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 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 periodic flows. Our novel approach yields integral representations for the symmetric and anti-symmetric parts of the effective diffusivity. These representations hold for dynamic and steady incompressible flows, and involve the spectral measure of a self-adjoint or normal linear operator. In the case of a steady fluid velocity field, 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 utilize the integral representations to obtain asymptotic behavior of the effective diffusivity as the molecular diffusivity tends to zero, for model 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 [29] with an effective diffusivity tensor $\mathcal{K}^*$. 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 [19]. 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 random, time-independent fluid velocity fields was treated in [16]. This reduced the analysis of turbulent diffusion to solving a diffusion equation involving a homogenized temperature and a (constant) effective diffusivity tensor $\mathcal{K}^*$. An important consequence of this analysis is that $\mathcal{K}^*$ is given in terms of a *curl-free* stationary stochastic process which satisfies a steady state diffusion equation, involving a skew-symmetric random matrix $\mathbf{H}$ [2, 1]. By adapting the analytic continuation method (ACM) of homogenization theory for composites [13], it was shown that the result in [16] leads to an integral representation for the symmetric part $\boldsymbol{\kappa}^*$ of $\mathcal{K}^*$, involving a spectral measure of a self-adjoint random operator [2, 1]. This integral representation of $\boldsymbol{\kappa}^*$ was generalized to the time-dependent case in [3, 7]. Remarkably, this method has also been extended to flows with incompressible *nonzero* effective drift [23], flows where particles diffuse according to linear collisions [24], and solute transport in porous media [6]. All these approaches yield integral representations of the symmetric and, when appropriate, the anti-symmetric part $\boldsymbol{\alpha}^*$ of $\mathcal{K}^*$.

Homogenization of the advection-diffusion equation for periodic or cellular, incompressible flow fields was treated in [8, 9]. As in the case of random flows, the effective diffusivity tensor $\mathcal{K}^*$ 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 integral representations for both the symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of $\mathcal{K}^*$, for both cases of steady and time-dependent flows. These integral representations involve a self-adjoint or normal linear 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 rectangle. 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 utilize the analytic structure of the integral representation for $\boldsymbol{\mathcal{K}}^*$ 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. To make this manuscript more self contained, we have include an appendix in Section A-1 which contains many of the technical details underlying this work.

2. Mathematical Methods

In this section, we formulate the effective parameter problem for enhanced diffusive transport by advective, periodic flows. We provide integral representations for the symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$, which hold for both steady and dynamic flows. The effective parameter problem [16, 8, 7] for such transport processes is reviewed in Section 2.1. Parallels existing between this problem of homogenization theory [4] and the ACM for representing transport in composites [13], are put into correspondence in Section 2.2. In particular, an abstract Hilbert space framework is provided in Section 2.2.1 which places these different effective parameter problems on common mathematical footing. Within this Hilbert space setting, we derive in Section 2.2.2 integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$, involving the molecular diffusivity ε and a *spectral measure* of a self-adjoint or normal linear operator. These integral representations are employed in Section 3 to obtain the asymptotic behavior of the components of $\boldsymbol{\mathcal{K}}^*$ in the scaling regime, where $\varepsilon \ll 1$.

2.1. **Effective transport by advective-diffusion.** Consider the advection enhanced diffusive transport of a passive tracer $\phi(t,\vec{x}\,)$, $t>0$, $\vec{x}\in\mathbb{R}^d$, as described by the advection-diffusion equation

$$\partial_t\phi = \kappa_0\Delta\phi + \vec{\nabla}\cdot(\vec{v}\phi), \quad \phi(0,\vec{x}) = \phi_0(\vec{x}), \tag{1}$$

with initial density $\phi_0(\vec{x})$ given. Here, ∂_t denotes partial differentiation with respect to time t, $\Delta = \vec{\nabla}\cdot\vec{\nabla} = \nabla^2$ is the Laplacian, and $\kappa_0 > 0$ is the molecular diffusivity. The fluid velocity field $\vec{v} = \vec{v}(t,\vec{x})$ in (1) is assumed to be incompressible and mean-zero

$$\vec{\nabla}\cdot\vec{v} = 0, \quad \langle\vec{v}\,\rangle = 0, \tag{2}$$

where we denote by 0 the null element of all linear spaces in question. The bounded sets $\mathcal{T}\subset\mathbb{R}$ and $\mathcal{V}\subset\mathbb{R}^d$, with $t\in\mathcal{T}$ and $\vec{x}\in\mathcal{V}$, define the space-time period cell ($(d+1)$–torus) $\mathcal{T}\otimes\mathcal{V}$. In equation (2), we denote by $\langle\cdot\rangle$ spatial averaging over $\mathcal{V}$ in the case of a time-independent velocity field, $\vec{v} = \vec{v}(\vec{x})$, and when the velocity field is time-dependent, $\vec{v} = \vec{v}(t,\vec{x})$, $\langle\cdot\rangle$ denotes space-time averaging over $\mathcal{T}\otimes\mathcal{V}$.

We non-dimensionalize equation (1) as follows. Let ℓ and $\tilde{t}$ be typical length and time scales associated with the problem of interest. Mapping to the non-dimensional variables $t\mapsto t/\tilde{t}$ and $x_j\mapsto x_j/\ell$, one finds that ϕ satisfies the advection-diffusion equation in (1) with a non-dimensional molecular diffusivity $\varepsilon = \tilde{t}\kappa_0/\ell^2$ and velocity field $\vec{u} = \tilde{t}\,\vec{v}/\ell$, where x_j is the j^{th} component of the vector $\vec{x}$.

For d-dimensional, mean-zero, incompressible flows $\vec{u}$, there is a (non-dimensional) skew-symmetric matrix $\mathbf{H}(t,\vec{x})$ such that (see Section A-1.1 for details)

$$\vec{u} = \vec{\nabla}\cdot\mathbf{H}, \qquad \mathbf{H}^T = -\mathbf{H}. \tag{3}$$

Using this representation of the velocity field $\vec{u}$, which also satisfies (2), equation (1) can be written as a diffusion equation,

$$\partial_t \phi = \vec{\nabla} \cdot \boldsymbol{\kappa} \vec{\nabla} \phi, \quad \phi(0, \vec{x}) = \phi_0(\vec{x}), \qquad \boldsymbol{\kappa} = \varepsilon \mathbf{I} + \mathbf{H}, \tag{4}$$

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

$$\kappa_{jk} = \varepsilon \delta_{jk} + H_{jk}, \quad j, k = 1, \cdots, d. \tag{5}$$

Here, δ_{jk} is the Kronecker delta and we denote by $\mathbf{I}$ the identity operator on all linear spaces in question.

We are interested in the dynamics of ϕ in (4) for *large* length and time scales, and when the initial density ϕ_0 is slowly varying relative to the velocity field $\vec{u}$. Anticipating that ϕ will have diffusive dynamics, we re-scale space and time by $\vec{x} \mapsto \vec{x}/\delta$ and $t \mapsto t/\delta^2$, respectively. For periodic diffusivity coefficients in (4) which are uniformly elliptic but not necessarily symmetric, it can be shown [8] that, as $\delta \to 0$, the associated solution $\phi^\delta(t, \vec{x})$ of (4) converges to $\bar{\phi}(t, \vec{x})$, which satisfies the following diffusion equation involving a (constant) effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$ (see Section A-1.2 for details)

$$\partial_t \bar{\phi} = \vec{\nabla} \cdot \boldsymbol{\mathcal{K}}^* \vec{\nabla} \bar{\phi}, \quad \bar{\phi}(0, \vec{x}) = \phi_0(\vec{x}). \tag{6}$$

The components $\mathcal{K}^*_{jk} = \boldsymbol{\mathcal{K}}^* \vec{e}_j \cdot \vec{e}_k$ of the tensor $\boldsymbol{\mathcal{K}}^*$ are obtained by solving the cell problem [8]

$$\partial_t \chi_k = \vec{\nabla} \cdot \boldsymbol{\kappa} (\vec{\nabla} \chi_k + \vec{e}_k\,), \quad \langle \vec{\nabla} \chi_k \rangle = 0, \tag{7}$$

for each standard basis vector $\vec{e}_k$, $k = 1, \ldots, d$, where $\chi_k = \chi_k(t, \vec{x}\,; \vec{e}_k)$. Equation (7) also holds [8] when the velocity field is time-independent $\vec{u} = \vec{u}(\vec{x})$, however, in this case χ_k is time-independent and $\partial_t \chi_k = 0$. The symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of $\boldsymbol{\mathcal{K}}^*$ are defined by

$$\boldsymbol{\mathcal{K}}^* = \boldsymbol{\kappa}^* + \boldsymbol{\alpha}^*, \qquad \boldsymbol{\kappa}^* = \frac{1}{2}(\boldsymbol{\mathcal{K}}^* + [\boldsymbol{\mathcal{K}}^*]^T), \quad \boldsymbol{\alpha}^* = \frac{1}{2}(\boldsymbol{\mathcal{K}}^* - [\boldsymbol{\mathcal{K}}^*]^T). \tag{8}$$

The components κ^*_{jk} and α^*_{jk}, $j, k = 1, \ldots, d$, of $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ are given by (see Section A-1.2)

$$\kappa^*_{jk} = \varepsilon(\delta_{jk} + \langle \vec{\nabla} \chi_j \cdot \vec{\nabla} \chi_k \rangle), \qquad \alpha^*_{jk} = \langle \mathbf{S} \vec{\nabla} \chi_j \cdot \vec{\nabla} \chi_k \rangle, \qquad \mathbf{S} = \mathbf{H} - (\boldsymbol{\Delta}^{-1}) \mathbf{T}, \quad \mathbf{T} = \partial_t \mathbf{I}. \tag{9}$$

Here, $\mathbf{T} = \text{diag}(\partial_t, \ldots, \partial_t)$ operates component-wise on vector fields, $\boldsymbol{\Delta}^{-1} = \text{diag}(\Delta^{-1}, \ldots, \Delta^{-1})$ is the inverse of the vector Laplacian, and the inverse operation Δ^{-1} is based on convolution with the Green's function for the Laplacian Δ [27]. Due to the fact that the vector field $\vec{\nabla} \chi_j$ is *real-valued*, we have that $\langle \vec{\nabla} \chi_j \cdot \vec{\nabla} \chi_k \rangle = \langle \vec{\nabla} \chi_k \cdot \vec{\nabla} \chi_j \rangle$. From equation (9) this clearly implies that the tensor $\boldsymbol{\kappa}^*$ is symmetric, $\kappa^*_{jk} = \kappa^*_{kj}$. Moreover, equation (9) demonstrates that the effective transport of the tracer ϕ in the principle directions $\vec{e}_k$, $k = 1, \ldots, d$, is always *enhanced* by the presence of an incompressible velocity field, $\mathcal{K}^*_{kk} = \kappa^*_{kk} \geq \varepsilon$. Similarly, the skew-symmetry of the operator $\mathbf{S} : \mathbb{R}^d \mapsto \mathbb{R}^d$ (see Section A-1.3) implies that the tensor $\boldsymbol{\alpha}^*$ is also skew-symmetric, $\alpha^*_{jk} = \langle \mathbf{S} \vec{\nabla} \chi_j \cdot \vec{\nabla} \chi_k \rangle = -\langle \mathbf{S} \vec{\nabla} \chi_k \cdot \vec{\nabla} \chi_j \rangle = -\alpha^*_{kj}$. In particular,

$$\alpha^*_{kk} = \langle \mathbf{S} \vec{\nabla} \chi_k \cdot \vec{\nabla} \chi_k \rangle = -\langle \mathbf{S} \vec{\nabla} \chi_k \cdot \vec{\nabla} \chi_k \rangle = 0. \tag{10}$$

In Section 2.2 we discuss the properties of the operator $\mathbf{S}$ and the vector field $\vec{\nabla} \chi_j$ in more detail, and recast equations (7) and (9) into a form which parallels the ACM for characterizing effective transport in composite media [13]. In particular, we provide an abstract Hilbert space formulation of the effective parameter problem in Section 2.2.1, which yields a resolvent representation of the vector field $\vec{\nabla} \chi_j$ involving an anti-symmetric (normal) integro-differential operator $\mathbf{A}$ which is closely related to $\mathbf{S}$, where we use the terms skew-symmetric and anti-symmetric interchangeably. In Section 2.2.2, we employ this mathematical framework to provide integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ in terms of a *spectral measure* associated with $\mathbf{A}$.

