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Spectral analysis and computation of effective diffusivities in space-time periodic incompressible flows

N. Benjamin Murphy, [*,†,‡] Elena Cherkaev, [†,‡] Jack Xin, [*]
Jingyi Zhu, [†,‡] and Kenneth M. Golden [†,‡]

The enhancement in diffusive transport of passive tracer particles by incompressible, turbulent flow fields is a challenging problem with theoretical and practical importance in many areas of science and engineering, ranging from the transport of mass, heat, and pollutants in geophysical flows to sea ice dynamics and turbulent combustion. The long time, large scale behavior of such systems is equivalent to an enhanced diffusive process with an effective diffusivity tensor D^*. Two different formulations of integral representations for D^* were developed for the case of *time-independent* fluid velocity fields, involving spectral measures of *bounded* self-adjoint operators acting on vector fields and scalar fields, respectively. Here, we extend both of these approaches to the case of *space-time periodic* velocity fields, with possibly chaotic dynamics, providing rigorous integral representations for D^* involving spectral measures of *unbounded* self-adjoint operators. We prove the different formulations are equivalent. Their correspondence follows from a one-to-one isometry between the underlying Hilbert spaces. We also develop a Fourier method for computing D^*, which captures the phenomenon of residual diffusion related to Lagrangian chaos of a model flow. This is reflected in the spectral measure by a concentration of mass near the spectral origin.

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1. Introduction

The long time, large scale motion of diffusing particles or tracers being advected by an incompressible flow field is equivalent to an enhanced diffusive process [96] with an effective diffusivity tensor D^*. Describing the associated transport properties is a challenging problem with a broad range of scientific and engineering applications, such as stellar convection [47, 83, 22, 23, 21], turbulent combustion [3, 17, 95, 101, 79, 103], and solute transport in porous media [14, 15, 100, 41, 48, 51, 49]. Time-dependent flows can have fluid velocity fields with chaotic dynamics, which gives rise to turbulence that greatly enhances the mixing, dispersion, and large scale transport of diffusing scalars. Here we develop a mathematical framework that provides an analytic representation of D^* for such time-dependent, chaotic flows. This representation is given in terms of a Stieltjes integral involving the spectral measure of an *unbounded* self-adjoint operator. We demonstrate that this approach provides an effective method for computing D^* for chaotic flows.

1.1. Advection enhanced diffusion in the climate system

In the climate system [27, 40], turbulence plays a key role in transporting mass, heat, momentum, energy, and salt in geophysical flows [67]. Turbulence enhances the dispersion of atmospheric gases [29] such as ozone [43, 80, 81, 82] and pollutants [26, 11, 88], as well as atmosphere-ocean transfers of carbon dioxide and other climatically important trace gas fluxes [105, 8]. Longitudinal dispersion of passive scalars in oceanic flows can be enhanced by horizontal turbulence due to shearing of tidal currents, wind drift, or waves [104, 50, 19]. Chaotic motion of time-dependent fluid velocity fields cause instabilities in large scale ocean currents, generating geostrophic eddies [33] which dominate the kinetic energy of the ocean [34]. Geostrophic eddies greatly enhance [33] the meridional mixing of heat, carbon and other climatically important tracers, typically more than one order of magnitude greater than the mean flow of the ocean [91]. Eddies also impact heat and salt budgets through lateral fluxes and can extend the area of high biological productivity offshore by both eddy chlorophyll advection and eddy nutrient pumping [24].

In sea ice dynamics, where the ice cover couples the atmosphere to the polar oceans [98], the transport of sea ice can also be enhanced by eddie fluxes and large scale coherent structures in the ocean [99, 54]. In sea ice thermodynamics, the temperature field of the atmosphere is coupled to the temperature field of the ocean through sea ice, which is a composite of pure ice with brine inclusions whose volume fraction and connectedness depend strongly on temperature. Convective brine flow through the porous microstructure can enhance thermal transport through the sea ice layer [55, 102, 52].

Both numerical and observational studies of scalar transport have suggested that tracers are advected over large scales by a fluid velocity field that is different from the mean flow [76]. This suggests that the effective diffusivity tensor D^* should be spatially and possibly also temporally inhomogeneous [76]. The mixing of eddy fluxes is typically non-divergent and unable to affect the evolution of the mean flow [64], and do not alter the tracer moments [39]. In this sense, the mixing is non-dissipative, reversible, and sometimes referred to as stirring [28, 39]. It has been noted in various geophysical contexts [81, 82] that eddy-induced skew-diffusive tracer fluxes directed normal to the tracer gradient [64]] are generally equivalent to *antisymmetric* components in the effective diffusivity tensor D^*, while the *symmetric* part of D^* represents irreversible diffusive effects [84, 89, 39] directed down the tracer gradient. Motivated by these observations, we provide in the ensuing sections analytic representations for both the *symmetric and antisymmetric* components of D^*.

