under the constraints given in equation (A-33). While  $F(s,\mu_J)$  and  $E(s,\eta_J)$  in (A-34) may not run over all points in  $A_J^\mu$  and  $A_J^\eta$  as the  $\alpha_i$  and  $\lambda_i$  vary, they run over the extreme points of these sets, which is sufficient due to their convexity. It is important to note that, as the effective complex conductivity  $\sigma^*$  is given by  $\sigma^* = \sigma_2 m(h,\mu) = \sigma_1/\tilde{m}(h,\eta)$ , the regions  $A_J^\mu$  and  $A_J^\eta$  have to be mapped to the common  $\sigma^*$ -plane to provide bounds for  $\sigma^*$ .

In this section we discuss two different bounds for  $\sigma^*$ . The first bound assumes that only the masses  $\mu^0 = p_1$  and  $\eta^0 = p_2$  of the measures  $\mu$  and  $\eta$  are known. While the second bound also assumes that the random medium is statistically isotropic, so that the first moments of these measures are also known, and are given by [17]

$$\mu^{1} = \frac{p_{1}p_{2}}{d}, \qquad \eta^{1} = \frac{p_{1}p_{2}(d-1)}{d}.$$
 (A-35)

Consider the first case, where J=0 in (A-34) and the volume fraction  $p_1=1-p_2$  is fixed with  $\mu^0=p_1$  and  $\eta^0=p_2=1-p_1$ , so that  $F(s,\mu_J)=p_1/(s-\lambda)$  and  $E(s,\eta_J)=p_2/(s-\tilde{\lambda})$ . By the above discussion, the values of  $F(s,\mu)$  and  $E(s,\eta)$  lie inside the circles  $C_0(\lambda)$  and  $\tilde{C}_0(\tilde{\lambda})$ , respectively, given by

$$C_0(\lambda) = \frac{\mu^0}{s - \lambda}, \quad -\infty \le \lambda \le \infty, \qquad \tilde{C}_0(\tilde{\lambda}) = \frac{\eta^0}{s - \tilde{\lambda}}, \quad -\infty \le \tilde{\lambda} \le \infty.$$
 (A-36)

In the  $\sigma^*$ -plane, the intersection of these two regions is bounded by two circular arcs corresponding to  $0 \le \lambda \le p_2$  and  $0 \le \tilde{\lambda} \le p_1$  in (A-36), and the values of  $\sigma^*$  lie inside this region [17]. These bounds are optimal [31, 4], and are obtained by a composite of uniformly aligned spheroids of material 1 in all sizes coated with confocal shells of material 2, and vice versa. The arcs are traced out as the aspect ratio varies. When the value of the component permittivities  $\sigma_1$  and  $\sigma_2$  are real and positive, the bounding region collapses to the interval  $1/(p_1/\sigma_1+p_2/\sigma_2) \le \sigma^* \le p_1\sigma_1+p_2\sigma_2$ , which are the Wiener bounds. The lower and upper bounds are obtained by parallel slabs of the two materials aligned perpendicular and parallel to the field  $\vec{E}_0$ , respectively [40].

Now consider the second case, where J=1 in (A-34), the volume fraction  $p_1=1-p_2$  is fixed, and the random medium is statistically isotropic so that the first moments  $\mu^1$  and  $\eta^1$  of the measures  $\mu$  and  $\eta$  are given, respectively, by that in equation (A-35). A convenient way of including this information is to use the transformations [4]

$$F_1(s) = \frac{1}{p_1} - \frac{1}{sF(s)}, \qquad E_1(s) = \frac{1}{p_2} - \frac{1}{sE(s)}.$$
 (A-37)

Due to the symmetries between  $F_1(s)$  and  $E_1(s)$  in (A-37) we will first focus on the function  $F_1(s)$  and introduce the function  $E_1(s)$  when appropriate. The function  $F_1(s)$  is an upper half plane function analytic off [0,1] and therefore has an integral representation [4, 17] analogous to that in equations (2.8) and (A-30), involving a measure  $\mu_1$ , say, which is supported in the interval [0,1]. Since only the mass  $\mu^0 = p_1$  and the first moment  $\mu^1 = p_1 p_2/d$  of the measure  $\mu$  are known, the transformation (A-37) determines only the mass  $\mu^0_1 = p_2/(p_1 d)$  of the measure  $\mu_1$  [4, 17]. This reveals the utility of the transformation  $F_1(s)$  in (A-37), it reduces the second case (J=1) for F(s) to the first case (J=0) for  $F_1(s)$ .