2.2. **The ACM for advection enhanced diffusion by periodic flows.** The ACM for representing transport in composites gives a Hilbert space formulation of the effective parameter problem and provides an integral representation for the effective transport coefficients of composite media, involving a *spectral measure* of a self-adjoint operator which depends only on the composite geometry [13, 20, 18]. Here we establish a correspondence between this effective parameter problem and that for enhanced diffusive transport by advective velocity fields. In Section 2.2.1, we formulate the Hilbert space framework associated with advective diffusion, and employ it to obtain a resolvent representation of the vector field $\vec{\nabla}\chi_k$ in (7). In Section 2.2.2 we utilize this mathematical framework to obtain integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ involving a spectral measure which depends only on the fluid velocity field $\vec{u}$.

Toward this goal, we rewrite the first formula in equation (7) in a more suggestive, divergence form. Using the notation $\vec{\nabla}\Delta^{-1}\partial_t = \boldsymbol{\Delta}^{-1}\mathbf{T}\vec{\nabla}$ (see Section A-1.3), where $\mathbf{T} = \partial_t\mathbf{I}$, we rewrite [8] $\partial_t\chi_k = \Delta\Delta^{-1}\partial_t\chi_k = \vec{\nabla}\cdot(\boldsymbol{\Delta}^{-1}\mathbf{T})\vec{\nabla}\chi_k$ and we define the vector field $\vec{E}_k = \vec{\nabla}\chi_k + \vec{e}_k$ and the operator $\boldsymbol{\sigma} = \boldsymbol{\kappa} - (\boldsymbol{\Delta}^{-1})\mathbf{T} = \varepsilon\mathbf{I} + \mathbf{S}$. In the case of steady fluid velocity fields, we have $\boldsymbol{\sigma} = \boldsymbol{\kappa} = \varepsilon\mathbf{I} + \mathbf{H}$. With these definitions, equation (7) may be written as $\vec{\nabla}\cdot\boldsymbol{\sigma}\vec{E}_k = 0$, $\langle\vec{E}_k\rangle = \vec{e}_k$, which is equivalent to

$$\vec{\nabla}\cdot\vec{J}_k = 0, \quad \vec{\nabla}\times\vec{E}_k = 0, \quad \vec{J}_k = \boldsymbol{\sigma}\vec{E}_k, \quad \langle\vec{E}_k\rangle = \vec{e}_k, \qquad \boldsymbol{\sigma} = \varepsilon\mathbf{I} + \mathbf{S} \tag{11}$$

The formulas in (11) are precisely the electrostatic version of Maxwell's equations for a conductive medium [13], 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 composites, the effective conductivity tensor $\boldsymbol{\sigma}^*$ is defined as

$$\langle\vec{J}_k\rangle = \boldsymbol{\sigma}^*\langle\vec{E}_k\rangle. \tag{12}$$

The linear constitutive relation $\vec{J}_k = \boldsymbol{\sigma}\vec{E}_k$ in (11) relates the local intensity and flux, while that in (12) relates the mean intensity and flux. Due to the skew-symmetry of $\mathbf{S}$, the intensity-flux relationship in (11) is similar to that of a Hall medium [14].

For the (constant) tensors $\boldsymbol{\mathcal{K}}^*$ and $\boldsymbol{\sigma}^*$ to be meaningfull, the averages which define these effective quantities in (9) and (12) must be well defined and finite. For example, in order for the diagonal components $\mathcal{K}^*_{kk}$, $k = 1, \ldots, d$, of the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$ to be well defined and finite, we must have that the vector field $\vec{\nabla}\chi_k$ is square integrable on $\mathcal{T}\times\mathcal{V}$ with respect to the Lebesgue measure. Moreover, for the components α^*_{jk}, $j \neq k = 1, \ldots, d$, of $\boldsymbol{\alpha}^*$ to be well defined and finite, we must also have that the integro-differential operator $\mathbf{S}$ is bounded in some sense so that $\mathbf{S}\vec{\nabla}\chi_j$ is Lebesgue measurable and $\mathbf{S}\vec{\nabla}\chi_j\cdot\vec{\nabla}\chi_k$ is Lebesgue integrable on $\mathcal{T}\times\mathcal{V}$. In other words, we must define the vector field $\vec{\nabla}\chi_j$ as a member of a suitable space of functions so that the components of the tensors $\boldsymbol{\mathcal{K}}^*$ and $\boldsymbol{\sigma}^*$ are well defined and have finite values.

In Section 2.2.1 we discuss these important details at length and prove the following theorem, which states that the definitions of the effective parameters in (12) and (9) are well defined and finite, and are equivalent up to transposition. This places these different effective parameter problems on common mathematical footing, for both cases of time-independent and time-dependent velocity fields $\vec{u}$, and follows by adapting the Hilbert space formulation of the ACM to treat the effective transport properties of advective diffusion.

Theorem 2.1. *Let the effective tensors $\boldsymbol{\mathcal{K}}^*$ and $\boldsymbol{\sigma}^*$ be defined as in equations (7)–(9) and equations (11) and (12), respectively. Then there exists a function space $\mathscr{H}$ on which $\boldsymbol{\sigma} = \varepsilon\mathbf{I}+\mathbf{S}$ is a bounded linear operator, and for $\vec{\nabla}\chi_j \in \mathscr{H}$, $j = 1, \ldots, d$, we have*

$$\boldsymbol{\sigma}^* = [\boldsymbol{\mathcal{K}}^*]^T. \tag{13}$$

In particular, the symmetric part $\boldsymbol{\kappa}^*$ of $\boldsymbol{\mathcal{K}}^*$ is equal to that of $\boldsymbol{\sigma}^*$ and the anti-symmetric part $\boldsymbol{\alpha}^*$ of $\boldsymbol{\mathcal{K}}^*$ is equal to the negative of that of $\boldsymbol{\sigma}^*$.

2.2.1. *Hilbert space and resolvent representation.* In this section we explore the mathematical properties of the skew-symmetric operator $\mathbf{S}$ introduced in equation (9), and provide an abstract Hilbert space formulation of the effective parameter problem for advective diffusion. We utilize this mathematical framework and equation (7) to obtain a resolvent representation of the vector field $\vec{\nabla}\chi_k$, involving an anti-symmetric operator $\mathbf{A}$ which is closely related to $\mathbf{S}$. Using the results of this section, we derive in Section 2.2.2 integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$, involving a *spectral measure* associated with $\mathbf{A}$.

Consider the Hilbert spaces $L^2_d(\mathcal{T}) = \otimes_{i=1}^d L^2(\mathcal{T})$ and $L^2_d(\mathcal{V}) = \otimes_{i=1}^d L^2(\mathcal{V})$ (over the complex field $\mathbb{C}$) of Lebesgue measurable, square integrable vector valued functions. Now consider the associated Hilbert spaces $\mathscr{H}_\mathcal{T} \subset L^2_d(\mathcal{T})$ and $\mathscr{H}_\mathcal{V} \subset L^2_d(\mathcal{V})$ of periodic vector valued functions with temporal periodicity T on the interval $\mathcal{T} = (0, T)$ and spatial periodicities V_j, $j = 1, \ldots, d$, on the d-dimensional region $\mathcal{V} = (0, V_1) \times \cdots \times (0, V_d)$, respectively, as well as their direct product $\mathscr{H}_{\mathcal{T}\mathcal{V}} = \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\mathcal{V}$,

$$\mathscr{H}_{\mathcal{T}\mathcal{V}} = \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\mathcal{V}, \quad \mathscr{H}_\mathcal{T} = \{\vec{\xi} \in L^2_d(\mathcal{T}) \mid \vec{\xi}(0) = \vec{\xi}(T)\}, \quad \mathscr{H}_\mathcal{V} = \{\vec{\xi} \in L^2_d(\mathcal{V}) \mid \vec{\xi}(0) = \vec{\xi}(\vec{V})\}, \tag{14}$$

where we have defined $\vec{V} = (V_1, \ldots, V_d)$. Denote by $\langle \cdot, \cdot \rangle$ the sesquilinear inner-product associated with the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}$, which is defined by $\langle \vec{\xi}, \vec{\zeta} \rangle = \langle \vec{\xi} \cdot \vec{\zeta} \rangle$ and $\langle \vec{\xi}, \vec{\zeta} \rangle = \overline{\langle \vec{\zeta}, \vec{\xi} \rangle}$, where $\bar{a}$ is the complex conjugate of $a \in \mathbb{C}$. By the Helmholtz theorem [15, 5], the Hilbert space $\mathscr{H}_\mathcal{V}$ in (14) can be decomposed into mutually orthogonal subspaces of curl-free $\mathscr{H}_\times$, divergence-free $\mathscr{H}_\bullet$, and constant $\mathscr{H}_0$ vector fields, with associated orthogonal projectors $\boldsymbol{\Gamma}_\times$, $\boldsymbol{\Gamma}_\bullet$, and $\boldsymbol{\Gamma}_0$, respectively, [8, 18]

$$\begin{gathered} \mathscr{H}_\mathcal{V} = \mathscr{H}_\times \oplus \mathscr{H}_\bullet \oplus \mathscr{H}_0, \qquad \mathbf{I} = \boldsymbol{\Gamma}_\times + \boldsymbol{\Gamma}_\bullet + \boldsymbol{\Gamma}_0, \\ \boldsymbol{\Gamma}_\times = \vec{\nabla}(\Delta^{-1})\vec{\nabla}\cdot, \quad \boldsymbol{\Gamma}_\bullet = -\vec{\nabla} \times (\boldsymbol{\Delta}^{-1})\vec{\nabla}\times, \quad \boldsymbol{\Gamma}_0 = \langle \cdot \rangle, \\ \mathscr{H}_\times = \{\vec{\xi} \mid \vec{\nabla} \times \vec{\xi} = 0 \text{ weakly}\}, \quad \mathscr{H}_\bullet = \{\vec{\xi} \mid \vec{\nabla} \cdot \vec{\xi} = 0 \text{ weakly}\}, \quad \mathscr{H}_0 = \{\vec{\xi} \mid \vec{\xi} = \langle \vec{\xi} \rangle\}. \end{gathered} \tag{15}$$

We are primarily concerned with fluid velocity fields $\vec{u}$ such that $0 < \mathcal{K}^*_{kk} < \infty$ for all $0 < \varepsilon < \infty$. Consequently, from equation (9) we have that the (weakly) curl-free vector field $\vec{\nabla}\chi_k$ is bounded in the norm $\| \cdot \|$ induced by the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$-inner-product, $\|\vec{\nabla}\chi_k\| < \infty$, and from equation (7) we have that $\vec{\nabla}\chi_k$ is mean-zero, so that $\vec{\nabla}\chi_k \in \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\times \subset \mathscr{H}_{\mathcal{T}\mathcal{V}}$. Defining the (weakly) divergence-free vector field $\vec{J}_k = \boldsymbol{\sigma}\vec{E}_k$ in (11) as a member of a subset of $\mathscr{H}_{\mathcal{T}\mathcal{V}}$ is technically difficult, due to the *unboundedness* of the operator $\boldsymbol{\sigma} = \boldsymbol{\kappa} - (\boldsymbol{\Delta}^{-1})\mathbf{T}$ on this space. We now explore the properties of this operator in more detail.

