SPECTRAL THEORY OF ADVECTIVE DIFFUSION BY DYNAMIC AND STEADY PERIODIC FLOWS

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Abstract. The analytic continuation method for representing transport in composites provides Stieltjes integral representations for the effective coefficients of two-phase random media. Here we adapt this method to characterize the effective thermal transport properties of advective diffusion, by steady and time-dependent, periodic flows. Our novel approach yields a Stieltjes integral representation for the effective diffusivity, which holds for dynamic and steady, incompressible flows, involving the spectral measure of a self-adjoint operator. In the case of steady fluid velocity fields, the spectral measure is associated with a Hermitian Hilbert-Schmidt integral operator, and in the case of dynamic flows, it is associated with an unbounded integro-differential operator. We utalize the integral representation to obtain asymptotic behavior of the effective diffusivity, as the molecular diffusivity tends to zero, for model, steady and dynamic flows. Our analytical results are supported by numerical computations of the spectral measures and effective diffusivities.

1. Introduction

The long time, large scale behavior of a diffusing particle or tracer being advected by an incompressible velocity field is equivalent to an enhanced diffusive process [40] with an effective diffusivity tensor $\boldsymbol{\kappa}^*$. Determining the effective transport properties of advection enhanced diffusion is a challenging problem with theoretical and practical importance in many fields of science and engineering, ranging from turbulent combustion to mass, heat, and salt transport in geophysical flows [30]. A broad range of mathematical techniques have been developed that reduce the analysis of complex fluid flows, with rapidly varying structures in space and time, to solving averaged or *homogenized* equations that do not have rapidly varying data, and involve an effective parameter.

Homogenization of the advection-diffusion equation for thermal transport by time-independent, random fluid velocity fields was treated in [25]. This reduced the analysis of turbulent diffusion to solving an anisotripic diffusion equation involving a homogenized temperature and an effective diffusivity tensor $\boldsymbol{\kappa}^*$. An important consequence of this analysis is that $\boldsymbol{\kappa}^*$ is given in terms of a *curl-free* stationary stochastic process which satisfies a steady state diffusion equation involving a skew-symmetric matrix $\mathbf{H}$ [3, 2]. By adapting the analytic continuation method (ACM) of homogenization theory for composites [19], it was shown that the result in [25] leads to a Stieltjes integral representation of $\boldsymbol{\kappa}^*$, involving a spectral measure of a self-adjoint random operator [3, 2]. This integral representation of $\boldsymbol{\kappa}^*$ was generalized to the time-dependent case in [4, 9]. Remarkably, this method has also been extended to flows with incompressible *nonzero* effective drift [33], flows where particles diffuse according to linear collisions [34], and solute transport in porous media [8]. All these approaches yield Stieltjes integral representations of the symmetric and, when appropriate, the antisymmetric part of $\boldsymbol{\kappa}^*$.

Homogenization of the advection-diffusion equation for periodic or cellular, incompressible flow fields was treated in [14, 15]. As in the case of random flows, the effective diffusivity tensor $\boldsymbol{\kappa}^*$ is given in terms of a *curl-free* vector field which satisfies a diffusion equation involving a skew-symmetric matrix $\mathbf{H}$. Here, we demonstrate that the ACM can be adapted to this periodic setting to provide a Stieltjes integral representation for $\boldsymbol{\kappa}^*$, both for steady and time-dependent flows, involving a self-adjoint operator and the (non-dimensional) molecular diffusivity ε. In the case of steady fluid velocity fields, the spectral measure is associated with a Hermitian Hilbert-Schmidt integral operator involving the Green's function of the Laplacian on a square. While in the case of dynamic flows, the spectral measure is associated with a Hermitian operator which is the sum of that for steady flows and an unbounded integro-differential operator.

We utalize the analytic structure of the Stieltjes integral representation for $\boldsymbol{\kappa}^*$ to obtain its asymptotic behavior for model flows, as the molecular diffusivity ε tends to zero. This is the high Péclet number regime that is important for the understanding of transport processes in real fluid flows, where the molecular diffusivity is often quite small in comparison. In particular, FINISH THIS PARAGRAPH WHEN WE HAVE CONCRETE RESULTS. necessary and sufficient conditions for steady periodic flow fields $\kappa^* \sim \epsilon^{1/2}$, generically, for steady flows and $\kappa^* \sim O(1)$ for "chaotic" time-dependent flows.

2. Mathematical Methods

Consider the advective difusion of the passive tracer $T(t,\vec{x})$, $\vec{x} \in \mathbb{R}^d$, described by the advection-diffusion equation

$$\partial_t T = \kappa_0 \Delta T + \vec{v} \cdot \vec{\nabla} T, \quad T(0,\vec{x}) = T_0(\vec{x}), \tag{1}$$

with $T(0,\vec{x})$ given. Here, ∂_t denotes partial differentiation with respect to time t, $\Delta = \vec{\nabla} \cdot \vec{\nabla} = \nabla^2$ is the Laplacian, $\kappa_0 > 0$ is the molecular diffusivity, and $\vec{v} = \vec{v}(t,\vec{x})$ is the fluid velocity field, which is assumed to be incompressible, i.e. $\vec{\nabla} \cdot \vec{v} = 0$. We non-dimensionalize equation (1) as follows. Let l and τ be typical length and time scales, respectively, of the problem of interest. Mapping to non-dimensional coordinates $t \mapsto t/\tau$ and $x_i \mapsto x_i/l$ in (1), we have that T satisfies the advection-diffusion equation in (1) with a non-dimensional molecular diffusivity $\varepsilon = \tau\kappa_0/l^2$ and velocity field $\vec{u} = \tau\vec{v}/l$, where x_i is the i^{th} component of the vector $\vec{x}$. Since $\vec{u}$ is incompressible, there is a (non-dimensional) skew-symmetric matrix $\mathbf{H}(t,\vec{x})$, $\mathbf{H}^T = -\mathbf{H}$, such that $\vec{u} = \vec{\nabla} \cdot \mathbf{H}$. Using this representation of the velocity field $\vec{u}$, equation (1) can be written in divergence form,

$$\partial_t T = \vec{\nabla} \cdot \boldsymbol{\kappa} \vec{\nabla} T, \quad T(0,\vec{x}) = T_0(\vec{x}), \tag{2}$$

where $\boldsymbol{\kappa}(t,\vec{x}) = \varepsilon\mathbf{I} + \mathbf{H}(t,\vec{x})$ can be viewed as a local diffusivity tensor with coefficients

$$\kappa_{jk} = \varepsilon\delta_{jk} + H_{jk}, \tag{3}$$

where $\mathbf{I}$ is the identity matrix on $\mathbb{R}^d$, and δ_{jk} is the Kronecker delta.

We are interested in the dynamics of T on *large* length and time scales. Anticipating that T will have diffusive dynamics, we rescale space and time by $\vec{x} \mapsto \vec{x}/\delta$ and $t \mapsto t/\delta^2$, respectively, while keeping the intital condition $T(0,\vec{x}) = T_0(\vec{x})$ independent of δ. This is equivalent to assuming that the initial data is slowly varying relative to the velocity field $\vec{u}$ in the unscaled variables [25, 14, 15]. For periodic diffusivity coefficients in (3) which are uniformly elliptic but not necessarily symmetric, it can be shown [14] that the associated solution $T^\delta(t,\vec{x}) = T(t/\delta^2, \vec{x}/\delta)$ of (2), involving the rescaled local diffusivity tensor $\boldsymbol{\kappa}(t/\delta^2, \vec{x}/\delta)$, converges to $\bar{T}(t,\vec{x})$, which satisfies an anisotropic diffusion equation involving the (constant) diffusivity tensor $\boldsymbol{\kappa}^*$

$$\partial_t \bar{T} = \vec{\nabla} \cdot \boldsymbol{\kappa}^* \vec{\nabla} \bar{T}, \quad \bar{T}(0,\vec{x}) = T_0(\vec{x}). \tag{4}$$

The convergence is in L^2 [14],

$$\lim_{\delta \to 0} \left[\sup_{0 \le t \le t_0} \int \left| T^\delta(t,\vec{x}) - \bar{T}(t,\vec{x}) \right|^2 d\vec{x} \right], \tag{5}$$

for all $t_0 < \infty$.