1.2. Mathematical characterization of effective diffusivity

Due to the computational intensity of detailed climate models [40, 98, 70], a coarse resolution is necessary in numerical simulations and *parameterization* is used to help resolve sub-grid processes, such as turbulent entrainment-mixing processes in clouds [53], atmospheric boundary layer turbulence [20], atmosphere-surface exchange over the sea [30] and sea ice [90, 1, 2, 97], and eddies in the ocean [60, 37]. In this way, only the effective or averaged behavior of these sub-grid processes are included in the models. Here, we study the effective behavior of advection enhanced diffusion by time-dependent fluid velocity fields, with possibly chaotic dynamics, which gives rise to such a parameterization, namely, the effective diffusivity tensor D^* of the flow.

In recent decades, a broad range of mathematical techniques have been developed which reduce the analysis of enhanced diffusive transport by complex fluid velocity fields with rapidly varying structures in both space and time, to solving averaged or *homogenized* equations that do not have rapidly varying data, and involve an effective parameter [72, 61, 9, 16, 31, 71, 32, 59, 76, 78, 25, 42, 44, 57, 58, 103]. Motivated by [73], it was shown [61] that the homogenized behavior of the advection-diffusion equation with a random, time-independent, incompressible, mean-zero fluid velocity field, is given by an inhomogeneous diffusion equation involving the symmetric part of an effective diffusivity tensor D^*. Moreover, a rigorous representation of D^* was given in terms of an auxiliary "cell problem" involving a curl-free random field [61]. We stress that the effective diffusivity tensor D^* is not symmetric in general. However, only its symmetric part appears in the homogenized equation for *this* formulation of the effective transport properties of advection enhanced diffusion [61].

The incompressibility condition of the time-independent fluid velocity field was used [4, 5] to transform the cell problem in [61] into the quasi-static limit of Maxwell's equations [46, 38], which describe the transport properties of an electromagnetic wave in a composite material [66]. The analytic continuation method for representing transport in composites [38] provides Stieltjes integral representations for the bulk transport coefficients of composite media, such as electrical conductivity and permittivity, magnetic permeability, and thermal conductivity [66]. This method is based on the spectral theorem [94, 85] and a resolvent formula for, say, the electric field, involving a random self-adjoint operator [38, 69] or matrix [68]. Based on the analytic continuation method [38], the cell problem for the advection diffusion equation was transformed into a resolvent formula involving a *bounded* self-adjoint operator, acting on the Hilbert space of curl-free random vector fields [4, 5]. This, in turn, led to a Stieltjes integral representation for the symmetric part of the effective diffusivity tensor D^*, involving the Péclet number Pe of the flow and a *spectral measure* $\boldsymbol{\mu}$ of the operator [4, 5]. A key feature of the method is that parameter information in Pe is *separated* from the complicated geometry of the time-independent flow, which is encoded in the measure $\boldsymbol{\mu}$. This property led to rigorous bounds [5] for the diagonal components of D^*. Bounds for D^* can also be obtained using variational methods [5, 31, 71, 32].

The mathematical framework developed in [61] was also adapted [76, 62, 57] to the case of a periodic, time-dependent, incompressible fluid velocity field with *non-zero* mean. The velocity field was modeled as a superposition of a large-scale mean flow with small-scale periodically oscillating fluctuations. It was shown [76] that, depending on the strength of the fluctuations relative to the mean flow, the effective diffusivity tensor D^* can be constant or a function of both space and time. When D^* is constant, only its symmetric part appears in the homogenized equation as an enhancement in the diffusivity. However, when D^* is a function of space and time, its antisymmetric part also plays a key role in the homogenized equation. In particular, the symmetric part of D^* appears as an enhancement in the diffusivity, while both the symmetric and antisymmetric parts of D^* contribute to an effective drift in the homogenized equation. The effective drift due to the antisymmetric part is purely sinusoidal, thus divergence-free [76]. This is consistent with what has been observed in geophysical flows in the climate system, as discussed in the final paragraph of Section 1.1.