Since $\mathcal{V}$ is a bounded domain, (Δ^{-1}) is a compact operator [27] on the Hilbert space $L^2(\mathcal{V})$. Hence $(\boldsymbol{\Delta}^{-1})$ is a compact operator on the Hilbert space $\mathscr{H}_\mathcal{V}$, and is consequently bounded in the operator norm $\| \cdot \|$ induced by the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$-inner-product [25, 28, 27], when considered as an operator on $\mathscr{H}_{\mathcal{T}\mathcal{V}}$. We have already assumed for the convergence $\phi^\delta \to \bar{\phi}$, as $\delta \to 0$, that the flow matrix $\mathbf{H}(t, \vec{x})$ is periodic on $\mathcal{T} \otimes \mathcal{V}$. We will also assume that it is (component-wise) mean-zero and bounded in operator norm, and that its component-wise time derivative $\mathbf{TH}$ is also bounded on $\mathscr{H}_{\mathcal{T}\mathcal{V}}$

$$\langle \mathbf{H} \rangle = 0, \quad \|\mathbf{H}\| < \infty, \quad \|\mathbf{TH}\| < \infty. \tag{16}$$

This implies that $\boldsymbol{\kappa} = \varepsilon\mathbf{I} + \mathbf{H}$ is also bounded for all $0 < \varepsilon < \infty$. Consequently, in the case of a time-independent velocity field $\vec{u}$, where $\boldsymbol{\sigma} = \boldsymbol{\kappa}$, the operator $\boldsymbol{\sigma}$ is bounded. This and $\|\vec{\nabla}\chi_k\| < \infty$ implies that $\vec{J}_k \in \mathscr{H}_\bullet$. Therefore, in the case of a time-dependent velocity field, under the assumptions of (16), the unboundedness of $\boldsymbol{\sigma}$ on $\mathscr{H}_{\mathcal{T}\mathcal{V}}$ is due to the unboundedness of $\mathbf{T}$ on $\mathscr{H}_\mathcal{T}$.

The unboundedness of $\mathbf{T}$ on $\mathscr{H}_\mathcal{T}$ can be understood by considering the orthonormal set of functions $\{\vec{\psi}_n\} \subset \mathscr{H}_\mathcal{T}$ with components $(\vec{\psi}_n)_j$, $j = 1, \ldots, d$, defined by

$$(\vec{\psi}_n)_j(t, \vec{x}) = \beta \sin((n + j)\pi t/T), \quad \beta = \sqrt{2/(Td)}, \qquad \langle \vec{\psi}_n \cdot \vec{\psi}_m \rangle = \delta_{nm}, \quad n, m \in \mathbb{N}. \tag{17}$$

The components $(\mathbf{T}\vec{\psi}_n)_j$, $j = 1, \ldots, d$, of the vector $\mathbf{T}\vec{\psi}_n$ and its norm $\|\mathbf{T}\vec{\psi}_n\|$ are given by

$$(\partial_t \vec{\psi}_n)_j(t,\vec{x}) = [\beta(n+j)\pi/T]\cos((n+j)\pi t/T), \quad \|\mathbf{T}\vec{\psi}_n\|^2 = \frac{1}{d}\sum_j [(n+j)\pi/T]^2. \tag{18}$$

Therefore, the norm of the members of the set $\{\mathbf{T}\vec{\psi}_n\}$ grows arbitrarily large as $n \to \infty$. This clearly demonstrates the unboundedness of the operator $\mathbf{T}$ on $\mathscr{H}_\mathcal{T}$.

The above analysis demonstrates that the domain $D(\mathbf{T})$ of the unbounded operator $\mathbf{T}$ is defined only on a proper subset of $\mathscr{H}_\mathcal{T}$, i.e. $D(\mathbf{T}) \subset \mathscr{H}_\mathcal{T}$. However, $D(\mathbf{T})$ can be defined as a *dense* subset of $\mathscr{H}_\mathcal{T}$ such that $\mathbf{T}$ is bounded [25, 28]. Toward this goal, consider the class $\mathscr{A}_\mathcal{T}$ of all functions $\xi \in L^2(\mathcal{T})$ such that $\xi(t)$ is *absolutely continuous* [26] on the interval $\mathcal{T}$ and has a derivative $\xi'(t)$ belonging to $L^2(\mathcal{T})$, i.e. [28, 26]

$$\mathscr{A}_\mathcal{T} = \left\{ \xi \in L^2(\mathcal{T}) \;\middle|\; \xi(t) = c + \int_0^t g(\tau)d\tau, \quad g \in L^2(\mathcal{T}) \right\}, \tag{19}$$

where the constant c and function $g(t)$ are arbitrary. Now, consider the set $\tilde{\mathscr{A}_\mathcal{T}}$ of all functions $\xi \in \mathscr{A}_\mathcal{T}$ that satisfy the periodic initial condition $\xi(0) = \xi(T)$, i.e. functions ξ satisfying the properties of equation (19) with $\int_0^T g(\tau)d\tau = 0$. To illustrate some important ideas later in this work, we also consider the set $\hat{\mathscr{A}_\mathcal{T}}$ of all functions $\xi \in \mathscr{A}_\mathcal{T}$ that satisfy the Dirichlet initial condition $\xi(0) = \xi(T) = 0$, i.e. functions ξ satisfying the properties of equation (19) with $c = 0$ and $\int_0^T g(\tau)d\tau = 0$. More concisely,

$$\tilde{\mathscr{A}_\mathcal{T}} = \{\xi \in \mathscr{A}_\mathcal{T} \,|\, \xi(0) = \xi(T)\}, \quad \hat{\mathscr{A}_\mathcal{T}} = \{\xi \in \mathscr{A}_\mathcal{T} \,|\, \xi(0) = \xi(T) = 0\}. \tag{20}$$

These function spaces satisfy $\hat{\mathscr{A}_\mathcal{T}} \subset \tilde{\mathscr{A}_\mathcal{T}} \subset \mathscr{A}_\mathcal{T}$ and are each everywhere dense in $L^2(\mathcal{T})$ [28].

It follows that $\mathscr{D}_\mathcal{T} = \otimes_{i=1}^d \tilde{\mathscr{A}_\mathcal{T}}$ is everywhere dense in the Hilbert space $\mathscr{H}_\mathcal{T}$. As an operator acting on the function space $\mathscr{D}_\mathcal{T} \otimes \mathscr{H}_\mathcal{V}$, $\boldsymbol{\sigma} = \boldsymbol{\kappa} - (\boldsymbol{\Delta}^{-1})\mathbf{T}$ is bounded. This, $\vec{\nabla}\chi_k \in \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\times$, and equation (7) suggest that we consider the curl-free, mean-zero vector field $\vec{\nabla}\chi_k$ as a member of the function space $\mathscr{H} \subset \mathscr{D}_\mathcal{T} \otimes \mathscr{H}_\mathcal{V}$,

$$\mathscr{H} = \{\vec{\xi} \in \mathscr{D}_\mathcal{T} \otimes \mathscr{H}_\times \,|\, \langle \vec{\xi} \rangle = 0\}, \tag{21}$$

which will be used extensively in the rest of this manuscript. We stress that $\mathscr{H}$ is *not* a Hilbert space, and is instead a dense subset of the Hilbert space $\mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\times$. We will henceforth assume that $\vec{\nabla}\chi_k \in \mathscr{H}$. In the case of a time-independent velocity field $\vec{u}$ we set $\mathscr{D}_\mathcal{T} = \emptyset$ in (21), so that $\vec{\xi} \in \mathscr{H}$ implies $\vec{\xi} \in \mathscr{H}_\times$ with $\langle \vec{\xi} \rangle = 0$. As $\boldsymbol{\sigma}$ is bounded on $\mathscr{H}$ and we have $\vec{\nabla}\chi_k \in \mathscr{H}$, the divergence-free vector field $\vec{J}_k = \boldsymbol{\sigma}\vec{\nabla}\chi_k$ is also bounded $\|\vec{J}_k\| < \infty$, thus $\vec{J}_k \in \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\bullet$. Now we are in a position to prove Theorem 2.1.

Proof of Theorem 2.1. We now employ the Hilbert space framework formulated above to verify that equation (13) holds. This puts the effective parameter problem of enhanced diffusive transport by advective velocity fields and that of transport in composite media on common mathematical footing, for both cases of time-independent and time-dependent velocity fields $\vec{u}$. By the mutual orthogonality of the Hilbert spaces $\mathscr{H}_\times$ and $\mathscr{H}_\bullet$ in (15), $\vec{\nabla}\chi_k \in \mathscr{H}$ and $\vec{J}_k \in \mathscr{H}_\mathcal{T} \otimes \mathscr{H}_\bullet$ imply that $\langle \vec{J}_j \cdot \vec{\nabla}\chi_k \rangle = 0$ for every $j, k = 1, \ldots, d$. This is trivially satisfied in the case of a time-independent velocity field $\vec{u}$, since in this case $\boldsymbol{\sigma} = \boldsymbol{\kappa}$ is bounded so that $\vec{J}_j \in \mathscr{H}_\bullet$ for $\vec{\nabla}\chi_k \in \mathscr{H}$. In either case, as $\vec{E}_k = \vec{\nabla}\chi_k + \vec{e}_k$, we have $\langle \vec{J}_j \cdot \vec{e}_k \rangle = \langle \vec{J}_j \cdot \vec{E}_k \rangle$. Equation (12) then implies that the components $\sigma^*_{jk} = \boldsymbol{\sigma}^* \vec{e}_j \cdot \vec{e}_k = \langle \boldsymbol{\sigma}\vec{E}_j \cdot \vec{e}_k \rangle$ of the effective tensor $\boldsymbol{\sigma}^*$ can be expressed as $\sigma^*_{jk} = \langle \boldsymbol{\sigma}\vec{E}_j \cdot \vec{E}_k \rangle$, with $\boldsymbol{\sigma} = \varepsilon\mathbf{I} + \mathbf{S}$ and $\mathbf{S} = \mathbf{H} - (\boldsymbol{\Delta}^{-1})\mathbf{T}$. Consequently,

$$\sigma^*_{jk} = \varepsilon\langle \vec{E}_j \cdot \vec{E}_k \rangle + \langle \mathbf{S}\vec{E}_j \cdot \vec{E}_k \rangle \tag{22}$$