The effective diffusivity tensor $\boldsymbol{\kappa}^*$ is obtained by solving the cell problem [14]

$$\partial_t \chi = \vec{\nabla} \cdot \boldsymbol{\kappa}(\vec{\nabla}\chi + \vec{e}_j\,), \quad \langle \vec{\nabla}\chi \rangle = 0, \tag{6}$$

for each standard basis vector $\vec{e}_j$, $j = 1, \dots, d$, where $\chi = \chi(t, \vec{x}\,; \vec{e}_j)$. Equation also holds [14] when the velocity field is time-independent, $\vec{u} = \vec{u}(\vec{x})$, however in this case χ is time-independent and $\partial_t \chi = 0$ in (6). The components $\kappa^*_{jk} = \boldsymbol{\kappa}^* \vec{e}_j \cdot \vec{e}_k$ of the effective diffusivity tensor are given by [14]

$$\kappa^*_{jk} = \varepsilon\langle(\vec{\nabla}\chi + \vec{e}_i\,) \cdot (\vec{\nabla}\chi + \vec{e}_j\,)\rangle = \varepsilon(\delta_{jk} + \langle\vec{\nabla}\chi \cdot \vec{\nabla}\chi\rangle). \tag{7}$$

Here, $\langle\cdot\rangle$ denotes spatial averaging over a period cell when the velocity field is time-independent, $\vec{u} = \vec{u}(\vec{x})$, and when the velocity field is time-dependent, $\vec{u} = \vec{u}(t, \vec{x})$, $\langle\cdot\rangle$ denotes time averaging over a temporal period in addition to spacial averaging. Equation (7) demonstrates that the effective thermal transport is always inhanced by the presence of a incompressible velocity field.

Writing $\partial_t\chi = \Delta\Delta^{-1}\partial_t\chi = \vec{\nabla}\cdot\Delta^{-1}\partial_t\vec{\nabla}\chi$ and defining $\vec{E}_k = \vec{\nabla}\chi + \vec{e}_k$ and $\boldsymbol{\sigma} = ((\varepsilon - \Delta^{-1}\partial_t)\mathbf{I} + \mathbf{H})$, equation (6) may be rewritten as [14]

$$\vec{\nabla} \times \vec{E}_k = 0, \quad \vec{\nabla} \cdot \vec{J}_k = 0, \quad \vec{J}_k = \boldsymbol{\sigma}\vec{E}_k, \quad \langle\vec{E}_k\rangle = \vec{e}_k, \tag{8}$$

where Δ^{-1} is based on convolution with the Green's function for the Laplacian Δ and $\boldsymbol{\sigma} = \boldsymbol{\kappa}$ in the time-independent case where $\vec{u} = \vec{u}(\vec{x})$. The formulas in (8) are precisely the electrostatic version of Maxwell's equations for a conductive medium [19], where $\vec{E}_k$ and $\vec{J}_k$ are the local electric field and current density, respectively, and $\boldsymbol{\sigma}$ is the local conductivity tensor of the medium. In the ACM for treating the effective transport properties of composites, the effective conductivity tensor $\boldsymbol{\sigma}^*$ is defined by

$$\langle\vec{J}_k\rangle = \boldsymbol{\sigma}^*\langle\vec{E}_k\rangle. \tag{9}$$

The linear constituative relation $\vec{J}_k = \boldsymbol{\sigma}\vec{E}_k$ in (8) relates the local intensity and flux, while the linear relation in (9) relates the mean intensity and mean flux. By the skew-symmetry of the operator $\mathbf{S} = -\Delta^{-1}\partial_t\mathbf{I} + \mathbf{H}$, the intensity-flux relationship in (8) is similar to that of a Hall medium [21]. Moreover, the skew-symmetry of this operator implies that $\langle\mathbf{S}\vec{E}_k \cdot \vec{E}_k\rangle = -\langle\mathbf{S}\vec{E}_k \cdot \vec{E}_k\rangle = 0$. Consequently by equation (8) and the Helmholtz theorem [12], equation (9) reduces to (7).

$$\sigma^*_{kk} = \langle\boldsymbol{\sigma}\vec{E}_k \cdot \vec{e}_k\rangle = \langle\boldsymbol{\sigma}\vec{E}_k \cdot \vec{E}_k\rangle = \langle[((\varepsilon - \Delta^{-1}\partial_t)\mathbf{I} + \mathbf{H})]\vec{E}_k \cdot \vec{E}_k\rangle = \varepsilon\langle\vec{E}_k \cdot \vec{E}_k\rangle. \tag{10}$$

We will discuss this connection to the ACM in more detail in Section 2.1, where we provide a Stieltjes integral representation for $\boldsymbol{\kappa}^*$, which holds for both for steady and time-dependent flows.

2.1. **Integral representation of the effective diffusivity for steady and dynamic flows.** In this section we adapt the ACM for representing transport incomposites to equations (8) and (9),

In two dimensions, $d = 2$, the matrix $\mathbf{H}$ is determined by a stream function $H(t, \vec{x})$

$$\mathbf{H} = \begin{bmatrix} 0 & H \\ -H & 0 \end{bmatrix}, \qquad \vec{u} = [\partial_{x_1}H,\ \partial_{x_2}H]. \tag{11}$$

Consider the following problems in electrostatics: find stationary random vector fields $\vec{E}(\vec{x},\omega)$ and $\vec{J}(\vec{x},\omega)$ such that

$$\vec{\nabla}\times\vec{E}=0,\quad \vec{\nabla}\cdot\vec{J}=0,\quad \vec{J}=\boldsymbol{\sigma}\vec{E},\quad \langle\vec{E}\,\rangle=\vec{E}_0, \tag{12}$$

$$\vec{\nabla}\times\vec{E}=0,\quad \vec{\nabla}\cdot\vec{J}=0,\quad \vec{E}=\boldsymbol{\rho}\vec{J},\quad \langle\vec{J}\,\rangle=\vec{J}_0, \tag{13}$$

where $\langle\cdot\rangle$ denotes ensemble average over Ω with respect to the measure P, we use the simplified vector notation $\langle\vec{\xi}\,\rangle=(\langle\xi_1\rangle,\ldots,\langle\xi_d\rangle)^T$, and ξ_i is the $i^{\,\mathrm{th}}$ component of the vector $\vec{\xi}$. Here, $\vec{E}$ and $\vec{J}$ are the electric field and current density within the polycrystalline medium, respectively. Their averages $\vec{E}_0$ and $\vec{J}_0$ are assumed to be *given*, and by stationarity they are independent of $\vec{x}\in\mathbb{R}^d$. More specifically, we seek stationary solutions $\vec{E}$ and $\vec{J}$ to each of equations (12) and (13) of the form

$$\vec{E}(\vec{x},\omega)=\vec{E}\,'(\tau_{-x}\,\omega),\quad \vec{J}(\vec{x},\omega)=\vec{J}\,'(\tau_{-x}\,\omega),\quad \forall\;\vec{x}\in\mathbb{R}^d,\;\omega\in\Omega, \tag{14}$$

where $\vec{E}\,'(\omega)=\vec{E}(0,\omega)$, $\vec{J}\,'(\omega)=\vec{J}(0,\omega)$, $\vec{E}_0=\langle\vec{E}\,'(\omega)\rangle$, and $\vec{J}_0=\langle\vec{J}\,'(\omega)\rangle$. Once suitable solutions are found, the effective conductivity and resistivity tensors $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$ are defined by [19]

$$\langle\vec{J}\,\rangle=\boldsymbol{\sigma}^*\vec{E}_0 \quad\text{and}\quad \langle\vec{E}\,\rangle=\boldsymbol{\rho}^*\vec{J}_0. \tag{15}$$

We will prove the existence and uniqueness of such solutions to equations (12) and (13), as the setup of the proof illustrates the mathematical properties of $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$ which lead to the desired integral representations of these effective parameters. From equations (**??**), (14) and (15), we see that the effective parameters are independent of $\vec{x}\in\mathbb{R}^d$.