By our previous analysis, the values of  $F_1(s)$  lie inside a circle  $p_2/(p_1d(s-\lambda))$ ,  $-\infty \le \lambda \le \infty$ . Similarly, the values of  $E_1(s)$  lie inside a circle  $p_1(d-1)/(p_2d(s-\tilde{\lambda}))$ ,  $-\infty \le \tilde{\lambda} \le \infty$ . Since F and E are fractional linear in  $F_1$  and  $E_1$ , respectively, these circles are transformed to the circles  $C_1(\lambda)$  in the F-plane and  $\tilde{C}_1(\tilde{\lambda})$  in the E-plane given by [17]

$$C_1(\lambda) = \frac{p_1(s-\lambda)}{s(s-\lambda-p_2/d)}, \quad \tilde{C}_1(\tilde{\lambda}) = \frac{p_2(s-\tilde{\lambda})}{s(s-\tilde{\lambda}-p_1(d-1)/d)}, \quad -\infty \le \lambda, \tilde{\lambda} \le \infty.$$
(A-38)

In the  $\sigma^*$ -plane the intersection of these two circular regions is bounded by two circular arcs [17] corresponding to  $0 \le \lambda \le (d-1)/d$  and  $0 \le \tilde{\lambda} \le 1/d$  in (A-38).

The vertices of the region,  $C_1(0) = p_1/(s - p_2/d)$  and  $\tilde{C}(0) = p_2/(s - p_1(d-1)/d)$ , are attained by the Hashin–Shtrikman geometries (spheres of all sizes of material 1 in the volume fraction  $p_1$  coated with spherical shells of material 2 in the volume fraction  $p_2$  filling all of  $\mathbb{R}^d$ , and vice versa), and lie on the arcs of the first order bounds [17]. While there are at least five points on the arc  $C_1(\lambda)$  in (A-38) that are attainable by composite microstructures [31], the arc  $\tilde{C}_1(\tilde{\lambda})$  in (A-38) violates [17] the interchange inequality  $m(h)m(1/h) \geq 1$  [29, 41], which becomes an equality in two dimensions [33]. Consequently the isotropic bounds in (A-38) are not optimal, but have been improved [30, 4] by incorporating the interchange inequality. When  $\sigma_1$  and  $\sigma_2$  are real and positive with  $\sigma_1 \leq \sigma_2$ , the region collapses to the interval

$$\sigma_1 + p_2 \left/ \left( \frac{1}{\sigma_2 - \sigma_1} + \frac{p_1}{d\sigma_1} \right) \le \sigma^* \le \sigma_1 + p_1 \left/ \left( \frac{1}{\sigma_1 - \sigma_2} + \frac{p_2}{d\sigma_2} \right) \right.\right)$$

which are the Hashin-Shtrikman bounds.

The higher moments  $\mu^n$ ,  $n \ge 2$  depend on (n+1)-point correlation functions [22] and cannot be calculated in general, although the interchange inequality forces relations among them [32]. If the moments  $\mu^0, \ldots, \mu^J$  are known then the transformation  $F_1$  in (A-37) can be iterated to produce an upper half plane function  $F_J$  with a integral representation, involving a positive measure  $\mu_J$  which is supported on the interval [0,1]. As in the case where J=1, the first J moments of the measure  $\mu$  determine only the mass  $\mu^0_J$  of the measure  $\mu_J$  [17], and the function  $F_J(s)$  can easily be extremized by the above procedure, and similarly for a function  $E_J(s)$  associated with the moments  $\eta^0, \ldots, \eta^J$ . The resulting bounds form a nested sequence of lens-shaped regions [17].