The property $\langle\vec{\nabla}\chi_k\rangle = 0$ in (7) and equation (9) imply that

$$\varepsilon\langle\vec{E}_j\cdot\vec{E}_k\rangle = \varepsilon[\langle\vec{\nabla}\chi_j\cdot\vec{\nabla}\chi_k\rangle + \langle\vec{\nabla}\chi_j\cdot\vec{e}_k\rangle + \langle\vec{e}_j\cdot\vec{\nabla}\chi_k\rangle + \langle\vec{e}_j\cdot\vec{e}_k\rangle] = \varepsilon(\langle\vec{\nabla}\chi_j\cdot\vec{\nabla}\chi_k\rangle + \delta_{jk}) = \kappa^*_{jk}. \tag{23}$$

From the definition of $\mathbf{S} = \mathbf{H} - (\mathbf{\Delta}^{-1})\mathbf{T}$ in equation (9) we have that $\mathbf{S}\vec{e}_j = \mathbf{H}\vec{e}_j$. Consequently, $\langle\mathbf{S}\vec{e}_j\cdot\vec{e}_k\rangle = \langle\mathbf{H}\vec{e}_j\cdot\vec{e}_k\rangle = 0$, since by equation (16) the matrix $\mathbf{H}$ is (component-wise) mean-zero. Also, by the definition $\vec{u} = \vec{\nabla}\cdot\mathbf{H}$ in (3) and the periodicity of $\mathbf{H}$ and χ_k, we also have $\langle\mathbf{H}\vec{e}_j\cdot\vec{\nabla}\chi_k\rangle = -\langle u_j\chi_k\rangle$ via integration by parts. Therefore, by the skew-symmetry of $\mathbf{S}$ on $\mathscr{H}$, the symmetries $\kappa^*_{kj} = \kappa^*_{jk}$ and $\alpha^*_{kj} = -\alpha^*_{jk}$, and equations (9) and (A-13), we have

$$\begin{aligned}\langle\mathbf{S}\vec{E}_j\cdot\vec{E}_k\rangle &= \langle\mathbf{S}\vec{\nabla}\chi_j\cdot\vec{\nabla}\chi_k\rangle + \langle\mathbf{S}\vec{\nabla}\chi_j\cdot\vec{e}_k\rangle + \langle\mathbf{S}\vec{e}_j\cdot\vec{\nabla}\chi_k\rangle + \langle\mathbf{S}\vec{e}_j\cdot\vec{e}_k\rangle \\ &= \alpha^*_{jk} - \langle\vec{\nabla}\chi_j\cdot\mathbf{H}\vec{e}_k\rangle + \langle\mathbf{H}\vec{e}_j\cdot\vec{\nabla}\chi_k\rangle \\ &= \alpha^*_{jk} + \langle\chi_j u_k\rangle - \langle u_j\chi_k\rangle \\ &= \alpha^*_{jk} + [\alpha^*_{kj} + \kappa^*_{kj} - \varepsilon\delta_{kj}] - [\alpha^*_{jk} + \kappa^*_{jk} - \varepsilon\delta_{jk}] \\ &= -\alpha^*_{jk}.\end{aligned} \tag{24}$$

In summary, from equations (22)–(24) and the symmetries $\kappa^*_{jk} = \kappa^*_{kj}$ and $\alpha^*_{jk} = -\alpha^*_{kj}$ we have that

$$\sigma^*_{jk} = \kappa^*_{jk} - \alpha^*_{jk} = \kappa^*_{kj} + \alpha^*_{kj} = \mathcal{K}^*_{kj}\,, \tag{25}$$

which is equivalent to equation (13). This concludes our proof of Theorem 2.1 □.

We conclude this section with a derivation of the following resolvent representation for $\vec{\nabla}\chi_k$, involving the orthogonal projection operator $\mathbf{\Gamma}_\times = \vec{\nabla}(\Delta^{-1})\vec{\nabla}\cdot$ onto curl-free fields in (15),

$$\vec{\nabla}\chi_j = (\varepsilon\mathbf{I} + \mathbf{A})^{-1}\vec{g}_j = (\varepsilon\mathbf{I} + \mathrm{i}\mathbf{M})^{-1}\vec{g}_j, \quad \mathbf{A} = \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}, \quad \mathbf{M} = -\mathrm{i}\mathbf{A}, \quad \vec{g}_j = -\mathbf{\Gamma}\mathbf{H}\vec{e}_j, \tag{26}$$

where $\mathrm{i} = \sqrt{-1}$ and we have defined $\mathbf{\Gamma} = \mathbf{\Gamma}_\times$ for notational simplicity. Equation (26) follows from applying the integro-differential operator $\vec{\nabla}(\Delta^{-1})$ to $\vec{\nabla}\cdot\boldsymbol{\sigma}\vec{E}_j = 0$ in equation (11), with $\vec{E}_j = \vec{\nabla}\chi_j + \vec{e}_j$ and $\boldsymbol{\sigma} = \varepsilon\mathbf{I} + \mathbf{S}$, yielding

$$\mathbf{\Gamma}(\varepsilon\mathbf{I} + \mathbf{S})\vec{\nabla}\chi_j = -\mathbf{\Gamma}\mathbf{H}\vec{e}_j, \tag{27}$$

since $\mathbf{\Gamma}\vec{e}_j = 0$ and $\mathbf{S}\vec{e}_j = \mathbf{H}\vec{e}_j$. The equivalence of equations (26) and (27) can be seen by noting that $\vec{\nabla}\chi_j \in \mathscr{H}$ implies $\mathbf{\Gamma}\vec{\nabla}\chi_j = \vec{\nabla}\chi_j$, and then writing $1 = -\mathrm{i}^2$. It is worth mentioning that the condition $\langle\vec{J}_j\cdot\vec{\nabla}\chi_k\rangle = 0$ is equivalent to equation (A-13). Moreover, taking the inner-product of both sides of equation (27) with $\vec{\nabla}\chi_k$, averaging, using the properties $\mathbf{\Gamma}\vec{\nabla}\chi_j = \vec{\nabla}\chi_j$ and $\langle\mathbf{\Gamma}\vec{\xi}\cdot\vec{\zeta}\rangle = \langle\vec{\xi}\cdot\mathbf{\Gamma}\vec{\zeta}\rangle$ for $\vec{\xi}, \vec{\zeta} \in \mathscr{H}_\mathcal{V}$ (see Section A-1.3), and integrating by parts, also yields equation (A-13).

In Section A-1.3 we show that $\mathbf{A}$ acts as an anti-symmetric linear operator on the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}$, $\langle\mathbf{A}\vec{\xi}\cdot\vec{\zeta}\rangle = \langle\vec{\xi}\cdot\mathbf{A}^*\vec{\zeta}\rangle = -\langle\vec{\xi}\cdot\mathbf{A}\vec{\zeta}\rangle$. Therefore, $\mathbf{A}$ commutes with its (Hilbert space) adjoint $\mathbf{A}^*$ (not to be confused with an effective tensor) and is therefore an example of a *normal* operator [28]. Consequently, due to the sesquilinearity of the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$-inner-product, $\mathbf{M} = -\mathrm{i}\mathbf{A}$ acts as a *symmetric* operator [25, 28]. Moreover, on the function space $\mathscr{H}$, $\mathbf{A}$ is a *maximal* normal operator and $\mathbf{M}$ is *self-adjoint* [28]. In Section 2.2.2 we examine these properties of $\mathbf{A}$ and $\mathbf{M}$ in more detail and demonstrate how equation (26) and the spectral theory of such operators lead to an integral representation for the symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of $\boldsymbol{\mathcal{K}}^*$.

2.2.2. *Integral representation of the effective diffusivity for steady and dynamic flows.* In this section, we employ the Hilbert space formulation of the effective parameter problem discussed in Section 2.2.1, to provide integral representations for the symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$. These integral representations involve a *spectral measure* associated with the (maximal) normal operator $\mathbf{A} = \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}$ on $\mathscr{H}$, or equivalently the self-adjoint operator $\mathbf{M}=-\mathrm{i}\mathbf{A}$, and follow from the spectral theorem for such operators [25, 28] and the resolvent formula for $\vec{\nabla}\chi_k$ given in equation (26). There are significant differences in the theory between the case of steady flows, where $\mathbf{S}=\mathbf{H}$ is *bounded* on the Hilbert space $\mathscr{H}_{\mathcal{V}}$, and the case of dynamic flows, where $\mathbf{S}=\mathbf{H}-(\boldsymbol{\Delta}^{-1})\mathbf{T}$ is *unbounded* on the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}$, as discussed in Section 2.2.1. It is therefore natural to start our discussion with a more detailed look into this distinction, in the present context.

Since $\boldsymbol{\Gamma}$ is an orthogonal projector from $\mathscr{H}_{\mathcal{V}}$ to $\mathscr{H}_{\times}$, it is bounded by unity in operator norm $\|\boldsymbol{\Gamma}\|\leq 1$ on $\mathscr{H}_{\mathcal{V}}$ and $\|\boldsymbol{\Gamma}\|=1$ on $\mathscr{H}_{\times}$ [25, 28]. Therefore by (16), in the case of steady flows, the operator $\mathbf{A}=\boldsymbol{\Gamma}\mathbf{H}\boldsymbol{\Gamma}$ is bounded on the Hilbert space $\mathscr{H}_{\mathcal{V}}$, with $\|\mathbf{A}\|\leq\|\mathbf{H}\|<\infty$. Let's first focus on this time-independent case. Since $\mathbf{M}=-\mathrm{i}\mathbf{A}$ we have $\|\mathbf{M}\|=\|\mathbf{A}\|$, so the domains of these two operators are identical, $D(\mathbf{M})=D(\mathbf{A})$. For simplicity we focus on the operator $\mathbf{M}$ now, re-introducing the operator $\mathbf{A}$ later. The (Hilbert space) adjoint $\mathbf{M}^*$ of $\mathbf{M}$ is defined by $\langle\mathbf{M}\vec{\xi},\vec{\zeta}\,\rangle=\langle\vec{\xi},\mathbf{M}^*\vec{\zeta}\,\rangle$, and is also a bounded operator on $\mathscr{H}_{\mathcal{V}}$ with $\|\mathbf{M}^*\|=\|\mathbf{M}\|$ [25]. Consequently, they have identical domains,

$$D(\mathbf{M})=D(\mathbf{M}^*), \tag{28}$$

which are the entire space, $D(\mathbf{M})=D(\mathbf{M}^*)=\mathscr{H}_{\mathcal{V}}$. In Section A-1.3 we show that $\mathbf{M}$ is symmetric,

$$\langle\mathbf{M}\vec{\xi}\cdot\vec{\zeta}\,\rangle=\langle\vec{\xi}\cdot\mathbf{M}\vec{\zeta}\rangle,\ \text{ for all }\ \vec{\xi},\vec{\zeta}\in D(\mathbf{M}). \tag{29}$$

By definition [25, 28], the two properties (28) and (29) together imply that the operator $\mathbf{M}$ is *self-adjoint*, i.e. $\mathbf{M}\equiv\mathbf{M}^*$ on $D(\mathbf{M})$.