2.1.1. *Existence and uniqueness for the electrostatic problem.* If the components of the vector fields $\vec{E}$ and $\vec{J}$ were continuously differentiable on all of $\mathbb{R}^d$, for every $\omega\in\Omega$, then, since $\vec{\nabla}\times\vec{E}=0$ and $\vec{\nabla}\cdot\vec{J}=0$, there would exist [22] scalar and vector potentials φ and $\vec{A}$ such that $\vec{E}=\vec{\nabla}\varphi$ and $\vec{J}=\vec{\nabla}\times\vec{A}$. In this case, equations (12) and (13) are equivalent to

$$\vec{\nabla}\cdot\boldsymbol{\sigma}\vec{\nabla}\varphi=0 \quad\text{and}\quad \vec{\nabla}\times(\boldsymbol{\rho}\,\vec{\nabla}\times\vec{A}\,)=0, \tag{16}$$

respectively, with $\langle\vec{\nabla}\varphi\rangle=\vec{E}_0$ and $\langle\vec{\nabla}\times\vec{A}\,\rangle=\vec{J}_0$. However, σ_{jk} and ρ_{jk} may be discontinuous, hence non-differentiable, across the crystallite boundaries of the polycrystalline medium, so the formulas in equation (16) may not exist in a classical sense. The weak forms [16] of these formulas are given by

$$\langle\boldsymbol{\sigma}\vec{\nabla}\varphi\cdot\vec{\nabla}\psi\rangle_V=0 \quad\text{and}\quad \langle\boldsymbol{\rho}\vec{\nabla}\times\vec{A}\cdot\vec{\nabla}\times\vec{\xi}\,\rangle_V=0, \tag{17}$$

where $\langle\cdot\rangle_V$ denotes integration over all of $\mathbb{R}^d$, ψ and ξ_i, $i=1,\ldots,d$, are infinitely differentiable functions with compact support, and we stress that $\vec{\nabla}\psi$ is a curl-free vector field and $\vec{\nabla}\times\vec{\xi}$ is a divergence-free vector field. This would directly bypass the non-differentiability of σ_{jk} and ρ_{jk}. Although, indirectly, the discontinuous nature of these functions might still render $\vec{E}$ and $\vec{J}$ non-differentiable, so that the differential equations in (12) and (13) involving these vector fields again, may not exist in a classical sense. We address such issues by providing abstract Hilbert space formulations [19] of the equations in (17).

The group of transformations τ_x acting on Ω induces a group of operators T_x on the Hilbert space $L^2(\Omega,P)$ defined by $(T_xf)(\omega)=f(\tau_{-x}\,\omega)$ for all $f\in L^2(\Omega,P)$. Since the group generated by τ_x (through composition) is measure preserving, the operators T_x form a unitary group and therefore have closed densely defined infinitesimal generators L_i in each direction $i=1,\ldots,d$ with domain $\mathscr{D}_i\subset L^2(\Omega,P)$ [19, 32]. Thus,

$$L_i=\frac{\partial}{\partial x_i}T_x\bigg|_{x=0},\quad i=1,\ldots,d, \tag{18}$$

where differentiation in (18) is defined in the sense of convergence in $L^2(\Omega,P)$ for elements of $\mathscr{D}_i$ [19, 32]. The closed subset $\mathscr{D}=\cap_{i=1}^d\mathscr{D}_i$ of $L^2(\Omega,P)$ is a Hilbert space with inner product $\langle\cdot,\cdot\rangle_D$ given by $\langle f,g\rangle_D=\langle f,g\rangle_{L^2}+\sum_{i=1}^d\langle L_if,L_ig\rangle_{L^2}$, where $\langle\cdot,\cdot\rangle_{L^2}$ is the $L^2(\Omega,P)$ inner product [19, 32].

Now consider the Hilbert space $\mathscr{H}=\bigotimes_{i=1}^d L^2(\Omega,P)$ with inner product $\langle\cdot,\cdot\rangle$ defined by $\langle\vec{\xi},\vec{\zeta}\rangle=\langle\vec{\xi}\cdot\vec{\zeta}\rangle$, and define the Hilbert spaces of "curl free" $\mathscr{H}_\times$ and "divergence free" $\mathscr{H}_\bullet$ random fields [19]

$$\mathscr{H}_\times=\left\{\vec{\xi}'(\omega)\in\mathscr{H}\ \mid\ L_i\xi_j'-L_j\xi_i'=0\ \text{ weakly and }\langle\vec{\xi}'\,\rangle=0\right\},\tag{19}$$

$$\mathscr{H}_\bullet=\left\{\vec{\zeta}'(\omega)\in\mathscr{H}\ \middle|\ \sum_{i=1}^d L_i\zeta_i'=0\ \text{ weakly and }\langle\vec{\zeta}'\,\rangle=0\right\}.$$

We write $\vec{E}(\vec{x},\omega)=\vec{E}_0+\vec{E}_f(\vec{x},\omega)$, where $\vec{E}_f$ is the fluctuating field of mean zero about the (constant) average $\vec{E}_0$, and similarly for $\vec{J}(\vec{x},\omega)=\vec{J}_0+\vec{J}_f(\vec{x},\omega)$. In view of equations (**??**), (14), and (17), we consider the following variational problems [19]: given $\boldsymbol{\sigma}'$ and $\vec{E}_0$, and $\boldsymbol{\rho}'$ and $\vec{J}_0$, find $\vec{E}_f'(\omega)\in\mathscr{H}_\times$ and $\vec{J}_f'(\omega)\in\mathscr{H}_\bullet$ such that

$$\langle\boldsymbol{\sigma}'(\omega)(\vec{E}_0+\vec{E}_f'(\omega))\cdot\vec{\xi}'(\omega)\rangle=0\quad\forall\ \vec{\xi}'(\omega)\in\mathscr{H}_\times\tag{20}$$

$$\langle\boldsymbol{\rho}'(\omega)(\vec{J}_0+\vec{J}_f'(\omega))\cdot\vec{\zeta}'(\omega)\rangle=0\quad\forall\ \vec{\zeta}'(\omega)\in\mathscr{H}_\bullet\,,\tag{21}$$

respectively. In order to apply the Lax-Milgram Theorem, we rewrite equations (20) and (21) as

$$\Phi(\vec{E}_f',\vec{\xi}')=\langle\boldsymbol{\sigma}'(\omega)\vec{E}_f'(\omega)\cdot\vec{\xi}'(\omega)\rangle=-\langle\boldsymbol{\sigma}'(\omega)\vec{E}_0\cdot\vec{\xi}'(\omega)\rangle=f_\sigma(\vec{\xi}'),\tag{22}$$

$$\Psi(\vec{J}_f',\vec{\zeta}')=\langle\boldsymbol{\rho}'(\omega)\vec{J}_f'(\omega)\cdot\vec{\zeta}'(\omega)\rangle=-\langle\boldsymbol{\rho}'(\omega)\vec{J}_0\cdot\vec{\zeta}'(\omega)\rangle=f_\rho(\vec{\zeta}'),\tag{23}$$

respectively.

By the Cauchy–Schwartz inequality and equation (**??**), each of the bilinear functionals Φ and Ψ are bounded, and f_σ and f_ρ are both bounded linear functionals on the Hilbert spaces $\mathscr{H}_\times$ and $\mathscr{H}_\bullet$, respectively. For now, we will assume that Φ and Ψ are coercive, e.g. that there exists a positive constant $\kappa>0$ such that $\Phi(\vec{\xi},\vec{\xi}\,)\geq\kappa\|\vec{\xi}\,\|^2$ for all $\vec{\xi}\in\mathscr{H}_\times$, where $\|\cdot\|$ denotes the norm induced by the $\mathscr{H}$-inner-product. Later, we will demonstrate that the coercivity condition determines analytic properties of the effective parameters. By the Lax-Milgram theorem, there exist unique $\vec{E}_f'\in\mathscr{H}_\times$ and $\vec{J}_f'\in\mathscr{H}_\bullet$ that satisfy equations (22) and (23) for all $\vec{\xi}'\in\mathscr{H}_\times$ and $\vec{\zeta}'\in\mathscr{H}_\bullet$, respectively. By construction,

$$\vec{E}'(\omega)=\vec{E}_0+\vec{E}_f'(\omega),\qquad\vec{J}'(\omega)=\boldsymbol{\sigma}'(\omega)\vec{E}'(\omega),\tag{24}$$

$$\vec{J}'(\omega)=\vec{J}_0+\vec{J}_f'(\omega),\qquad\vec{E}'(\omega)=\boldsymbol{\rho}'(\omega)\vec{J}'(\omega),\tag{25}$$

are the unique solutions of equations (12) and (13), respectively, via equations (**??**) and (14). To simplify notation, we will henceforth drop the distinction between the primed variables $\vec{E}_f'(\omega)$ and $\vec{E}_f(\vec{x},\omega)$, for example, as the context of each notation is now clear.