Based on [14], the cell problem discussed in [76] was transformed into a resolvent formula involving a self-adjoint operator, acting on the Sobolev space [63, 35] of spatially periodic scalar fields, which is also a Hilbert space. In the case where the mean flow and periodic fluctuations are time-independent, the self-adjoint operator is compact [14], hence *bounded* [92]. This led to a discrete Stieltjes integral representation for the antisymmetric part of D^*, involving the Péclet number of the steady flow and a spectral measure of the operator.

The incompressibility of the fluid velocity field was a central property of the mathematical frameworks described above. However, in [62], these results were extended to weakly compressible, anelastic, stratified, time-independent, fluid velocity fields. Homogenization of the convection-reaction-diffusion equation with compressible velocity field was treated in [74].

1.3. Summary of Results

Here, we generalize both of the approaches described in [4, 5] and [76] to the case of an incompressible, periodic, *time-dependent* fluid velocity field, allowing for chaotic dynamics. In particular, for each approach, we provide Stieltjes integral representations for both the symmetric and antisymmetric parts of the effective diffusivity tensor D^*, involving a spectral measure of a self-adjoint operator. In this time-dependent setting, the underlying operator becomes *unbounded*. The spectral theory of unbounded operators is more subtle and technically challenging than the spectral theory of bounded operators, since the domain of an unbounded operator and its adjoint plays a central role in the spectral characterization of the operator. Neglecting such important mathematical details, the Stieltjes integral representation for D^* given in [4, 5] was extended to the time-dependent setting in [6]. Here, we provide a mathematically rigorous formulation of Stieltjes integral representations for D^* in the time-dependent, unbounded operator setting. We prove that the two approaches described in [4, 5] and [76] are equivalent in this setting, and that their correspondence follows from a one-to-one isometry between the underlying Hilbert spaces. We also establish a direct correspondence between the effective parameter problem for D^* and the effective parameter problem arising in the analytic continuation method for composite media.

Analytical calculations of the spectral measure underlying the effective diffusivity tensor D^* have been obtained only for a handful of simple models of periodic fluid velocity fields to date such as shear flows. We develop a Fourier method for the computation of D^*. In particular, we compute the effective properties for the following space-time periodic flow in two spatial dimensions, with $\boldsymbol{x} = (x, y)$,

$$\boldsymbol{u}(t, \boldsymbol{x}) = (\cos y, \cos x) + \theta \cos t \, (\sin y, \sin x), \quad \theta \in (0, 1]. \tag{1}$$

The steady part $(\cos y, \cos x)$ of the flow is subject to a time-periodic perturbation that gives rise to a transition to Lagrangian chaos for $\theta > 0$ [16, 106]. In the advection dominated regime, we shall compare our computations of the effective diffusivity for the steady $\theta = 0$ and dynamic $\theta = 1$ settings.

The rest of the paper is organized as follows. In Section 2 the theory of homogenization for the advection-diffusion equation for space-time periodic flows is reviewed. Novel Stieltjes integral representations for the effective diffusivity tensor are also obtained for a large class of fluid velocity fields, involving a spectral measure of an *unbounded* self-adjoint operator. In Section 3 we use Fourier methods to transform the eigenvalue problem for the operator into an infinite set of algebraic equations. This provides a rigorous mathematical foundation for the computation of effective diffusivities for such flows. This framework is employed in Section 4 to compute the discrete component of the spectral measure associated with the fluid velocity field in (1) for time-independent flow $\theta = 0$ and time-dependent flow $\theta = 1$. Our computations highlight that the behavior of the measure near the spectral origin governs the behavior of the effective diffusivity in the advection dominated regime of small molecular diffusion. In particular, we demonstrate that for $\theta = 0$ there is a *spectral gap* in the measure near a limit point at the spectral origin, giving rise to the known vanishing asymptotic behavior of 2D cell flows [31, 71]. However in the time dependent setting, a strong concentration of measure mass near the spectral origin gives rise to the phenomenon of residual diffusivity in the limit of vanishing molecular diffusion.

Technical background information and proofs of the key results of the paper are differed to the appendices. The spectral theory of unbounded self-adjoint operators in Hilbert space is reviewed in Appendix A and Appendix B. Two mathematical formulations of the effective parameter problem for advection enhanced diffusion are presented in Appendix C, leading to novel integral representations for the symmetric and antisymmetric components of the effective diffusivity tensor. In Appendix D we use powerful methods of functional analysis to prove that the two approaches are equivalent, which follows from a one-to-one isometry between the associated Hilbert spaces. In Appendix D.1 we derive an explicit formula for the discrete component of the spectral measure, which is employed in our numerical computations.