Conversely, the Hellinger–Toeplitz theorem [25] states, if the operator $\mathbf{M}$ satisfies equation (29) for *every* $\vec{\xi},\vec{\zeta}\in\mathscr{H}_{\mathcal{V}}$, then $\mathbf{M}$ is bounded. This suggests that, in the time-dependent case when $\mathbf{M}$ is unbounded on the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}$, it is defined as a self-adjoint operator only on a proper subset of $\mathscr{H}_{\mathcal{T}\mathcal{V}}$. However, as discussed in Section 2.2.1, the domain $D(\mathbf{M})$ can be defined as a *dense* subset of $\mathscr{H}_{\mathcal{T}\mathcal{V}}$ such that $\mathbf{M}$ is bounded. Moreover, on this domain, $\mathbf{M}$ can be extended to a *closed* symmetric operator [25, 28]. Although even in this case, in general [25], the domain $D(\mathbf{M}^*)$ of the associated adjoint $\mathbf{M}^*$ does not coincide with $D(\mathbf{M})$, and in such circumstances $\mathbf{M}$ is *not* self-adjoint on $D(\mathbf{M})$. Only for self-adjoint (or maximal normal) operators does the spectral theorem hold [25], which provides the existence of the promised integral representation for $\boldsymbol{\mathcal{K}}^*$, involving a spectral measure associated with $\mathbf{M}$. It is therefore necessary that we find a domain $D(\mathbf{M})$ on which $\mathbf{M}$ is self-adjoint.

As $\boldsymbol{\Gamma}$ is bounded on $\mathscr{H}_{\mathcal{V}}$ and $\mathbf{M}=-\mathrm{i}\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$, our discussion in Section 2.2.1 indicates that the unboundedness of $\mathbf{M}$ on $\mathscr{H}_{\mathcal{T}\mathcal{V}}$ is due to the unboundedness of the underlying operator $\mathbf{T}$ on the Hilbert space $\mathscr{H}_{\mathcal{T}}$. It is therefore necessary that we find a domain $D(\mathbf{T})$ for which $\mathbf{T}$ is a self-adjoint operator. Toward this goal, and to illustrate these ideas, we consider the operator $\mathrm{i}\partial_t$ with the three different domains $\mathscr{A}_{\mathcal{T}}$, $\tilde{\mathscr{A}_{\mathcal{T}}}$, and $\hat{\mathscr{A}_{\mathcal{T}}}$ defined in equations (19) and (20), which are everywhere dense in $L^2(\mathcal{T})$ [28]. Let the operators B, $\tilde{B}$, and $\hat{B}$ be identified as $\mathrm{i}\partial_t$ with domains $\mathscr{A}_{\mathcal{T}}$, $\tilde{\mathscr{A}_{\mathcal{T}}}$, and $\hat{\mathscr{A}_{\mathcal{T}}}$, respectively. Then, $\hat{B}$ is a closed linear symmetric operator with adjoint $\hat{B}^*\equiv B$, and the operator $\tilde{B}$ is a *self-adjoint* extension of $\hat{B}$ [28]. In symbols, this means that $\tilde{B}=\tilde{B}^*$ on $\tilde{\mathscr{A}_{\mathcal{T}}}$ and $D(\tilde{B})=D(\tilde{B}^*)=\tilde{\mathscr{A}_{\mathcal{T}}}$, i.e. $\tilde{B}\equiv\tilde{B}^*$ on $\tilde{\mathscr{A}_{\mathcal{T}}}$.

Since the operator $\tilde{B}=\mathrm{i}\partial_t$ with domain $\tilde{\mathscr{A}_{\mathcal{T}}}$ is self-adjoint, it follows that the operator $\mathrm{i}\mathbf{T}=\mathrm{i}\partial_t\mathbf{I}$ with domain $D(\mathbf{T})=\mathscr{D}_{\mathcal{T}}=\otimes_{i=1}^{d}\tilde{\mathscr{A}_{\mathcal{T}}}$ is self-adjoint. This is seen as follows. By noting that $\mathrm{i}\mathbf{T}\vec{\xi}=(\tilde{B}\xi_1,\ldots,\tilde{B}\xi_d)$ and, for all $\vec{\xi},\vec{\zeta}\in\mathscr{D}_{\mathcal{T}}$ with components $\xi_j,\zeta_j\in\tilde{\mathscr{A}_{\mathcal{T}}}$, $j=1,\ldots,d$, the self-adjointness of $\tilde{B}$ implies that $\mathbf{T}$ is symmetric, $\mathbf{T}=\mathbf{T}^*$, on $\mathscr{D}_{\mathcal{T}}$,

$$\langle\mathbf{T}\vec{\xi}\cdot\vec{\zeta}\rangle=\sum_j\langle\tilde{B}\xi_j,\zeta_j\rangle_2=\sum_j\langle\xi_j,\tilde{B}\zeta_j\rangle_2=\langle\vec{\xi}\cdot\mathbf{T}\vec{\zeta}\rangle, \tag{30}$$

where $\langle\cdot,\cdot\rangle_2$ denotes the $L^2(\mathcal{T})$ inner-product. Moreover, since we have $D(\tilde{B}) = D(\tilde{B}^*) = \tilde{\mathscr{A}}_{\mathcal{T}}$, we also have $D(\mathbf{T}) = D(\mathbf{T}^*) = \mathscr{D}_{\mathcal{T}}$, i.e. $\mathbf{T} \equiv \mathbf{T}^*$ on $\mathscr{D}_{\mathcal{T}}$.

We now summarize what we have discussed so far, and discuss the implications thereof. We have discussed that the operators $(\mathbf{\Delta}^{-1})$ and $\mathbf{\Gamma}$ are bounded on the Hilbert space $\mathscr{H}_{\mathcal{V}}$. In Section A-1.3 we show that they are also symmetric, hence self-adjoint on $\mathscr{H}_{\mathcal{V}}$. Due to the sesquilinearity of the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$-inner-product, and equations (16) and (3) with $\mathbf{H}^* = \mathbf{H}^T$, the operator $\mathrm{i}\mathbf{H}$ is bounded and symmetric, hence self-adjoint on the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}$. Consequently, the operator $\mathrm{i}\mathbf{\Gamma}\mathbf{H}\mathbf{\Gamma}$ is also self-adjoint on $\mathscr{H}_{\mathcal{T}\mathcal{V}}$. The differential and integral operators $\mathrm{i}\mathbf{T}$ and $(\mathbf{\Delta}^{-1})$ are bounded on the function space $\mathscr{D}_{\mathcal{T}}$ and Hilbert space $\mathscr{H}_{\mathcal{V}}$, respectively, and they are consequently commutable operations on the function space $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$ [10]. Moreover, as $\mathrm{i}\mathbf{T}$ and $(\mathbf{\Delta}^{-1})$ are self-adjoint on $\mathscr{D}_{\mathcal{T}}$ and $\mathscr{H}_{\mathcal{V}}$, respectively, the operator $\mathrm{i}(\mathbf{\Delta}^{-1})\mathbf{T}$, hence $\mathrm{i}\mathbf{\Gamma}[(\mathbf{\Delta}^{-1})\mathbf{T}]\mathbf{\Gamma}$ is self-adjoint on $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$. It is now clear that the operator $\mathbf{M} = \mathrm{i}\mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}$, with $\mathbf{S} = \mathbf{H} - (\mathbf{\Delta}^{-1})\mathbf{T}$, is self-adjoint on $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$. Finally, since $\mathbf{M} = -\mathrm{i}\mathbf{A}$ is self-adjoint on $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$ and an operator is self-adjoint if and only if it is a maximal normal operator [28], we have that $\mathbf{A}$ is a maximal normal operator on $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$. In view of the resolvent representation of $\vec{\nabla}\chi_j \in \mathscr{H}$ in (26) involving these operators, we will henceforth take the domain of these operators to be $D(\mathbf{A}) = D(\mathbf{M}) = \mathscr{H}$ in (21), which is a *closed* subset of $\mathscr{D}_{\mathcal{T}} \times \mathscr{H}_{\mathcal{V}}$.

In terms of a general, maximal normal operator $\mathbf{N}$ on $\mathscr{H}$, i.e. $\mathbf{N}\mathbf{N}^* = \mathbf{N}^*\mathbf{N}$, the spectral theorem states that $\mathbf{N}$ can be decomposed as $\mathbf{N} = \mathbf{H}_1 + \mathrm{i}\mathbf{H}_2$, where $\mathbf{H}_1$ and $\mathbf{H}_2$ are self-adjoint and commute on $\mathscr{H}$ [28]. Moreover, there is a one-to-one correspondence between $\mathbf{H}_i$, $i = 1, 2$, and a family $\{\mathbf{Q}_i(\lambda)\}$, $-\infty < \lambda \leq \infty$, of self-adjoint projection operators - the resolution of the identity - with domain $\mathscr{H}$ which satisfies $\lim_{\lambda\to-\infty}\mathbf{Q}_i(\lambda) = 0$, $\lim_{\lambda\to+\infty}\mathbf{Q}_i(\lambda) = \mathbf{I}$, and the $\mathbf{Q}_i(\lambda)$, $i = 1, 2$, commute [25, 28]. Consequently, there is a one-to-one correspondence between the maximal normal operator $\mathbf{N}$ and a family $\{\mathbf{Q}(z)\}$, $\mathbf{Q}(z) = \mathbf{Q}_1(\mathrm{Re}(z))\mathbf{Q}_2(\mathrm{Im}(z))$, $z = \lambda_1 + \mathrm{i}\lambda_2$, $-\infty < \lambda_1, \lambda_2 \leq \infty$, of self-adjoint projection operators - the *complex* resolution of the identity - which satisfies $\mathbf{Q}(z) \to 0$ when $\mathrm{Re}(z) \to -\infty$ and when $\mathrm{Im}(z) \to -\infty$, and $\mathbf{Q}(z) \to \mathbf{I}$ when $\mathrm{Re}(z) \to +\infty$ and when $\mathrm{Im}(z) \to +\infty$ [28].

The spectral theorem also provides an operational calculus in Hilbert space which yields integral representations associated with $\mathbf{Q}(z)$-measurable functions of $\mathbf{N}$ [28]. The details are as follows. Let $\vec{\xi}, \vec{\zeta} \in \mathscr{H}$ and consider the *complex-valued* function $\mu_{\xi\zeta}(z) = \langle\mathbf{Q}(z)\vec{\xi}\cdot\vec{\zeta}\rangle$, $\vec{\xi} \neq \vec{\zeta}$. By the sesquilinearity of the inner-product and the self-adjointness of the projection operator $\mathbf{Q}(z)$ we have $\mu_{\zeta\xi}(z) = \overline{\mu}_{\xi\zeta}(z)$, where $\overline{\mu}_{\xi\zeta}$ denotes complex conjugation of the function $\mu_{\xi\zeta}$. Moreover, the function $\mu_{\xi\xi}$ is real-valued $\mu_{\xi\xi}(z) = \langle\mathbf{Q}(z)\vec{\xi}\cdot\vec{\xi}\rangle = \langle\mathbf{Q}(z)\vec{\xi}\cdot\mathbf{Q}(z)\vec{\xi}\rangle = \|\mathbf{Q}(z)\vec{\xi}\|^2$. We associate with these functions of *bounded variation* Radon–Stieltjes measures $\mathrm{d}\mu_{\xi\zeta}(z)$ and $\mathrm{d}\mu_{\xi\xi}(z)$ [28]

$$\mathrm{d}\mu_{\xi\zeta}(z) = \mathrm{d}\langle\mathbf{Q}(z)\vec{\xi}\cdot\vec{\zeta}\rangle, \quad \mathrm{d}\mu_{\xi\xi}(z) = \mathrm{d}\|\mathbf{Q}(z)\vec{\xi}\|^2. \tag{31}$$

Let $F(z)$ be an arbitrary complex-valued function and denote by $\mathscr{D}(F)$ the set of all $\vec{\xi} \in \mathscr{H}$ such that $F \in L^2(\mu_{\xi\xi})$, the class of functions that are square integrable with respect to the measure $\mathrm{d}\mu_{\xi\xi}$. Then $\mathscr{D}(F)$ is a linear manifold and there exists a linear transformation $F(\mathbf{N})$ with domain $\mathscr{D}(F)$ defined in terms of the Radon–Stieltjes integrals [28]

$$\begin{aligned} \langle F(\mathbf{N})\vec{\xi}\cdot\vec{\zeta}\rangle &= \int_I F(z)\,\mathrm{d}\mu_{\xi\zeta}(z), \qquad \forall\vec{\xi} \in \mathscr{D}(F),\ \vec{\zeta} \in \mathscr{H} \\ \langle F(\mathbf{N})\vec{\xi}\cdot G(\mathbf{N})\vec{\zeta}\rangle &= \int_I F(z)\overline{G}(z)\,\mathrm{d}\mu_{\xi\zeta}(z), \quad \forall\vec{\xi} \in \mathscr{D}(F),\ \vec{\zeta} \in \mathscr{D}(G), \end{aligned} \tag{32}$$

where the operator $G(\mathbf{N})$ and set $\mathscr{D}(G)$ are defined analogously to that for F. The domain of integration I in (32) is determined by the support of the spectrum of the operator $\mathbf{N}$, and in general $I \subseteq (-\infty, \infty] \times (-\mathrm{i}\infty, \mathrm{i}\infty]$. A Radon–Stieltjes integral representation of the functional

$\|F(\mathbf{N})\vec{\xi}\|^2$ follows from the second equation in (32) with $G = F$ and $\vec{\xi} = \vec{\zeta}$, and involves the measure $\mathrm{d}\mu_{\xi\xi}$ in equation (31) [28].