We conclude this section by noting that since $\vec{E}_f\in\mathscr{H}_\times$ and $\vec{J}_f\in\mathscr{H}_\bullet$, equations (20) and (21) yield the energy (power) [22] constraints $\langle\vec{J}\cdot\vec{E}_f\rangle=0$ and $\langle\vec{E}\cdot\vec{J}_f\rangle=0$, respectively, which lead to the following reduced energy representations $\langle\vec{J}\cdot\vec{E}\,\rangle=\langle\vec{J}\,\rangle\cdot\vec{E}_0$ and $\langle\vec{E}\cdot\vec{J}\,\rangle=\langle\vec{E}\,\rangle\cdot\vec{J}_0$. By equation (15), we have the following energy representations involving the effective parameters

$$\langle\vec{J}\cdot\vec{E}\rangle=\boldsymbol{\sigma}^*\vec{E}_0\cdot\vec{E}_0=\boldsymbol{\rho}^*\vec{J}_0\cdot\vec{J}_0.\tag{26}$$

2.1.2. *Representation formulas for uniaxial polycrystalline media.* In Section 2.1.1 we proved that equations (12) and (13) have unique solutions when the tensors $\boldsymbol{\sigma}(\vec{x},\omega)$ and $\boldsymbol{\rho}(\vec{x},\omega)$ satisfy the boundedness conditions in (**??**), and when the bilinear functionals Φ and Ψ of equations (22) and (23) are coercive. In this section, we derive Stieltjes integral representations for the effective conductivity and resistivity tensors $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$, involving spectral measures of self-adjoint random operators, which depend only on the geometry of the polycrystallin medium. We will also show that the coercivity conditions on Φ and Ψ determine analytic properties these representations.

In this section, we restrict our attention to polycrystalline media with uniaxial microscopic asymmetry. We also interpret the electrostatic version of Maxwell's equations in (12) and (13) as the quasi-static limit of the full set of Maxwell's equations. In this case, these equations describe the transport properties of an electromagnetic wave through a polycrystalline medium, when the wavelength is very large compared to the typical size of the crystallites within the random medium. Under these assumptions, the local conductivity along one of the crystallite axes has the *complex* value σ_1, while the conductivity along all the other axes have the complex value σ_2. The conductivity and resistivity tensors of such polycrystalline media are given by

$$\begin{aligned}\boldsymbol{\sigma}(\vec{x},\omega) &= R^T(\vec{x},\omega)\mathrm{diag}(\sigma_1,\sigma_2,\ldots,\sigma_2)R(\vec{x},\omega),\\ \boldsymbol{\rho}(\vec{x},\omega) &= R^T(\vec{x},\omega)\mathrm{diag}(1/\sigma_1,1/\sigma_2,\ldots,1/\sigma_2)R(\vec{x},\omega),\end{aligned} \tag{27}$$

where $R(\vec{x},\omega)$ is a rotation matrix satisfying $R^T = R^{-1}$. For example, when $d = 2$ we have

$$\boldsymbol{\sigma} = R^T \begin{bmatrix} \sigma_1 & 0 \\ 0 & \sigma_2 \end{bmatrix} R, \quad \boldsymbol{\rho} = R^T \begin{bmatrix} 1/\sigma_1 & 0 \\ 0 & 1/\sigma_2 \end{bmatrix} R, \qquad R = \begin{bmatrix} \cos\theta & -\sin\theta \\ \sin\theta & \cos\theta \end{bmatrix}, \tag{28}$$

where $\theta = \theta(\vec{x},\omega)$ is the orientation angle, measured from the direction $\vec{e}_1$, of the polycrystallite which has an interior containing $\vec{x} \in \mathbb{R}^d$ for $\omega \in \Omega$. Here, $\vec{e}_j$, $j = 1,\ldots,d$, are standard basis vectors with components $(\vec{e}_j)_k = \delta_{jk}$ and δ_{jk} is the Kronecker delta. In higher dimensions, $d \geq 3$, the rotation matrix R is a composition of "basic" rotation matrices R_i, e.g. $R = \prod_{j=1}^d R_j$, where the matrix $R_j(\vec{x},\omega)$ rotates vectors in $\mathbb{R}^d$ by an angle $\theta_j = \theta_j(\vec{x},\omega)$ about the $\vec{e}_j$ axis. For example, in three dimensions

$$R_1 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\theta_1 & -\sin\theta_1 \\ 0 & \sin\theta_1 & \cos\theta_1 \end{bmatrix}, \quad R_2 = \begin{bmatrix} \cos\theta_2 & 0 & \sin\theta_2 \\ 0 & 1 & 0 \\ -\sin\theta_2 & 0 & \cos\theta_2 \end{bmatrix}, \quad R_3 = \begin{bmatrix} \cos\theta_3 & -\sin\theta_3 & 0 \\ \sin\theta_3 & \cos\theta_3 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{29}$$

Equation (27) can be written in a more suggestive form in terms of the matrix $C = \mathrm{diag}(1,0,\ldots,0)$

$$\boldsymbol{\sigma}(\vec{x},\omega) = \sigma_1 X_1(\vec{x},\omega) + \sigma_2 X_2(\vec{x},\omega), \qquad \boldsymbol{\rho}(\vec{x},\omega) = X_1(\vec{x},\omega)/\sigma_1 + X_2(\vec{x},\omega)/\sigma_2. \tag{30}$$

Here $X_1 = R^T C R$ and $X_2 = R^T(I - C)R$, where I is the identity matrix on $\mathbb{R}^d$. Since $R^T = R^{-1}$ and C is a diagonal projection matrix satisfying $C^2 = C$, it is clear that the X_i, $i = 1,2$, are mutually orthogonal projection matrices satisfying

$$X_j^T = X_j, \quad X_j X_k = X_j \delta_{jk}, \quad X_1 + X_2 = I. \tag{31}$$

By equation (**??**), we have the existence of measurable functions $[X_i'(\omega)]_{jk}$, $j,k = 1,\ldots,d$, such that $[X_i(\vec{x},\omega)]_{jk} = [X_i'(\tau_{-x}\,\omega)]_{jk}$ for all $\vec{x} \in \mathbb{R}^d$ and $\omega \in \Omega$. From equations (15) and (30), we see that $\sigma^*_{jk}(a\sigma_1, a\sigma_2) = a\sigma^*_{jk}(\sigma_1,\sigma_2)$, for any complex number a, and similarly for the ρ^*_{jk}. Due to this homogeneity of these functions, 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

$$m_{jk}(h) = \sigma^*_{jk}/\sigma_2, \quad w_{jk}(z) = \sigma^*_{jk}/\sigma_1, \quad \tilde{m}_{jk}(h) = \sigma_1\rho^*_{jk}, \quad \tilde{w}_{jk}(z) = \sigma_2\rho^*_{jk}. \tag{32}$$

where $z = 1/h$. For example, from equations (15) and (30) we have

$$\boldsymbol{\sigma}^*(h)\vec{E}_0 = \sigma_2 \left\langle (hX_1 + X_2)\vec{E} \right\rangle, \quad \boldsymbol{\rho}^*(h)\vec{J}_0 = (1/\sigma_1) \left\langle (X_1 + hX_2)\vec{J} \right\rangle \tag{33}$$

We now show that the coercivity conditions on the bilinear functionals Φ and Ψ of equations (22) and (23) imply that

$$\begin{aligned}\sigma^*_{jk}(h) \text{ and } \rho^*_{jk}(h) &\text{ are analytic functions of the complex variable } h \\ &\text{everywhere except on the negative real axis.}\end{aligned} \tag{34}$$

We only prove the statement in (34) for $\sigma^*_{jk}(h)$, as the proof involving $\rho^*_{jk}(h)$ is analogous. The bounded bilinear functional Φ is coercive if there exists a $\kappa>0$ such that $|\Phi(\vec{\xi},\vec{\xi})|\geq\kappa\|\vec{\xi}\,\|^2$ for all $\vec{\xi}\in\mathscr{H}_\times$ such that $\|\vec{\xi}\,\|\neq 0$. From equations (22) and (30) this is true only if

$$\left|\left\langle (hX_1+X_2)\vec{\xi}\cdot\overline{\vec{\xi}}\right\rangle\right|\geq(\kappa/|\sigma_2|)\|\vec{\xi}\,\|^2, \tag{35}$$

where $\vec{\xi}$ is complex-valued when h is complex and $\overline{\vec{\xi}}$ is the complex conjugate of the vector $\vec{\xi}$. Define ν by the ratio

$$\nu=\frac{\left\langle X_1\vec{\xi}\cdot\overline{\vec{\xi}}\right\rangle}{\|\vec{\xi}\,\|^2}. \tag{36}$$