2. Effective transport by advective-diffusion

The density ϕ of a cloud of passive tracer particles diffusing along with molecular diffusivity ε and being advected by an incompressible velocity field $\boldsymbol{u}$ satisfies the advection-diffusion equation

$$\partial_t \phi(t,\boldsymbol{x}) = \boldsymbol{u}(t,\boldsymbol{x})\cdot\boldsymbol{\nabla}\phi(t,\boldsymbol{x}) + \varepsilon\Delta\phi(t,\boldsymbol{x}), \quad \phi(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}), \tag{2}$$

for $t>0$ and $\boldsymbol{x}\in\mathbb{R}^d$. Here, the initial density $\phi_0(\boldsymbol{x})$ and the fluid velocity field $\boldsymbol{u}$ are assumed given, and $\boldsymbol{u}$ satisfies $\boldsymbol{\nabla}\cdot\boldsymbol{u}=0$. In equation (2), the molecular diffusion constant $\varepsilon>0$, d is the spatial dimension of the system, ∂_t denotes partial differentiation with respect to time t, and $\Delta=\boldsymbol{\nabla}\cdot\boldsymbol{\nabla}=\nabla^2$ is the Laplacian. Moreover, $\boldsymbol{\psi}\cdot\boldsymbol{\varphi}=\boldsymbol{\psi}^T\overline{\boldsymbol{\varphi}}$, $\boldsymbol{\psi}^T$ denotes transposition of the vector $\boldsymbol{\psi}$, and $\overline{\boldsymbol{\varphi}}$ denotes component-wise complex conjugation, with $\boldsymbol{\psi}\cdot\boldsymbol{\psi}=|\boldsymbol{\psi}|^2$. Later, we will extensively use this form of the dot product over complex fields, with built in complex conjugation. However, we stress that all quantities considered in this section are *real-valued.*

We consider enhanced diffusive transport by a periodic fluid velocity field and non-dimensionalize equation (2) as follows. Let ℓ and T be typical length and time scales associated with the problem of interest. Mapping to the non-dimensional variables $t\mapsto t/T$ and $\boldsymbol{x}\mapsto\boldsymbol{x}/\ell$, one finds that ϕ satisfies the advection-diffusion equation in (2) with a non-dimensional molecular diffusivity $\varepsilon\mapsto T\,\varepsilon/\ell^2$ and velocity field $\boldsymbol{u}\mapsto T\,\boldsymbol{u}/\ell$. There are several different non-dimensionalizations possible for the advection-diffusion equation. A detailed discussion of various non-dimensionalizations involving the Strouhal number, the Péclet number, and the periodic Péclet number is given in [62, 57]. Here, we focus on the long time, large scale transport characteristics of equation (2) as a function of ε. To this end, we simply take T to be the temporal periodicity of the velocity field $\boldsymbol{u}$ and assume that the spatial periodicity of $\boldsymbol{u}$ is ℓ in all spatial dimensions, i.e.,

$$\boldsymbol{u}(t+T,\boldsymbol{x}) = \boldsymbol{u}(t,\boldsymbol{x}), \qquad \boldsymbol{u}(t,\boldsymbol{x}+\ell\,\boldsymbol{e}_j) = \boldsymbol{u}(t,\boldsymbol{x}), \quad j=1,\ldots,d, \tag{3}$$

where $\boldsymbol{e}_j$ is a standard basis vector in the jth direction.

2.1. Mean-zero flow

In this section we will discuss the effective transport properties of advection enhanced diffusion, as described by the advection diffusion equation in (2). We will assume in this section that the fluid velocity field is mean-zero. The effects of a large-scale mean flow will be discussed in Section 2.2.