The spectral theorem in (32) for the maximal normal operator $\mathbf{N}$ on $\mathscr{H}$ generalizes that for self-adjoint and maximal anti-symmetric operators, with purely real and imaginary spectrum, respectively. More specifically, the case $F(z) = z = \lambda_1 + \mathrm{i}\lambda_2$ corresponds to $F(\mathbf{N}) = \mathbf{H}_1 + \mathrm{i}\mathbf{H}_2$ with $I \subseteq (-\infty, \infty] \times (-\mathrm{i}\infty, \mathrm{i}\infty]$ and $\mathbf{Q}(z) = \mathbf{Q}_1(\mathrm{Re}(z))\mathbf{Q}_2(\mathrm{Im}(z))$, the case $F(z) = \mathrm{Re}(z)$ corresponds to the self-adjoint operator $F(\mathbf{N}) = \mathbf{H}_1$ with $I \subseteq (-\infty, \infty]$ and $\mathbf{Q}(z) = \mathbf{Q}_1(\mathrm{Re}(z))\mathbf{Q}_2(0)$, and the case $F(z) = \mathrm{i}\,\mathrm{Im}(z)$ corresponds to the maximal anti-symmetric operator $F(\mathbf{N}) = \mathrm{i}\mathbf{H}_2$ with $I \subseteq (-\mathrm{i}\infty, \mathrm{i}\infty]$ and $\mathbf{Q}(z) = \mathbf{Q}_1(0)\mathbf{Q}_2(\mathrm{Im}(z))$.

We now employ equation (26) and the spectral theorem in (32) to provide Radon–Stieltjes integral representations for the symmetric $\boldsymbol{\kappa}^*$ and anti-symmetric $\boldsymbol{\alpha}^*$ parts of the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$, for both cases of time-independent and time-dependent velocity fields $\vec{u}$. These representations are summarized by the following theorem.

Theorem 2.2. *Let $z = \mathrm{i}\lambda$, $\vec{g}_j = -\boldsymbol{\Gamma}\mathbf{H}\vec{e}_j$ be defined as in (26), and $\mathbf{Q}(z) := \mathbf{Q}(\lambda) = \mathbf{Q}_1(0)\mathbf{Q}_2(\lambda)$ be the complex resolution of the identity associated with the maximal anti-symmetric operator $\mathbf{A}$ defined in (26) with domain $\mathscr{H}$ defined in (21). Define the complex-valued function $\mu_{jk}(\lambda) = \langle \mathbf{Q}(\lambda)\vec{g}_j \cdot \vec{g}_k \rangle$, $j \neq k = 1, \ldots, d$, with $\mu_{kj} = \overline{\mu}_{jk}$, and the real-valued function $\mu_{kk}(\lambda) = \|\mathbf{Q}(\lambda)\vec{g}_k\|^2$. Moreover, consider the real-valued functions*

$$\mathrm{Re}\,\mu_{jk}(\lambda) = \frac{1}{2}\left(\mu_{jk}(\lambda) + \overline{\mu}_{jk}(\lambda)\right), \quad \mathrm{Im}\,\mu_{jk}(\lambda) = \frac{1}{2\,\mathrm{i}}\left(\mu_{jk}(\lambda) - \overline{\mu}_{jk}(\lambda)\right). \tag{33}$$

Corresponding to each of these functions, of bounded variation for $\vec{g}_j \in \mathscr{H}$ [28], we consider the associated Radon–Stieltjes measures $\mathrm{d}\mu_{jk}(\lambda)$, $\mathrm{d}\mu_{kk}(\lambda)$, $\mathrm{dRe}\,\mu_{jk}(\lambda)$, and $\mathrm{dIm}\,\mu_{jk}(\lambda)$. Then there exist Radon–Stieltjes integral representations for the components κ^*_{kk}, κ^*_{jk}, and α^*_{jk}, $j \neq k = 1, \ldots, d$, of the effective tensors $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ defined in (9) given by

$$\kappa^*_{kk} = \varepsilon\left(1 + \int_{-\infty}^{\infty} \frac{\mathrm{d}\mu_{kk}(\lambda)}{\varepsilon^2 + \lambda^2}\right), \quad \kappa^*_{jk} = \varepsilon \int_{-\infty}^{\infty} \frac{\mathrm{dRe}\,\mu_{jk}(\lambda)}{\varepsilon^2 + \lambda^2}, \quad \alpha^*_{jk} = \int_{\infty}^{\infty} \frac{\lambda\,\mathrm{dIm}\,\overline{\mu}_{jk}(\lambda)}{\varepsilon^2 + \lambda^2}. \tag{34}$$

Here, the domain of integration $I \subseteq (-\infty, \infty]$ is determined by the support of the spectrum of the operator $\mathbf{A}$, where $I \subseteq [-\|\mathbf{A}\|, \|\mathbf{A}\|]$ for the case of time-independent velocity fields $\vec{u}$ for which $\|\mathbf{A}\| \leq \|\mathbf{H}\| < \infty$ [25].

Proof of Theorem 2.2 We first note that from $\vec{\nabla}\chi_j \in \mathscr{H}$, we have the property $\vec{\nabla}\chi_j = \boldsymbol{\Gamma}\vec{\nabla}\chi_j$, so that the components α^*_{jk}, $j, k = 1, \ldots, d$, of the effective tensor $\boldsymbol{\alpha}^*$ in equation (9) can re-expressed as $\alpha^*_{jk} = \langle \mathbf{S}\vec{\nabla}\chi_j \cdot \vec{\nabla}\chi_k \rangle = \langle \boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}\vec{\nabla}\chi_j \cdot \vec{\nabla}\chi_k \rangle = \langle \mathbf{A}\vec{\nabla}\chi_j \cdot \vec{\nabla}\chi_k \rangle$, where we have used that $\boldsymbol{\Gamma}$ is self-adjoint on $\mathscr{H}$. From this and (26), equation (9) can be rewritten as

$$\kappa^*_{jk} = \varepsilon(\delta_{jk} + \langle (\varepsilon\mathbf{I} + \mathbf{A})^{-1}\vec{g}_j \cdot (\varepsilon\mathbf{I} + \mathbf{A})^{-1}\vec{g}_k \rangle), \quad \alpha^*_{jk} = \langle \mathbf{A}(\varepsilon\mathbf{I} + \mathbf{A})^{-1}\vec{g}_j \cdot (\varepsilon\mathbf{I} + \mathbf{A})^{-1}\vec{g}_k \rangle, \tag{35}$$

where $\vec{g}_j = -\boldsymbol{\Gamma}\mathbf{H}\vec{e}_j$. The integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ follow from equation (35) and the spectral theorem of operational calculus in Hilbert space for maximal normal operators, or equivalently that for self-adjoint operators, since $\mathbf{M} = -\mathrm{i}\mathbf{A}$ [25, 28].

We prove that equation (34) holds by applying equation (32) to (35), showing that the conditions of the spectral theorem are met, and using the symmetry properties $\langle \vec{\nabla}\chi_j \cdot \vec{\nabla}\chi_k \rangle = \langle \vec{\nabla}\chi_k \cdot \vec{\nabla}\chi_j \rangle$ and $\langle \mathbf{A}\vec{\nabla}\chi_j \cdot \vec{\nabla}\chi_k \rangle = \langle \vec{\nabla}\chi_k \cdot \mathbf{A}\vec{\nabla}\chi_j \rangle$, as the vector field $\vec{\nabla}\chi_j$ is real-valued and $\mathbf{A} : \mathbb{R}^d \to \mathbb{R}^d$.

These functions are of bounded variation, since $\vec{g}_k = -\boldsymbol{\Gamma}\mathbf{H}\vec{e}_k$ is bounded $\|\vec{g}_k\| \leq \|\mathbf{H}\| < \infty$.

follow from equations (9) and (26), and the second formula in (32) with

$$F(\lambda) = G(\lambda) = (\varepsilon - \lambda)^{-1} = -\mathrm{i}(-\mathrm{i}\varepsilon - \lambda)^{-1}, \quad \vec{\xi} = \vec{g}_j, \quad \vec{\zeta} = \vec{g}_k, \quad \vec{g}_j = \boldsymbol{\Gamma}\mathbf{H}\vec{e}_j, \quad j, k = 1, \ldots, d. \tag{36}$$

To apply the spectral theory

$\vec{g}_j \in \mathscr{D}(F)$, for all $\varepsilon > 0$ and $j = 1, \ldots, d$

More specifically, the orthogonality of the projection operators $\mathbf{\Gamma}_\times = \mathbf{\Gamma}$ and $\mathbf{\Gamma}_0$ in (15) implies that the vector field $\vec{g}_j(t,\vec{x}) = \mathbf{\Gamma}\mathbf{H}(t,\vec{x})\vec{e}_j$ is curl-free and mean-zero. Moreover, by equation (16), $\vec{g}_j$ and $\partial_t\vec{g}_j$ are bounded on $\mathscr{H}$, with $\|\vec{g}_j\| \leq \|\mathbf{H}\|$ and $\|\partial_t\vec{g}_j\| \leq \|\partial_t\mathbf{H}\|$, so that $\vec{g}_j \in \mathscr{H}$ for all $j = 1, \ldots, d$. We now show that Also, in the case of the spectral theorem involving $\mathbf{A}$, $\varepsilon \notin I \subseteq (-\mathrm{i}\infty, \mathrm{i}\infty]$, and in the case of the spectral theorem involving $\mathbf{M}$, $\mathrm{i}\varepsilon \notin I \subseteq (-\infty, \infty]$ for all $\varepsilon > 0$ and the measure $\mu_{\xi\xi}(d\lambda)$ is of bounded mass [28]

$$\mu^0_{\xi\xi} = \int_I \mu_{\xi\xi}(d\lambda) = \int_I \langle \mathbf{Q}(d\lambda)\vec{\xi}\cdot\vec{\xi}\,\rangle = \|\vec{\xi}\,\|^2 = \langle \mathbf{H}^T\mathbf{\Gamma}\mathbf{H}\vec{e}_k\cdot\vec{e}_k\rangle \leq \|\mathbf{H}\|^2 < \infty, \tag{37}$$

we have that $F(\lambda)$ in (36) satisfies $F \in L^2(\mu_{\xi\xi})$, hence $\vec{g}_j \in \mathscr{D}(F)$, for all $\varepsilon > 0$ and $j = 1, \ldots, d$. Therefore, the following Radon-Stieltjes integral representation for the components κ^*_{jk} of $\boldsymbol{\kappa}^*$, involving the components $\mu_{jk}(d\lambda)$ of the matrix-valued measure $\boldsymbol{\mu}(d\lambda)$, holds for all $\varepsilon > 0$