Since X_1 is a projection matrix, it is bounded by 1 in operator norm. Moreover, we have that $\nu\geq 0$ as $\langle X_1\vec{\xi}\cdot\overline{\vec{\xi}}\,\rangle=\langle X_1\vec{\xi}\cdot X_1\overline{\vec{\xi}}\,\rangle=\|X_1\vec{\xi}\,\|^2\geq 0$. Therefore, the Cauchy–Schwartz inequality and the definition of the operator norm [17] implies that $0\leq\nu\leq 1$. Since $X_2=I-X_1$, equation (35) can now be written as

$$|h\nu+1-\nu|\geq\kappa/|\sigma_2|>0. \tag{37}$$

Let $h=h_r+\mathrm{i}h_i$, where h_r and h_i are the real and imaginary parts of the complex number h, respectively. Since $|h\nu+1-\nu|^2=|1+\nu(h_r-1)|^2+\nu^2|h_i|^2$, the formula in (37) always holds when $h_i\neq 0$. If h is real, then $1+\nu(h-1)=0$ if and only if $h=1-1/\nu$ for $0<\nu\leq 1$, i.e. for h on the negative real axis. In conclusion, equation (37) holds if and only if h is *off* of the negative real axis including zero [19]. For any such complex value of h, Φ is coercive and there exists a unique solution $\vec{E}_f$ to equation (22), hence to equation (12). By differentiation with respect to h, one deduces easily (WHY?) that $\vec{E}_f$ is analytic in h off the negative real axis with values in $\mathscr{H}_\times$. Therefore by equation (33), $\sigma^*_{jk}(h)$ is analytic off the negative real axis in the complex h-plane. This concludes our proof of the statement in equation (34).

The above analysis [19] demonstrates that the dimensionless functions $m_{jk}(h)$ and $\tilde{m}_{jk}(h)$ in (32) 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. By equation (33) and its counterpart involving $z=1/h$, each take the corresponding upper half plane to the upper half plane and are therefore examples of Herglotz functions [11, 19].

A key step in the ACM is obtaining Stieltjes integral representations for $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$. These follow from resolvent representations for the electric field $\vec{E}$ [19] and current density $\vec{J}$ [31]

$$\begin{aligned}\vec{E}&=s(sI-\Gamma X_1)^{-1}\vec{E}_0=t(tI-\Gamma X_2)^{-1}\vec{E}_0, \quad s\in\mathbb{C}\backslash[0,1],\\ \vec{J}&=t(tI-\Upsilon X_1)^{-1}\vec{J}_0=s(sI-\Upsilon X_2)^{-1}\vec{J}_0, \quad t\in\mathbb{C}\backslash[0,1],\end{aligned} \tag{38}$$

where we have defined the complex variables $s=1/(1-h)$ and $t=1/(1-z)=1-s$. The (non-random) operator $\Gamma=\vec{\nabla}(\Delta^{-1})\vec{\nabla}\cdot$ is based on convolution with the free-space Green's function for the Laplacian $\Delta=\vec{\nabla}\cdot\vec{\nabla}=\nabla^2$, and the (non-random) operator $\Upsilon=-\vec{\nabla}\times(\vec{\nabla}\times\vec{\nabla}\times)^{-1}\vec{\nabla}\times$ involves the vector Laplacian $\boldsymbol{\Delta}=-\vec{\nabla}\times\vec{\nabla}\times+\vec{\nabla}\vec{\nabla}\cdot$ for $d=2,3$ [19, 31]. These (projection) operators are discussed in more detail below.

If the current density $\vec{J}(\vec{x},\omega)$ and the electric field $\vec{E}(\vec{x},\omega)$ are sufficiently smooth for all $\vec{x}\in\mathbb{R}^d$ when $\omega\in\Omega$, equation (38) is obtained as follows. The operator Δ^{-1} is well defined in terms of convolution with respect to the free-space Green's function of the Laplacian Δ [19, 16]. Similarly, on the Hilbert space $\mathscr{H} = \bigotimes_{i=1}^{d} L^2(\Omega, P)$, the inverse $\boldsymbol{\Delta}^{-1}$ of the vector Laplacian $\boldsymbol{\Delta}$ 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 (12) yields $\Gamma\vec{J} = 0$, where $\Gamma = \vec{\nabla}(\Delta^{-1})\vec{\nabla}\cdot$ is an orthogonal projection [19] 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 $\vec{\xi} \in \mathscr{H}_\times$ there exists [22] a scalar potential φ which is unique up to a constant such that $\vec{\xi} = \vec{\nabla}\varphi$. Consequently, it is clear that $\Gamma\vec{\xi} = \vec{\xi}$ for all such $\vec{\xi} \in \mathscr{H}_\times$.

In order to discuss analogous properties of divergence free vector fields, we restrict our attention to $d = 2, 3$, and avoid a more involved discussion regarding differential forms. Applying the operator $\vec{\nabla} \times (\vec{\nabla} \times \vec{\nabla}\times)^{-1}$ to the formula $\vec{\nabla} \times \vec{E} = 0$, we have $\Upsilon\vec{E} = 0$, where $\Upsilon = -\vec{\nabla} \times (\boldsymbol{\Delta}^{-1})\vec{\nabla}\times$ is an orthogonal projection from $\mathscr{H}$ onto the Hilbert space $\mathscr{H}_\bullet$ of divergence-free random fields (of transverse gauge) [31]. 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 the vector potential associated with $\vec{\zeta}$ and the arbitrary vector field $\vec{C}$ satisfies $\vec{\nabla} \times \vec{C} = 0$ [22]. Without loss of generality, $\vec{C}$ can be chosen so that $\vec{A}$ satisfies $\vec{\nabla} \cdot \vec{A} = 0$ [22]. Hence, $\vec{\nabla} \times \vec{\zeta} = \vec{\nabla} \times \vec{\nabla} \times \vec{A} = \vec{\nabla}(\vec{\nabla} \cdot \vec{A}) - \boldsymbol{\Delta}\vec{A} = -\boldsymbol{\Delta}\vec{A}$. The vector $\vec{C}$ chosen in this manner gives the transverse *gauge* of $\vec{\zeta}$ [22]. Choosing the members of $\mathscr{H}_\bullet$ to have transverse gauge, the action of $\vec{\nabla} \times \vec{\nabla}\times$ on $\mathscr{H}_\bullet$ is given by that of $-\boldsymbol{\Delta}$. Therefore, the action of Υ on $\mathscr{H}_\bullet$ is given by that of

$$\Upsilon = \vec{\nabla} \times (\vec{\nabla} \times \vec{\nabla}\times)^{-1}\vec{\nabla}\times = -\vec{\nabla} \times (\boldsymbol{\Delta}^{-1})\vec{\nabla}\times, \tag{39}$$

and it is clear from the above discussion that $\Upsilon\vec{\zeta} = \vec{\zeta}$ for all such $\vec{\zeta} \in \mathscr{H}_\bullet$.

We now derive the formulas in equation (38). Using $h = 1 - 1/s$, $z = 1 - 1/t$ and $X_1 + X_2 = I$, we write σ and ρ in equation (30) as $\boldsymbol{\sigma} = \sigma_2(I - X_1/s) = \sigma_1(I - X_2/t)$ and $\boldsymbol{\rho} = (I - X_2/s)/\sigma_1 = (I - X_1/t)/\sigma_2$. Recall 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 (38)

$$\vec{E}_f = \frac{1}{s}\Gamma X_1\vec{E} = \frac{1}{t}\Gamma X_2\vec{E}, \qquad \vec{J}_f = \frac{1}{t}\Upsilon X_1\vec{J} = \frac{1}{s}\Upsilon X_2\vec{J}. \tag{40}$$

In general, the differential opperators $\vec{\nabla}$, $\vec{\nabla}\cdot$, and $\vec{\nabla}\times$ are interpreted in a weak sense in terms of the operators L_i in (18).