The long time, large scale dispersion of diffusing tracer particles being advected by an incompressible fluid velocity field is equivalent to an enhanced diffusive process [96] with an effective diffusivity tensor D^*. In recent decades, methods of homogenization theory [61, 31, 71, 57] have been used to provide an explicit representation for D^*. In particular, these methods have demonstrated that the averaged or *homogenized* behavior of the advection-diffusion equation in (2), with space-time periodic velocity field $\boldsymbol{u}$, is determined by a diffusion equation involving an averaged scalar density $\bar{\phi}$ and an effective diffusivity tensor D^* [57]

$$\partial_t \bar{\phi}(t,\boldsymbol{x}) = \boldsymbol{\nabla}\cdot[\mathsf{D}^* \boldsymbol{\nabla}\bar{\phi}(t,\boldsymbol{x})], \quad \bar{\phi}(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}). \tag{4}$$

Equation (4) follows from the assumption that the initial tracer density ϕ_0 varies slowly relative to the variations of the fluid velocity field $\boldsymbol{u}$ [61, 32, 57]. This information is incorporated into equation (2) by introducing a small dimensionless parameter $\delta \ll 1$ and writing [61, 32, 57]

$$\phi(0,\boldsymbol{x}) = \phi_0(\delta \boldsymbol{x}). \tag{5}$$

Anticipating that ϕ will have diffusive dynamics as $t \to \infty$, space and time are rescaled according to the standard diffusive relation

$$\boldsymbol{\xi} = \boldsymbol{x}/\delta, \quad \tau = t/\delta^{\gamma}, \qquad \gamma = 2. \tag{6}$$

The rescaled form of equation (2) is given by [57]

$$\partial_t \phi^\delta(t,\boldsymbol{x}) = \delta^{-1}\boldsymbol{u}(t/\delta^2,\boldsymbol{x}/\delta)\cdot\boldsymbol{\nabla}\phi^\delta(t,\boldsymbol{x}) + \varepsilon\Delta\phi^\delta(t,\boldsymbol{x}), \quad \phi^\delta(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}), \tag{7}$$

where we have denoted $\phi^\delta(t,\boldsymbol{x}) = \phi(t/\delta^2,\boldsymbol{x}/\delta)$. The convergence of ϕ^δ to $\bar{\phi}$ can be rigorously established in the following sense [57]

$$\lim_{\delta\to 0} \sup_{0\le t\le t_0} \sup_{\boldsymbol{x}\in\mathbb{R}^d} |\phi^\delta(t,\boldsymbol{x}) - \bar{\phi}(t,\boldsymbol{x})| = 0, \tag{8}$$

for every finite $t_0 > 0$, provided that ϕ_0 and $\boldsymbol{u}$ obey some mild smoothness and boundedness conditions, and that $\boldsymbol{u}$ is *mean-zero*. We will discuss the consequences of a fluid velocity field $\boldsymbol{u}$ with a large scale mean flow in Section 2.2.

An explicit representation of the effective diffusivity tensor D^* is given in terms of the (unique) mean zero, space-time periodic solution χ_j of the following *cell problem* [16, 57],

$$\partial_\tau \chi_j(\tau, \boldsymbol{\xi}) - \varepsilon \Delta_\xi \chi_j(\tau, \boldsymbol{\xi}) - \boldsymbol{u}(\tau, \boldsymbol{\xi}) \cdot \boldsymbol{\nabla}_\xi \chi_j(\tau, \boldsymbol{\xi}) = u_j(\tau, \boldsymbol{\xi}), \tag{9}$$

where the subscript ξ in Δ_ξ and $\boldsymbol{\nabla}_\xi$ indicates that differentiation is with respect to the fast variable $\boldsymbol{\xi}$ defined in equation (6). Specifically, the components D^*_{jk}, $j, k = 1, \ldots, d$, of the matrix D^* are given by [61, 31, 71, 57]

$$\mathsf{D}^*_{jk} = \varepsilon \delta_{jk} + \langle u_j \chi_k \rangle, \tag{10}$$

where δ_{jk} is the Kronecker delta and u_j is the jth component of the vector $\boldsymbol{u}$. The averaging $\langle\cdot\rangle$ in (10) is with respect to the fast variables defined in equation (6). The averaging is over the bounded sets $\mathcal{T} \subset \mathbb{R}$ and $\mathcal{V} \subset \mathbb{R}^d$, with $\tau \in \mathcal{T}$ and $\boldsymbol{\xi} \in \mathcal{V}$, which define the space-time period cell ($(d{+}1)$–torus) $\mathcal{T} \times \mathcal{V}$. For example, in Section 4 we compute D^* for a fluid velocity field $\boldsymbol{u}$ with temporal periodicity $\mathcal{T} = [0, 2\pi]$ and spatial periodicity $\mathcal{V} = [0, 2\pi]^d$. In the case of a time-dependent fluid velocity field, $\langle\cdot\rangle$ denotes space-time averaging over $\mathcal{T} \times \mathcal{V}$. In the special case of a time-independent fluid velocity field, the function χ_j is time-independent and satisfies equation (9) with $\partial_\tau \chi_j \equiv 0$, and $\langle\cdot\rangle$ in (10) denotes spatial averaging over $\mathcal{V}$ [31, 71, 57]. Since the remainder of the analysis involves only the fast variables, for notational simplicity, we will drop the subscripts ξ shown in equation (9).