**A-1.5.2. Finite lattice setting** In this section we discuss the bounding procedure of the ACM for the finite lattice setting. For explicitness, we again discuss the procedure in terms of the components  $\sigma_{kk}^*$ ,  $k=1,\ldots,d$ , of the effective complex conductivity tensor  $\sigma^*$ . Recall that  $N_1^k(\omega) = \operatorname{Trace}(\chi_1^k(\omega))$  denotes the total number

of type-one bonds in the positive  $k^{\text{th}}$  direction for  $\omega \in \Omega$ , and similarly for  $N_2^k(\omega)$ . A key difference in the ACM between the continuum and finite lattice settings is that the masses  $\mu_{kk}^0$  and  $\eta_{kk}^0$  of the measures  $\mu_{kk}$  and  $\eta_{kk}$  are given by  $\mu_{kk}^0 = p_1$  and  $\eta_{kk}^0 = p_2$  in the continuum case, while  $\mu_{kk}^0 = d\langle N_1^k(\omega)\rangle/N$  and  $\eta_{kk}^0 = d\langle N_2^k(\omega)\rangle/N$  in the finite lattice case (WE NEED TO RESOLVE THIS FOR THE INFINITE LATTICE SETTING). The measure masses of both cases are the same for locally isotropic media, where  $N_1^k(\omega) = N_1/d$  for all  $k = 1, \ldots, d$  and  $\omega \in \Omega$ , and for statistically isotropic media, where  $\langle N_1^k(\omega) \rangle = N_1/d$ , with  $p_1 = N_1/N$  denoting the number fraction of type-one bonds (see the paragraph following the statement of Theorem A-1.1 for more details).

While the bounding procedure is the same for both cases, this fundamental difference between the continuum and finite lattice settings effects the bounds given in Section (A-1.5.1). The bounds given in equation (A-36) depend only on the masses  $\mu_{kk}^0$  and  $\eta_{kk}^0$  of the measures  $\mu_{kk}$  and  $\eta_{kk}$ . They are related as follows. Note that  $\chi_1^k(\omega) + \chi_2^k(\omega) = I_{L^d}$ , where  $I_{L^d}$  is the identity matrix of size  $L^d \times L^d$ . By the linearity of the trace operation, we therefore have  $\operatorname{Trace}(\chi_1^k(\omega)) + \operatorname{Trace}(\chi_2^k(\omega)) = \operatorname{Trace}(I_{L^d})$  or equivalently  $N_1^k(\omega) + N_2^k(\omega) = L^d = N/d$ . Averaging this formula over  $\Omega$  and rearranging yields

$$\mu_{kk}^0 + \eta_{kk}^0 = 1, \quad k = 1, \dots, d.$$

By the discussion in Section A-1.5.1, the values of  $F_{kk}(s)$  and  $E_{kk}(s)$  lie inside the circles  $C_0^k(\lambda)$  and  $\tilde{C}_0^k(\tilde{\lambda})$ , respectively, given by equation (A-36)

$$C_0^k(\lambda) = \frac{\mu_{kk}^0}{s - \lambda}, \quad -\infty \le \lambda \le \infty, \qquad \tilde{C}_0^k(\tilde{\lambda}) = \frac{\eta_{kk}^0}{s - \tilde{\lambda}}, \quad -\infty \le \tilde{\lambda} \le \infty. \tag{A-39}$$

In the  $\sigma_{kk}^*$ -plane, the intersection of these two regions is bounded by two circular arcs corresponding to  $0 \le \lambda \le \eta_{kk}^0$  and  $0 \le \tilde{\lambda} \le \mu_{kk}^0$  in (A-39), and the values of  $\sigma_{kk}^*$  lie inside this region [17]. The isotropic bounds given in equation (A-38) do not hold for anisotropic media, where  $\langle N_1^j(\omega) \rangle \ne \langle N_1^k(\omega) \rangle$  for some  $j \ne k$ . However, in Section 3 we show that the isotropic bounds in (A-38) capture the data for locally isotropic and statistically isotropic random media in the finite lattice setting.

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