We now derive integral representations for the effective parameters of uniaxial polycrystalline media, involving the resolvent formulas in (38). For the formulation of the effective parameter problem involving $\mathscr{H}_\times$ and $\boldsymbol{\sigma}^*$, define the coordinate system so that in (15) the constant vector $\vec{E}_0 = \langle\vec{E}\rangle$ is given by $\vec{E}_0 = E_0\,\vec{e}_j$. In the other formulation involving $\mathscr{H}_\bullet$ and $\boldsymbol{\rho}^*$, define $\vec{J}_0 = J_0\,\vec{e}_j$. Recalling that $X_1 + X_2 = I$ yields $\boldsymbol{\sigma} = \sigma_2(I - X_1/s)$ and $\boldsymbol{\rho} = (I - X_2/s)/\sigma_1$ in (30), equations (33) and (38) imply that $\sigma^*_{jk} = \boldsymbol{\sigma}^*\vec{e}_j \cdot \vec{e}_k$ and $\rho^*_{jk} = \boldsymbol{\rho}^*\vec{e}_j \cdot \vec{e}_k$ satisfy

$$\sigma^*_{jk} = \sigma_2(\delta_{jk} - \langle X_1(sI - \Gamma X_1)^{-1}\vec{e}_j \cdot \vec{e}_k\rangle), \quad \rho^*_{jk} = (1/\sigma_1)(\delta_{jk} - \langle X_2(sI - \Upsilon X_2)^{-1}\vec{e}_j \cdot \vec{e}_k\rangle), \tag{41}$$

and similar formulas involving the contrast parameter t. Therefore, 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 [19].

On the Hilbert space $\mathscr{H}_\times$, the operators Γ and X_i, $i = 1, 2$, act as projectors. Therefore, $M_i = X_i\Gamma X_i$, $i = 1, 2$, are compositions of projection operators on $\mathscr{H}_\times$, and are consequently positive definite and bounded by 1 in the underlying operator norm [36]. They are self-adjoint with respect to the $\mathscr{H}$-inner-product $\langle\cdot, \cdot\rangle$. Therefore, on the Hilbert space $\mathscr{H}_\times$ with weight X_1 in the inner-product, $\langle\cdot,\cdot\rangle_1 = \langle X_1\,\cdot,\cdot\rangle$ for example, ΓX_1 is a bounded linear self-adjoint operator with spectrum contained in the interval $[0,1]$ [19, 16, 36]. Hence the resolvent operator $(sI-\Gamma X_1)^{-1}$ in (38) is also a linear self-adjoint operator with respect to the same inner-product, and is bounded for $s\in\mathbb{C}\backslash[0,1]$ [39]. Similarly, $(tI-\Upsilon X_1)^{-1}$ in (38) is a linear self-adjoint operator on $\mathscr{H}_\bullet$ with respect to the inner-product $\langle\cdot,\cdot\rangle_1$, and is bounded for $t\in\mathbb{C}\backslash[0,1]$.

By the spectral theorem for such operators [35, 39], there exists an increasing family of self-adjoint projection operators $\{Q(\lambda)\}$ - the resolution of the identity - that satisfy $Q(0)=0$ and $Q(1)=I$ such that

$$f(M_1)=\int_0^1 f(\lambda)Q(d\lambda),\quad \langle f(M_1)\,\vec{e}_j\cdot\vec{e}_k\rangle_1=\int_0^1 f(\lambda)\mu_{jk}(d\lambda), \tag{42}$$

for example, 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, $Q(d\lambda)$ is the projection valued measure associated with the operator $Q(\lambda)$ [35], and $\mu_{jk}(d\lambda)=\langle Q(d\lambda)\,\vec{e}_j\cdot\vec{e}_k\rangle_1$, $j,k=1,\dots,d$, are the components of the matrix valued *spectral measure* $\boldsymbol{\mu}(d\lambda)$ in the $(\vec{e}_j,\vec{e}_k)$ state [19, 35, 39]. As the spectrum of the operator M_1 is contained in the interval $[0,1]$, the support Σ_{jk} of the measure μ_{jk} satisfies $\Sigma_{jk}\subseteq[0,1]$ [35]. Analogous results hold for the operators M_2 and $K_i=X_i\Upsilon X_i$, $i=1,2$. In view of the formulas in equation (41) and their counterparts involving the contrast parameter t, setting $f(\lambda)=(s-\lambda)^{-1}$ in (42), for example, yields the following integral representations for the effective parameters σ^*_{jk} and ρ^*_{jk}

$$\begin{aligned}
m_{jk}(h)&=\delta_{jk}-F_{jk}(s), &\quad F_{jk}(s)&=\langle X_1(sI-\Gamma X_1)^{-1}\vec{e}_j\cdot\vec{e}_k\rangle=\int_0^1\frac{\mu_{jk}(d\lambda)}{s-\lambda}\,,\\
w_{jk}(z)&=\delta_{jk}-G_{jk}(t), &\quad G_{jk}(t)&=\langle X_2(tI-\Gamma X_2)^{-1}\vec{e}_j\cdot\vec{e}_k\rangle=\int_0^1\frac{\alpha_{jk}(d\lambda)}{t-\lambda}\,,\\
\tilde{m}_{jk}(h)&=\delta_{jk}-E_{jk}(s), &\quad E_{jk}(s)&=\langle X_2(sI-\Upsilon X_2)^{-1}\vec{e}_j\cdot\vec{e}_k\rangle=\int_0^1\frac{\eta_{jk}(d\lambda)}{s-\lambda}\,,\\
\tilde{w}_{jk}(z)&=\delta_{jk}-H_{jk}(t), &\quad H_{jk}(t)&=\langle X_1(tI-\Upsilon X_1)^{-1}\vec{e}_j\cdot\vec{e}_k\rangle=\int_0^1\frac{\kappa_{jk}(d\lambda)}{t-\lambda}\,.
\end{aligned}\tag{43}$$

Here, μ_{jk} and α_{jk} are *spectral measures* associated with the random operators $X_1\Gamma X_1$ and $X_2\Gamma X_2$, respectively, while η_{jk} and κ_{jk} are spectral measures associated with the random operators $X_2\Upsilon X_2$ and $X_1\Upsilon X_1$, respectively.

By the Stieltjes–Perron inversion theorem [20, 29], the spectral measure $\boldsymbol{\mu}$, for example, is given by the weak limit $\boldsymbol{\mu}(d\lambda)=-\lim_{\epsilon\downarrow0}\mathrm{Im}(\mathbf{F}(\lambda+\mathrm{i}\epsilon))(d\lambda/\pi)$, i.e.

$$\int_0^1\xi(\lambda)\;\boldsymbol{\mu}(d\lambda)=-\frac{1}{\pi}\lim_{\epsilon\downarrow0}\int_0^1\xi(\lambda)\;\mathrm{Im}(\mathbf{F}(\lambda+\mathrm{i}\epsilon))\,d\lambda, \tag{44}$$

for all smooth scalar test functions $\xi(\lambda)$, where $(\mathbf{F}(s))_{jk}=F_{jk}(s)$. From equation (44) and the identities $m_{jk}(h)=h\,w_{jk}(z)$ and $\tilde{m}_{jk}(h)=h\,\tilde{w}_{jk}(z)$, which follow from equation (32), it can be shown [31] that the measures μ_{jk} and α_{jk}, and the measures η_{jk} and κ_{jk} are related by

$$\begin{aligned}
\lambda\alpha_{jk}(\lambda)&=(1-\lambda)\mu_{jk}(1-\lambda)+\lambda\,(\,m_{jk}(0)\delta_0(d\lambda)+w_{jk}(0)(\lambda-1)\delta_1(d\lambda)\,),\\
\lambda\kappa_{jk}(\lambda)&=(1-\lambda)\eta_{jk}(1-\lambda)+\lambda\,(\,\tilde{m}_{jk}(0)\delta_0(d\lambda)+\tilde{w}_{jk}(0)(\lambda-1)\delta_1(d\lambda)\,).
\end{aligned}\tag{45}$$

Here, $m(0)=m(h)|_{h=0}$ and $w(0)=w(z)|_{z=0}$, for example, and $\delta_a(d\lambda)$ is the delta measure concentrated at $\lambda=a$. Equations (43) and (45) 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 μ_{jk}, α_{jk}, η_{ij}, and κ_{jk}. Because of these symmetries, for simplicity, we will focus on $m_{jk}(h)$ and μ_{jk}, and will refer to the other functions and measures where appropriate.