We conclude this section with a few remarks regarding the integral representation in (34). The Radon measure $\mu_{jk}(d\lambda)$ is a *spectral measure* associated with the self-adjoint linear operator $\mathbf{M}$ in the $(\vec{g}_j, \vec{g}_k)$ state [25]. Since $\mathbf{Q}(\lambda)$ is a projection operator, the diagonal components of $\boldsymbol{\mu}(d\lambda)$ are *positive* measures, $\mu_{kk}(d\lambda) = \|\mathbf{Q}(d\lambda)\vec{g}_k\|^2$. In the case of a time-independent velocity field $\vec{u}$, where $\mathbf{M} = \mathbf{\Gamma}\mathbf{H}\mathbf{\Gamma}$, the range of integration in equation (34) is given by $-\|\mathbf{M}\| \leq \lambda \leq \|\mathbf{M}\|$, with $\|\mathbf{M}\| \leq \|\mathbf{H}\| < \infty$, by (16). A key feature of the integral representation for $\boldsymbol{\kappa}^*$ in (34) is that parameter information in ε is *separated* from the geometry and dynamics of the velocity field, which is encapsulated in the measure $\boldsymbol{\mu}$. In Section 3 we employ the integral representation for $\boldsymbol{\kappa}^*$ in (34) to obtain asymptotic behavior of κ^*_{jk} as $\varepsilon \to 0$.

3. Asymptotic analysis of effective diffusivity

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

$$\mathbf{H} = \begin{bmatrix} 0 & H \\ -H & 0 \end{bmatrix}, \qquad \vec{u} = [\partial_{x_1}H,\ \partial_{x_2}H]. \tag{38}$$

4. Numerical Results

Since we are focusing on flows which are periodic on the spatial region $\mathcal{V}$, it is convenient to consider the Fourier representation of such a vector field $\vec{\xi}(t,\vec{x})$,

$$\vec{\xi}(t,\vec{x}) = \sum_{\vec{k}\in\mathbb{Z}^d} \hat{Y}(t,\vec{k})\mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{x}}, \qquad \hat{Y}(t,\vec{k}) = \frac{1}{(2\pi)^d}\int_{\mathcal{V}} \hat{Y}(t,\vec{x})\mathrm{e}^{-\mathrm{i}\vec{k}\cdot\vec{x}}\mathrm{d}\vec{x}. \tag{39}$$

The associated action of the above projection operators on a function $\vec{\xi} \in \mathscr{H}$ is given by [8]

$$\mathbf{\Gamma}_0\vec{\xi}(t,\vec{x}) = \langle\vec{\xi}(t,\vec{x})\rangle_x = \hat{Y}(t,0), \qquad \mathbf{\Gamma}_\times\vec{\xi}(t,\vec{x}) = \sum_{\vec{k}\neq 0} \frac{\vec{k}(\vec{k}\cdot\hat{Y}(t,\vec{k}))}{|\vec{k}|^2}\mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{x}}, \tag{40}$$

$$\mathbf{\Gamma}_\bullet\vec{\xi}(t,\vec{x}) = \sum_{\vec{k}\neq 0} \frac{\vec{k}\times(\vec{k}\times\hat{Y}(t,\vec{k}))}{|\vec{k}|^2}\mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{x}} = \sum_{\vec{k}\neq 0}\left(I - \frac{\vec{k}\vec{k}\cdot}{|\vec{k}|^2}\right)\hat{Y}(t,\vec{k})\mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{x}},$$

where $\langle\cdot\rangle_x$ denotes spatial averaging over $\mathcal{V}$. From equation (40) it is clear that $\mathbf{\Gamma}_\times + \mathbf{\Gamma}_\bullet + \mathbf{\Gamma}_0 = \mathbf{I}$.

A-1. Appendix

A-1.1. **The flow matrix H.**

A-1.2. **Multiple scale method.** In this section we provide the details of the multiple scale method [16, 21, 22, 4] which leads to equations (6)–(9). We assume that equation (1) has already been non-dimensionalized so that $\kappa_0 \mapsto \varepsilon$ and $\vec{v} \mapsto \vec{u}$. The key assumption of the method is that the initial density ϕ_0 in (1) is slowly varying relative to the velocity field $\vec{u}$, which introduces a small parameter $\delta \ll 1$ such that

$$\phi(0,\vec{x}) = \phi_0(\delta\vec{x}). \tag{A-2}$$

The variable changes $\vec{x} \mapsto \vec{y} = \vec{x}/\delta$ and $t \mapsto \tau = t/\delta^2$, along with equations (2) and (A-2), transforms equation (1) into [16]

$$\partial_t\phi^\delta(t,\vec{x}) = \varepsilon\Delta\phi^\delta(t,\vec{x}) + \delta^{-1}\,\vec{u}(\tau,\vec{y})\cdot\vec{\nabla}\phi^\delta(t,\vec{x}), \quad \phi^\delta(0,\vec{x}) = \phi_0(\vec{x}). \tag{A-3}$$

We now expand ϕ^δ in powers of δ [16]

$$\phi^\delta(t,\vec{x}) = \bar{\phi}(t,\vec{x}) + \delta\phi^{(1)}(t,\vec{x},\tau,\vec{y}) + \delta^2\phi^{(2)}(t,\vec{x},\tau,\vec{y}) + \cdots. \tag{A-4}$$

Writing

$$\partial_t\phi^{(i)} = [\partial_t + \delta^{-2}\partial_\tau]\phi^{(i)}, \quad \vec{\nabla}\phi^{(i)} = [\vec{\nabla}_x + \delta^{-1}\vec{\nabla}_y]\phi^{(i)}, \quad \Delta\phi^{(i)} = [\Delta_x + 2\delta^{-1}\vec{\nabla}_x\cdot\vec{\nabla}_y + \delta^{-2}\Delta_y]\phi^{(i)},$$

for the functions $\phi^{(i)}$, $i = 1, 2, \ldots$, of the fast $(\tau,\vec{y})$ and slow $(t,\vec{x})$ variables, we find that

$$\begin{aligned}
\partial_t\phi^\delta &= \delta^{-2}[\partial_\tau\bar{\phi}] + \delta^{-1}[\partial_\tau\phi^{(1)}] + \delta^0[\partial_t\bar{\phi} + \partial_\tau\phi^{(2)}] + O(\delta),\\
\vec{\nabla}\phi^\delta &= \delta^{-2}[0] + \delta^{-1}[\vec{\nabla}_y\bar{\phi}] + \delta^0[\vec{\nabla}_x\bar{\phi} + \vec{\nabla}_y\phi^{(1)}] + \delta^1[\vec{\nabla}_x\phi^{(1)} + \vec{\nabla}_y\phi^{(2)}] + O(\delta^2),\\
\Delta\phi^\delta &= \delta^{-2}[\Delta_y\bar{\phi}] + \delta^{-1}[2\vec{\nabla}_x\cdot\vec{\nabla}_y\bar{\phi} + \Delta_y\phi^{(1)}] + \delta^0[\Delta_x\bar{\phi} + 2\vec{\nabla}_x\cdot\vec{\nabla}_y\phi^{(1)} + \Delta_y\phi^{(2)}] + O(\delta).
\end{aligned} \tag{A-5}$$

Inserting this into equation (A-3) and setting the coefficients associated with the various powers of δ to zero, yields a sequence of problems.

Due to the dependence of $\bar{\phi}(t,\vec{x})$ on only the slow variables, the coefficients of δ^{-2} vanish. Equating the coefficients of δ^{-1} and δ^0 to zero we, respectively, obtain

$$\partial_\tau\phi^{(1)} - \varepsilon\Delta_y\phi^{(1)} - \vec{u}\cdot\vec{\nabla}_y\phi^{(1)} = \vec{u}\cdot\vec{\nabla}_x\bar{\phi}, \tag{A-6}$$

$$\partial_\tau\phi^{(2)} - \vec{u}\cdot\vec{\nabla}_y\phi^{(2)} - \varepsilon\Delta_y\phi^{(2)} = -\partial_t\bar{\phi} + \vec{u}\cdot\vec{\nabla}_x\phi^{(1)} + \varepsilon[\Delta_x\bar{\phi} + 2\vec{\nabla}_x\cdot\vec{\nabla}_y\phi^{(1)}]. \tag{A-7}$$

By the linearity of equation (A-6), we may separate the fast and slow variables by writing [16]

$$\phi^{(1)}(t,\vec{x},\tau,\vec{y}) = \vec{\chi}(\tau,\vec{y})\cdot\vec{\nabla}_x\bar{\phi}(t,\vec{x}). \tag{A-8}$$

When the components χ_k, $k = 1, \ldots, d$, of $\vec{\chi}$ satisfy

$$\partial_\tau\chi_k - \varepsilon\Delta_y\chi_k - \vec{u}\cdot\vec{\nabla}_y\chi_k = \vec{u}\cdot\vec{e}_k, \tag{A-9}$$

equation (A-6) is automatically satisfied [16]. Equation (A-9) along with (3) is equivalent to the cell problem (7), where the distinction of fast variables was dropped for notational simplicity. In order for $\phi^{(1)}(t,\vec{x},\tau,\vec{y})$ in (A-8) to be periodic in $(\tau,\vec{y})$ for each fixed $(t,\vec{x})$, we must have that the functions $\chi_k(\tau,\vec{y})$, $k = 1, \ldots, d$, are periodic. This and the fundamental theorem of calculus implies that $\langle\vec{\nabla}_y\chi_k\rangle = 0$. Here, $\langle\cdot\rangle$ denotes space-time averaging with respect to the *fast variables*.

Due to the incompressibility of the velocity field $\vec{\nabla}_y\cdot\vec{u}(\tau,\vec{y}) = 0$ and the *a priori* fast variable periodicity of the functions $\phi^{(i)}$, $i = 1, 2$, the fundamental theorem of calculus and the divergence theorem shows that the average of the left-hand-sides of equations (A-6) and (A-7) are zero. For the equations to have solutions, the average of the right-hand-sides must also vanish. The resulting solvability conditions are $\langle\vec{u}\rangle = 0$ and the following equation which governs the large-scale (slow variable) dynamics

$$\partial_t\bar{\phi} = \varepsilon\Delta_x\bar{\phi} + \langle\vec{u}\cdot\vec{\nabla}_x\phi^{(1)}\rangle. \tag{A-10}$$

Here, we have used that $\bar{\phi}$ is a *constant* with respect to the fast variables and, by the divergence theorem and the fast variable periodicity of $\phi^{(1)}$, we have $\langle\vec{\nabla}_y\cdot\vec{\nabla}_x\phi^{(1)}\rangle=0$. The convergence of ϕ^δ to $\bar{\phi}$ as $\delta\to 0$ is in L^2 [8],

$$\lim_{\delta\to 0}\left[\sup_{0\le t\le t_0}\int\left|\phi^\delta(t,\vec{x})-\bar{\phi}(t,\vec{x})\right|^2\mathrm{d}\vec{x}\right]=0, \tag{A-11}$$

for all $t_0<\infty$, where we have used the notation $\mathrm{d}\vec{x}=\mathrm{d}x_1\cdots\mathrm{d}x_d$ for the product Lebesgue measure.