In general, the effective diffusivity tensor D^* has a symmetric S^* and antisymmetric A^* part defined by

$$\mathsf{D}^* = \mathsf{S}^* + \mathsf{A}^*, \qquad \mathsf{S}^* = \frac{1}{2}\left(\mathsf{D}^* + [\mathsf{D}^*]^T\right), \quad \mathsf{A}^* = \frac{1}{2}\left(\mathsf{D}^* - [\mathsf{D}^*]^T\right), \tag{11}$$

where $[\mathsf{D}^*]^T$ denotes transposition of the matrix D^*. Denote by S^*_{jk} and A^*_{jk}, $j, k = 1, \ldots, d$, the components of S^* and A^* in (11). In Appendix C.1 we show that they have the following functional representations [76]

$$\mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle \chi_j, \chi_k \rangle_{1,2}), \quad \mathsf{A}^*_{jk} = \langle A\chi_j, \chi_k \rangle_{1,2}, \quad A = (-\Delta)^{-1}(\partial_\tau - \boldsymbol{u}\cdot\boldsymbol{\nabla}), \tag{12}$$

where $\langle f, h\rangle_{1,2} = \langle \boldsymbol{\nabla} f \boldsymbol{\cdot} \boldsymbol{\nabla} h\rangle$ is a Sobolev-type *sesquilinear* inner-product [63] and the operator $(-\Delta)^{-1}$ is based on convolution with respect to the Green's function for the Laplacian Δ [92]. Since the function χ_j is *real-valued* we have $\langle \chi_j, \chi_k\rangle_{1,2} = \langle \chi_k, \chi_j\rangle_{1,2}$, which implies that S^* is a symmetric matrix. The function $A\chi_j$ is also real-valued. We establish in Appendix C.1 that the operator A is skew-symmetric on a suitable Hilbert space, which implies that $\mathsf{A}^*_{kj} = \langle A\chi_k, \chi_j\rangle_{1,2} = -\langle \chi_k, A\chi_j\rangle_{1,2} = -\langle A\chi_j, \chi_k\rangle_{1,2} = -\mathsf{A}^*_{jk}$ which, in turn, implies that A^* is an antisymmetric matrix, hence $\mathsf{A}^*_{kk} = \langle A\chi_k, \chi_k\rangle_{1,2} = 0$.

Applying the linear operator $(-\Delta)^{-1}$ to both sides of the cell problem in equation (9) yields the following resolvent formula for χ_j

$$\chi_j = (\varepsilon + A)^{-1} g_j, \qquad g_j = (-\Delta)^{-1} u_j. \tag{13}$$

From equations (12) and (13) we have the following functional formulas for S^*_{jk} and A^*_{jk} involving the antisymmetric operator A

$$\begin{aligned} \mathsf{S}^*_{jk} &= \varepsilon\left(\delta_{jk} + \langle (\varepsilon + A)^{-1} g_j, (\varepsilon + A)^{-1} g_k\rangle_{1,2}\right), \\ \mathsf{A}^*_{jk} &= \langle A(\varepsilon + A)^{-1} g_j, (\varepsilon + A)^{-1} g_k\rangle_{1,2}. \end{aligned} \tag{14}$$

Since A is a skew-symmetric operator, it can be written as $A = \imath M$ where M is a symmetric operator. We demonstrate in Appendix C.1 that M is *self-adjoint* on an appropriate, dense subset of a Hilbert space.