A key feature of equations (15), (32), and (43) is that parameter information in h and E_0 is *separated* from the geometry of the polycrystalline medium, which is encoded in the spectral measure μ_{jk} via its moments

$$\mu^n_{jk} = \int_0^1 \lambda^n \mu_{jk}(d\lambda) = \langle X_1[\Gamma X_1]^n\, \vec{e}_j \cdot \vec{e}_k\rangle, \quad n = 0, 1, 2, \ldots, \tag{46}$$

which clearly contains statistical information regarding the random projection matrix X_1. The mass μ^0_{jk} of the measure μ_{jk} is given by

$$\mu^0_{jk} = \langle X_1\vec{e}_j \cdot \vec{e}_k\rangle, \quad \mu^0_{kk} = \langle |X_1\vec{e}_k|^2\rangle, \tag{47}$$

where we used equation (31) in the second formula. From this we see that the diagonal components μ_{kk}, $k = 1 \ldots, d$, of $\boldsymbol{\mu}$ are positive measures. Since the projection matrix X_1 is bounded by one in the operator norm, the Cauchy–Schwartz inequality and (47) imply that $0 \le \mu^0_{kk} \le 1$. For example, in the case of two-dimensional polycrystalline media, $d = 2$, equation (28) implies that

$$\mu^0_{11} = \langle \cos^2\theta\rangle, \quad \mu^0_{22} = \langle \sin^2\theta\rangle, \quad \mu^0_{12} = \langle \sin\theta\cos\theta\rangle. \tag{48}$$

Equation (47) follows by setting $f(M_1) = I$ $(f(\lambda) = 1)$ in equation (42), which implies that the projection valued measure satisfies $\int_0^1 Q(d\lambda) = I$ [35, 39]. Moreover, recall that the associated operator $Q(\lambda)$ is a *self-adjoint projector* on $\mathscr{H}_\times$ for $\lambda \in [0, 1]$ [35, 39]. Consequently, we have

$$\begin{aligned} \mu^0_{jk} &= \int_0^1 \langle Q(d\lambda)\vec{e}_j \cdot \vec{e}_k\rangle_1 = \langle \vec{e}_j \cdot \vec{e}_k\rangle_1 = \langle X_1\vec{e}_j \cdot \vec{e}_k\rangle \\ \mu_{kk}(d\lambda) &= \langle Q(d\lambda)\vec{e}_k \cdot \vec{e}_k\rangle_1 = \langle Q(d\lambda)\vec{e}_k \cdot Q(d\lambda)\vec{e}_k\rangle_1 = \|Q(d\lambda)\vec{e}_k\|_1^2, \end{aligned} \tag{49}$$

where we have used a Fubini theorem [17] and $\|\cdot\|_1$ denotes the norm induced by the inner-product $\langle\cdot,\cdot\rangle_1$. From equation (49) we see, generically, that the diagonal components μ_{kk}, $k = 1, \ldots, d$, of $\boldsymbol{\mu}$ are positive measures, while the off-diagonal components μ_{jk}, $j \ne k = 1, \ldots, d$, may be signed measures [17, 36]. Generalizing equation (29), with $R = \prod_{j=1}^d R_j$, to dimensions $d \ge 3$ shows that μ^0_{jk} is a linear combination of averages of the form $\langle \prod_i \cos^{n_i}\theta_i \sin^{m_i}\theta_i\rangle$, where $n_i, m_i = 0, 1, 2, \ldots$.

A principal application of the ACM is to derive *forward bounds* on the diagonal components σ^*_{kk} of the tensor $\boldsymbol{\sigma}^*$, $k = 1, \ldots, d$, given partial information on the microgeometry [6, 26, 19, 7]. This information may be given in terms of the moments μ^n_{kk}, $n = 0, 1, 2, \ldots$, of the measure μ_{kk} [28, 19]. Provided this information, the bounds on σ^*_{kk} follow from the special structure of $F_{kk}(s)$ in (43). More specifically, it is a *linear* functional of the *positive* measure μ_{kk}. The bounds are obtained by fixing the contrast parameter s, varying over an admissible set of measures μ_{kk} (or geometries) which is determined by the known information regarding the polycrystalline medium. Knowledge of the moments μ^n_{kk} for $n = 1, \ldots, J$ confines σ^*_{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 [28, 18]. When all the moments are known the measure μ_{kk} is uniquely determined [1], hence σ^*_{kk} is explicitly known. The bounding procedure [6, 26, 19, 7] is reviewed and extended to the polycrystalline setting in Section 3.

2.2. **Lattice Setting.**

2.2.1. *Infinite Lattice Setting.*

2.2.2. *Finite Lattice Setting.*

3. Bounding Procedure

An important property of the integral representation for $F_{jk}(s)$, $j,k = 1,\ldots,d$, displayed in equation (43), is that parameter information in s and E_0 is *separated* from the geometry of the composite, which is encoded in the spectral measure μ_{jk} via its moments μ^n_{jk}, $n = 0,1,2,\ldots$ [10, 19]. Another important property of the representation for $F_{jk}(s)$ is that it is a *linear* functional of the measure μ_{jk}. Moreover, the diagonal components μ_{kk} are *positive* measures. These properties are also shared by the function $E_{jk}(s)$ given in equation (43). These important properties may be exploited to obtain rigorous bounds for the diagonal components of the effective parameters [5, 6, 26, 19, 7]. In this section we review a bounding procedure which is presented in [19, 18]. The bounds incorporate the moments μ^n_{kk} and η^n_{kk}, $n = 0,1,2,\ldots$, of the measures μ_{kk} and η_{kk} associated with the functions $F_{kk}(s)$ and $E_{kk}(s)$, respectively.

In this section, we will discuss the bounding procedure in terms of the diagonal components σ^*_{kk}, $k = 1,\ldots,d$, of the effective complex conductivity tensor $\boldsymbol{\sigma}^*$. 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 $F(s)$ and $E(s)$ in equation (43) and initially focus on the function $F(s)$ and the measure μ, introducing the function $E(s)$ and the measure η when appropriate.

Bounds on σ^* are obtained as follows. The support of the measure μ is contained in the interval $[0,1]$ and its mass is given by $\mu^0 = \langle |X_1\vec{e}_k|^2\rangle$, where $0 \le \mu^0 \le 1$. Consider the set $\mathscr{M}$ of positive Borel measures on $[0,1]$ with mass ≤ 1. By equation (43), for fixed $s \in \mathbb{C}\backslash[0,1]$, $F(s)$ is a linear functional of the measure μ, $F : \mathscr{M} \mapsto \mathbb{C}$, and we write $F(s) = F(s,\mu)$ and $m(h) = m(h,\mu)$. Suppose that we know the moments μ^n of the measure μ for $n = 0,\ldots,J$. Define the set $\mathscr{M}^\mu_J \subset \mathscr{M}$ by

$$\mathscr{M}^\mu_J = \left\{ \nu \in \mathscr{M} \;\middle|\; \int_0^1 \lambda^n \nu(d\lambda) = \mu^n,\ n = 0,\ldots,J \right\}. \tag{50}$$

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

$$A^\mu_J = \left\{\ m(h,\mu) \in \mathbb{C} \mid\ h \notin (-\infty,0],\ \mu \in \mathscr{M}^\mu_J \right\}. \tag{51}$$

The set of measures $\mathscr{M}^\mu_J$ is a compact, convex subset of $\mathscr{M}$ with the topology of weak convergence [19]. Since the mapping $F(s,\mu)$ in (43) is linear in μ, it follows that A^μ_J is a compact convex subset of the complex plane $\mathbb{C}$. The extreme points of $\mathscr{M}^\mu_0$ are the one point measures $a\delta_b$, $0 \le a,b \le 1$ [13], while the extreme points of $\mathscr{M}^\mu_J$ for $J > 0$ are weak limits of convex combinations of measures of the form [23, 19]

$$\mu_J(d\lambda) = \sum_{i=1}^{J+1} a_i \delta_{b_i}(d\lambda), \quad a_i \ge 0, \quad 0 \le b_1 < \cdots < b_{J+1} < 1, \quad \sum_{i=1}^{J+1} a_i b_i^n = \mu^n, \tag{52}$$

for $n = 0,1,\ldots,J$. (WHAT CAN WE SAY ABOUT UNIAXIAL POLYCRYSTALLINE MEDIA WHICH IS ANALOGOUS TO THE FOLLOWING PROPERTIES OF TWO-COMPONENT COMPOSITE MEDIA? For the case of two-component random media in the continuous setting, every measure $\mu \in \mathscr{M}^\mu_J$ gives rise to a function $m(h,\mu)$ that is the effective (relative) conductivity of a multi-rank laminate [29]. However, in general [19], not every measure $\mu \in \mathscr{M}^\mu_J$ gives rise to such a function $m(h,\mu)$. Therefore, the set A^μ_J will only *contain* the exact range of values of the effective conductivity [19]. This is sufficient for the bounding procedure discussed in this section.