Inserting equation (A-8) into (A-10) yields equation (6) with the components $\mathcal{K}^*_{jk}=\boldsymbol{\mathcal{K}}^*\vec{e}_j\cdot\vec{e}_k$ of the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$ given by

$$\mathcal{K}^*_{jk}=\varepsilon\delta_{jk}+\langle u_j\chi_k\rangle. \tag{A-12}$$

By inserting the representation for u_j in (A-9) into equation (A-12), the functional $\langle u_j\chi_k\rangle$ can be represented in terms of $\vec{\nabla}_y\chi_j$ and the *skew-symmetric* operator $\mathbf{S}=\mathbf{H}-\Delta_y^{-1}\mathbf{T}$, where the inverse operation Δ_y^{-1} is based on convolution with the Green's function for the Laplacian Δ_y, $\mathbf{T}=\partial_\tau\mathbf{I}$, and the $\mathbf{I}$ in this definition is to remind us that the derivative ∂_τ operates component-wise. Indeed, writing $\partial_\tau\chi_j=\vec{\nabla}_y\cdot(\Delta_y^{-1}\mathbf{T})\vec{\nabla}_y\chi_j$, $\Delta_y\chi_j=\vec{\nabla}_y\cdot\vec{\nabla}_y\chi_j$, and $\vec{u}=\vec{\nabla}_y\cdot\mathbf{H}$ in (3), we have

$$\begin{aligned}\langle u_j\chi_k\rangle&=\langle[\partial_\tau\chi_j-\varepsilon\Delta_y\chi_j-\vec{u}\cdot\vec{\nabla}_y\chi_j]\chi_k\rangle\\&=\langle\vec{\nabla}_y\cdot[(\Delta_y^{-1}\mathbf{T}-\varepsilon\mathbf{I}-\mathbf{H})\vec{\nabla}_y\chi_j]\chi_k\rangle\\&=\langle[(\mathbf{H}-\Delta_y^{-1}\mathbf{T}+\varepsilon\mathbf{I})\vec{\nabla}_y\chi_j]\cdot\vec{\nabla}_y\chi_k\rangle\\&=\langle\mathbf{S}\vec{\nabla}_y\chi_j\cdot\vec{\nabla}_y\chi_k\rangle+\varepsilon\langle\vec{\nabla}_y\chi_j\cdot\vec{\nabla}_y\chi_k\rangle,\end{aligned} \tag{A-13}$$

where we have used the periodicity of χ_k and $\mathbf{H}$ to obtain the third equality. Equations (A-12) and (A-13) are equivalent to equations (8) and (9), where the distinction of fast variables was dropped for notational simplicity.

The above analysis shows that the main part of the study of effective, diffusive transport enhanced by periodic, incompressible flows, is the study of equation (A-9), from which the effective diffusivity tensor $\boldsymbol{\mathcal{K}}^*$ emerges. In Section 2.2.1, we use the analytical structure of the cell problem (A-9) to derive a resolvent representation for $\vec{\nabla}_y\chi_k$, involving an anti-symmetric integro-differential operator $\mathbf{A}$ which is related to $\mathbf{S}=\mathbf{H}-\boldsymbol{\Delta}^{-1}\partial_t\mathbf{I}$. In Section 2.2.2, we employ this representation for $\vec{\nabla}_y\chi_k$ and the spectral theorem, to provide integral representations for $\boldsymbol{\kappa}^*$ and $\boldsymbol{\alpha}^*$ involving a *spectral measure* associated with the operator $\mathbf{A}$ acting on a suitable Hilbert space.

A-1.3. **Symmetries and commutivity.** asdf

A-1.4. **Existance and Uniqueness.** Before we discuss how the Hilbert space framework presented above leads to an integral representation for $\boldsymbol{\mathcal{K}}^*$, we first discuss some key differences in the theory between the cases of steady and dynamic velocity fields $\vec{u}$. These differences are reflected in the measure underlying this integral representation for $\boldsymbol{\kappa}^*$ and stem from the *unboundedness* of the operator ∂_t on the Hilbert space $\mathscr{H}_\mathcal{T}$ [25, 28]. For steady $\vec{u}$, in general, equation (12) reduces to (9) for diagonal components of the effective parameter. However, for dynamic $\vec{u}$, this is not true in general. The details are as follows. For dynamic $\vec{u}$, the operator $\boldsymbol{\sigma}$ in (11) can be written as $\boldsymbol{\sigma}=\varepsilon\mathbf{I}+\mathbf{S}$, where $\mathbf{S}=\mathbf{H}-\boldsymbol{\Delta}^{-1}\partial_t\mathbf{I}$ is skew-symmetric $\langle\mathbf{S}\vec{\xi},\vec{\zeta}\rangle=-\langle\vec{\xi},\mathbf{S}\vec{\zeta}\rangle$ for all $\vec{\xi},\vec{\zeta}\in\mathscr{H}$ such that $|\langle\partial_t\vec{\xi},\vec{\zeta}\rangle|,|\langle\vec{\xi},\partial_t\vec{\zeta}\rangle|<\infty$ (see Section A-1 for details). This property of the operator $\mathbf{S}$ implies that

$$\langle\mathbf{S}\vec{\xi}\cdot\vec{\xi}\rangle=-\langle\mathbf{S}\vec{\xi}\cdot\vec{\xi}\rangle=0,\qquad\mathbf{S}=\mathbf{H}-(\boldsymbol{\Delta}^{-1})\partial_t\mathbf{I}, \tag{A-14}$$

for all such $\vec{\xi}\in\mathscr{H}$. In this dynamic setting, equation (10) does not hold for every $\vec{\xi}\in\mathscr{H}$, as the unbounded operator ∂_t is defined only on a proper (dense) subset of the Hilbert space $\mathscr{H}_\mathcal{T}$ [25], and it may be that $|\langle\partial_t\vec{\xi},\vec{\xi}\rangle|=\infty$. In the case of a steady velocity field we have $\mathbf{S}\equiv\mathbf{H}$ and, by equation (16) and the Cauchy Schwartz inequality, $|\langle \mathbf{S}\vec{\xi}, \vec{\xi}\rangle| \leq \|\mathbf{H}\|\|\vec{\xi}\|^2 < \infty$ for all $\vec{\xi} \in \mathscr{H}$, so equation (10) holds for all $\vec{\xi} \in \mathscr{H}$.

Another immediate consequence of equation (10), for steady $\vec{u}$, is the coercivity of the bilinear functional $\Phi(\vec{\xi}, \vec{\zeta}) = \langle \boldsymbol{\sigma}\vec{\xi} \cdot \vec{\zeta}\rangle$ for all $\varepsilon > 0$. By equation (16), this functional is also bounded in the case of steady $\vec{u}$ for all $\varepsilon < \infty$. Therefore, the Lax-Milgram theorem [17] provides the existence and uniqueness of a solution $\vec{\nabla}\chi_k \in \mathscr{H}$ satisfying the cell problem (7), or equivalently equation (11), in this time-independent case. The details are as follows.

The distributional form of equation (7), written as $\vec{\nabla} \cdot \boldsymbol{\sigma}\vec{E}_k = 0$, is given by $\langle \boldsymbol{\sigma}(\vec{\nabla}\chi_k + \vec{e}_k) \cdot \vec{\nabla}\zeta\rangle = 0$, where ζ is a compactly supported, infinitely differentiable function on $\mathcal{T} \otimes \mathcal{V}$, and we stress that $\vec{\nabla}\zeta$ is *curl-free*. Motivated by this, we consider the following variational problem: find $\vec{\nabla}\chi_k \in \mathscr{H}$ such that

$$\langle \boldsymbol{\sigma}(\vec{\nabla}\chi_k + \vec{e}_k) \cdot \vec{\xi}\rangle = 0, \text{ for all } \vec{\xi} \in \mathscr{H}. \tag{A-15}$$

In order to directly apply the Lax-Milgram Theorem, we rewrite equation (A-15) as

$$\Phi(\vec{\nabla}\chi_k, \vec{\xi}) = \langle \boldsymbol{\sigma}\vec{\nabla}\chi_k \cdot \vec{\xi}\rangle = -\langle \boldsymbol{\sigma}\vec{e}_k \cdot \vec{\xi}\rangle = f(\vec{\xi}). \tag{A-16}$$

By equation (10) Φ is coercive, i.e.

$$\Phi(\vec{\xi}, \vec{\xi}) = \langle [(\varepsilon\mathbf{I} + \mathbf{S})]\vec{\xi} \cdot \vec{\xi}\rangle = \varepsilon\|\vec{\xi}\|^2 > 0, \text{ for all } \vec{\xi} \in \mathscr{H} \tag{A-17}$$

such that $\|\vec{\xi}\| \neq 0$ and $\varepsilon > 0$, where $\|\cdot\|$ is the norm induced by the inner-product $\langle \cdot, \cdot \rangle$. Recall that $\mathbf{S} = \mathbf{H}$ in this time-independent case. This, equation (16), the triangle inequality, and the Cauchy-Schwartz inequality imply that Φ is also bounded for all $\varepsilon < \infty$

$$\Phi(\vec{\xi}, \vec{\zeta}) \leq (\varepsilon + \|H\|)\|\vec{\xi}\|\|\vec{\zeta}\| < \infty, \text{ for all } \vec{\xi} \in \mathscr{H}. \tag{A-18}$$

For the same reasons, the linear functional $f(\vec{\xi})$ in (A-16) is bounded for all $\vec{\xi} \in \mathscr{H}$. Therefore, the Lax-Milgram theorem [17] provides the existence of a unique $\vec{\nabla}\chi_k \in \mathscr{H}$ satisfying (7) in this time-independent case.

In the time-dependent case, equation (10) hence (A-17) does not hold for all $\vec{\xi} \in \mathscr{H}$. Moreover, the operator ∂_t hence $\boldsymbol{\sigma}$ is not bounded on $\mathscr{H}$ [25, 27], so (A-18) does not hold. Consequently, the Lax-Milgram theorem cannot be directly applied, and alternate techniques [11, 12] must be used to prove the existence and uniqueness of a solution $\vec{\nabla}\chi_k \in \mathscr{H}$ satisfying the cell problem (7). This discussion illustrates key differences in the analytic structure of the effective parameter problem for $\boldsymbol{\kappa}^*$, between the cases of steady and dynamic velocity fields $\vec{u}$, which stem from the unboundedness of the operator ∂_t on $\mathscr{H}_\mathcal{T}$, hence $\boldsymbol{\sigma}$ on $\mathscr{H}$. In Section 2.2.2, we will discuss other consequences of this boundedness/unboundedness property of the operator $\boldsymbol{\sigma}$, and demonstrate that it leads to significant differences in the spectral measure underlying an integral representation of $\boldsymbol{\kappa}^*$.

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