The spectral theorem for self-adjoint operators states that there is a one-to-one correspondence between the self-adjoint operator M and a family of self-adjoint projection operators $\{Q(\lambda)\}_{\lambda\in\Sigma}$ — the resolution of the identity — that satisfies $\lim_{\lambda\to\inf\Sigma} Q(\lambda) = 0$ and $\lim_{\lambda\to\sup\Sigma} Q(\lambda) = I$ [94]. Here, Σ is the *spectrum* of the operator M, while 0 and I denote the null and identity operators. Define the *complex valued* function $\mu_{jk}(\lambda) = \langle Q(\lambda) g_j, g_k\rangle_{1,2}$, $j, k = 1, \ldots, d$, where $g_j = (-\Delta)^{-1} u_j$ is defined in (13). The real, $\mathrm{Re}\,\mu_{jk}(\lambda)$, and imaginary, $\mathrm{Im}\,\mu_{jk}(\lambda)$, parts of the function $\mu_{jk}(\lambda)$ are strictly increasing and of bounded variation, and therefore have Stieltjes measures $\mathrm{Re}\,\mu_{jk}$ and $\mathrm{Im}\,\mu_{jk}$ associated with them [94]. The function $\mu_{kk}(\lambda)$ is positive hence μ_{kk} is a positive measure, while $\mathrm{Re}\,\mu_{jk}$ and $\mathrm{Im}\,\mu_{jk}$ are signed measures. Given certain regularity conditions on the components u_j of the fluid velocity field $\boldsymbol{u}$, the functional formulas for S^*_{jk} and A^*_{jk} in (14) have the following Radon–Stieltjes integral representations, for all $0 < \varepsilon < \infty$ (see Appendix C.1 for details)

$$\mathsf{S}^*_{jk} = \varepsilon\left(\delta_{jk} + \int_{-\infty}^{\infty} \frac{\mathrm{dRe}\ \mu_{jk}(\lambda)}{\varepsilon^2 + \lambda^2}\right), \qquad \mathsf{A}^*_{jk} = -\int_{-\infty}^{\infty} \frac{\lambda\, \mathrm{dIm}\ \mu_{jk}(\lambda)}{\varepsilon^2 + \lambda^2}. \tag{15}$$

A key feature of equation (15) is that parameter information in ε is *separated* from the complicated geometry and dynamics of the time-dependent flow, which are encoded in the spectral measure μ_{jk}. This important property of the integrals in (15) follows from the non-dimensionalization of the advection-diffusion equation discussed in the paragraph leading to equation (3), yielding a spectral measure μ_{jk} that is *independent* of the molecular diffusivity ε. An alternate formulation of the effective parameter problem for advection-diffusion by time-dependent flows was discussed in [6], which used a different non-dimensionalization, yielding a Stieltjes integral representation for S^*_{kk} involving the Péclet number Pe of the flow and a spectral measure that depends on the Strouhal number. However, as pointed out in [18], the Strouhal number dependence of the measure led to an implicit dependence of the spectral measure on Pe. This restricts the utility of the integral representations, such as rigorous bounds [7, 38] which depend explicitly on Pe but also implicitly on Pe through the moments of the measure. Our formulation has no such restrictions.

2.2. The effect of large scale mean flow

The periodic homogenization theorem summarized by equations (3)–(10), as well as its many variations [9, 72, 13, 15, 61, 5, 76, 77, 78, 62, 57], depend on the detailed nature of the fluid velocity field $\boldsymbol{u}$. They also depend on the temporal scaling used [15, 76, 57], i.e., what value of γ is used in equation (6). However, the mathematical structure of the cell problem in (9) and the functional form of D^* shown in equation (10) remain unchanged for the space-time periodic setting. One of the key goals of the present work is to develop a rigorous mathematical framework that provides the Stieltjes integral representations for effective diffusivity tensor D^* shown in (15), for space-time periodic $\boldsymbol{u}$. Due to the time-dependence of the fluid velocity field, one must employ the spectral theory of *unbounded* self-adjoint operators, which is much more subtle and challenging than the spectral theory of bounded operators. We will demonstrate that this mathematical framework depends only on the structure of the cell problem in (9) and the presence of an inner-product in the functional form of D^* in (10). In particular, the theoretical development is insensitive to the detailed nature of $\boldsymbol{u}$ and depends only on its boundedness properties (See Corollary **??** for details). Consequently, our results given here apply in many of the well studied systems and will likely apply to many of the homogenization results of the future.

In order to illustrate the rich behaviors that can arise in the effective diffusivity tensor D^* for more general velocity fields and alternate temporal scalings, we now briefly discuss some key variations of the theory described above. When the fluid velocity field is mean-zero, as discussed above, then equation (8) holds and the effective diffusivity tensor D^* defined in (10) is constant [57]. Consequently, only the symmetric part of D^* plays a role in the effective transport equation shown in (4). Now consider a more general fluid velocity field

$$\boldsymbol{u}(t,\boldsymbol{x}) = \delta^{\alpha}\boldsymbol{u}_0(\delta^{\gamma}t, \delta\boldsymbol{x}) + \boldsymbol{u}_1(t,\boldsymbol{x}), \qquad \alpha = 1, \quad \gamma = 2, \tag{16}$$