By the symmetries between the formulas in equation (43) and $X_1 + X_2 = I$ in (31), the support of the measure η is contained in the interval $[0,1]$ and its mass is given by $\eta^0 = \langle |X_2\,\vec{e}_k|^2\rangle = \langle X_2\,\vec{e}_k \cdot \vec{e}_k\rangle = 1 - \mu^0$, which implies that $0 \le \eta^0 \le 1$. We can therefore define compact, convex sets $\mathscr{M}_J^\eta \subset \mathscr{M}$ and $A_J^\eta \subset \mathbb{C}$ which are analogous to those defined in equations (50) and (51), respectively, involving the function $\tilde{m}(h,\eta) = 1 - E(s,\eta)$. Moreover, the extreme points of $\mathscr{M}_0^\eta$ are the one point measures $c\delta_d$, $0 \le c, d \le 1$ while the extreme points of $\mathscr{M}_J^\eta$ are weak limits of convex combinations of measures of the form given in equation (52).

Consequently, in order to determine the extreme points of the sets A_J^μ and A_J^η 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

$$F(s,\mu_J) = \sum_{i=1}^{J+1} \frac{a_i}{s - b_i}\,, \qquad E(s,\eta_J) = \sum_{i=1}^{J+1} \frac{c_i}{s - d_i}\,, \tag{53}$$

as the a_i, b_i, c_i, and d_i vary under the constraints given in equation (52). While $F(s,\mu_J)$ and $E(s,\eta_J)$ in (53) may not run over all points in A_J^μ and A_J^η as these parameters 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 σ^* is given by $\sigma^* = \sigma_2 m(h,\mu) = \sigma_1/\tilde{m}(h,\eta)$, the regions A_J^μ and A_J^η have to be mapped to the common σ^*-plane to provide bounds for σ^*.

In this section we discuss two different bounds for σ^*. The first bound assumes that only the masses μ^0 and η^0 of the measures μ and η 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, as described by the following lemma.

Lemma 3.1. *If the orientations of the crystallites are statistically isotropic then*

$$\mu^1 = \frac{d-1}{d^3}\,, \qquad \eta^1 = \frac{p_1 p_2 (d-1)}{d}\,. \tag{54}$$

Consider the first case, where $J = 0$ in (53) 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

$$C_0(\lambda) = \frac{\mu^0}{s-\lambda}\,, \quad -\infty \le \lambda \le \infty, \qquad \tilde{C}_0(\tilde{\lambda}) = \frac{\eta^0}{s-\tilde{\lambda}}\,, \quad -\infty \le \tilde{\lambda} \le \infty. \tag{55}$$

In the σ^*-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 (55), and the values of σ^* lie inside this region [18]. These bounds are optimal [27, 7], 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 σ_1 and σ_2 are real and positive, the bounding region collapses to the interval

$$\left(\frac{\mu^0}{\sigma_1} + \frac{\eta^0}{\sigma_2}\right)^{-1} \le \sigma^* \le \mu^0\sigma_1 + \eta^0\sigma_2, \tag{56}$$

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 [37].

Now consider the second case where $J = 1$ in (53), the volume fraction $p_1 = 1 - p_2$ is fixed, and the random medium is statistically isotropic so that the first moments μ^1 and η^1 of the measures μ and η are given, respectively, by that in equation (54). A convenient way of including this information is to use the transformations [7]

$$F_1(s) = \frac{1}{p_1} - \frac{1}{sF(s)}\,, \qquad E_1(s) = \frac{1}{p_2} - \frac{1}{sE(s)}\,. \tag{57}$$

Due to the symmetries between $F_1(s)$ and $E_1(s)$ in (57) 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 [7, 18] analogous to that in equation (43), involving a measure μ_1, say, which is supported in the interval $[0, 1]$. Since only the mass μ^0 and the first moment $\mu^1 = (d-1)/d^3$ of the measure μ are known, the transformation (57) determines only the mass $\mu_1^0 = (d-1)/d$ of the measure μ_1 [7, 18]. This reveals the utility of the transformation $F_1(s)$ in (57), 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 $p_2/(p_1 d(s-\lambda))$, $-\infty \le \lambda \le \infty$. Similarly, the values of $E_1(s)$ lie inside a circle $p_1(d-1)/(p_2 d(s-\tilde{\lambda}))$, $-\infty \le \tilde{\lambda} \le \infty$. 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 [18]

$$C_1(\lambda) = \frac{s-\lambda}{sd(s-\lambda-(d-1)/d^2)}, \quad \tilde{C}_1(\tilde{\lambda}) = \frac{(d-1)(s-\tilde{\lambda})}{sd(s-\tilde{\lambda}-(d-1)/d^2)}, \qquad -\infty \le \lambda, \tilde{\lambda} \le \infty. \tag{58}$$

In the σ^*-plane the intersection of these two circular regions is bounded by two circular arcs [18] corresponding to $0 \le \lambda \le (d-1)/d$ and $0 \le \tilde{\lambda} \le 1/d$ in (58).

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 [18]. While there are at least five points on the arc $C_1(\lambda)$ in (58) that are attainable by composite microstructures [27], the arc $\tilde{C}_1(\tilde{\lambda})$ in (58) violates [18] the interchange inequality $m(h)m(1/h) \ge 1$ [24, 38], which becomes an equality in two dimensions [29]. Consequently the isotropic bounds in (58) are not optimal, but have been improved [26, 7] by incorporating the interchange inequality. When σ_1 and σ_2 are real and positive with $\sigma_1 \le \sigma_2$, the region collapses to the interval

$$\sigma_1 - \frac{\sigma_1}{sd-(d-1)/d}. \le \sigma^* \le \frac{\sigma_1}{1-((d-1)/(sd-(d-1)/d))} \tag{59}$$

which are the Hashin–Shtrikman bounds.

The higher moments μ^n, $n \ge 2$ depend on $(n+1)$-point correlation functions [19] and cannot be calculated in general, although the interchange inequality forces relations among them [28]. If the moments $\mu^0, \dots, \mu^J$ are known then the transformation F_1 in (57) can be iterated to produce an upper half plane function F_J with a integral representation, involving a positive measure μ_J which is supported on the interval $[0, 1]$. As in the case where $J = 1$, the first J moments of the measure μ determine only the mass μ_J^0 of the measure μ_J [18], 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 $\eta^0, \dots, \eta^J$. The resulting bounds form a nested sequence of lens-shaped regions [18].

4. Numerical Results

In Section 2.2.2 we extended the ACM for representing transport in composites to the finite lattice setting. Here, we demonstrate how this mathematical framework can be utilized to compute spectral measures and the associated effective parameters for such two-phase random media. In particular, in the finite lattice setting, the operators Γ, Υ, and χ_i, $i = 1, 2$, are represented as real-symmetric matrices and the spectral measures of the associated random matrices $M_i = \chi_i \Gamma \chi_i$ and $K_i = \chi_i \Upsilon \chi_i$ are explicitly determined by their eigenvalues and eigenvectors, as displayed in equation (**??**). In Section 2.2.2 we also introduced a projection method, summarized by equations (**??**) and (**??**), which provides a numerically efficient way to accomplish these computations. Furthermore, in the paragraph following the statement of Theorem **??**, we introduced three classes of locally isotropic, statistically isotropic, and anisotropic random media. In this section we employ the projection method to directly calculate the spectral measures and effective parameters for such composite media.

Acknowledgements. We gratefully acknowledge support from the Division of Mathematical Sciences and the Office of Polar Programs at the U.S. National Science Foundation (NSF) through Grants DMS-1009704, ARC-0934721, and DMS-0940249. We are also grateful for support from the Office of Naval Research (ONR) through Grants N00014-13-10291 and N00014-12-10861. Finally, we would like to thank the NSF Math Climate Research Network (MCRN) for their support of this work.

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