which is the superposition of a *weak*, large-scale mean flow $\delta\boldsymbol{u}_0(\delta^2 t, \delta\boldsymbol{x})$ that varies on large spatial and slow time scales, with a mean-zero periodic flow $\boldsymbol{u}_1(t,\boldsymbol{x})$ that rapidly fluctuates in space and time [57]. If $\boldsymbol{u}_0(t,\boldsymbol{x})$ is smooth and bounded, the homogenization theorem for purely periodic velocity fields discussed above can be rigorously extended to the present setting and the effective transport equation in (4) is replaced by [57]

$$\partial_t\bar{\phi}(t,\boldsymbol{x}) = \boldsymbol{u}_0(t,\boldsymbol{x})\boldsymbol{\cdot}\boldsymbol{\nabla}\bar{\phi}(t,\boldsymbol{x}) + \boldsymbol{\nabla}\boldsymbol{\cdot}[\mathsf{D}^*\boldsymbol{\nabla}\bar{\phi}(t,\boldsymbol{x})], \quad \bar{\phi}(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}), \tag{17}$$

which includes an advective enhancement in transport by the large-scale mean flow $\boldsymbol{u}_0$ [57]. In this case, the effective diffusivity tensor D^* is completely independent of the mean flow $\boldsymbol{u}_0$, and is determined by the same formula in equation (10) and the same cell problem in (9) with $\boldsymbol{u} \to \boldsymbol{u}_1$ [57]. Consequently, D^* is again constant and only the symmetric part of D^* plays a role in the effective transport equation shown in (17).

This problem was studied in [76] for scalings in (16) different than $\alpha = 1$ and $\gamma = 2$. The parameter α determines the strength of the mean flow $\boldsymbol{u}_0$ relative to the small scale periodic fluctuations $\boldsymbol{u}_1$. When the mean flow is weak compared to the fluctuations, to leading order, D^* is constant and independent of the mean flow, which only determines the transport velocity on large length and long time scales. Consequently, only the symmetric part of D^* plays a role in the effective transport equation, which is similar to the effective transport equation in (17) [76]. Regardless of the values of α and γ, in the weak mean flow regime, the components D^*_{jk} of the effective diffusivity tensor are given by a formula analogous to equation (10) and the structure of the cell problem is analogous to equation (9). There are three distinct behaviors that arise as the values of α and γ vary, and the function χ_j in (9) can be time-dependent or time-independent ($\partial_\tau\chi_j \equiv 0$) [76].

As we discussed in Section 1, the constancy of the effective diffusivity tensor D^* is not consistent with measurements and numerical simulations of passive tracer transport in the ocean and atmosphere. However, when the fluid velocity is active on both the slow and fast time scales, with $\gamma = 1$, and the mean flow is equal in strength or stronger than the periodic fluctuations, then D^* *is a function of space and time* [76]. Consequently, in the effective transport equation, the *antisymmetric* part of D^* contributes to a purely rotational (divergence-free) enhancement in advective transport, while the symmetric part of D^* contributes to an enhancement in advective and diffusive transport [76]. This is consistent with observations and direct numerical simulations of geophysical flows in the climate system.

In Appendix C.1 we provide a mathematically rigorous framework that leads to the Stieltjes integral representations in (15), for both the symmetric and antisymmetric parts of the effective diffusivity tensor D^*. This formulation is based on the spectral theorem for *unbounded* self-adjoint operators in Hilbert space, which is based on an axiomatic construction of Hilbert space. Consequently, the integral representations for D^* depend only on abstract properties of the underlying self-adjoint operator and, in particular, on boundedness properties shared by a large class of fluid velocity fields $\boldsymbol{u}$, including all those discussed in this section. In Appendix A, we review the spectral theory of unbounded operators. In Appendix C we give two natural Hilbert space formulations of the effective parameter problem for D^* which lead to its promised integral representations. In Appendix D we use powerful methods of functional analysis to prove that the two formulations are equivalent and discuss the theoretical and computational advantages of each approach.

3. Fourier methods

In this section we discuss how Fourier methods can be employed to convert the eigenvalue problem $A\varphi_l = \imath\lambda_l\varphi_l$, $\lambda_l \in \mathbb{R}$, $l \in \mathbb{N}$, into an infinite set of algebraic equations involving the Fourier coefficients of the eigenfunction φ_l. This will be used in Section 4 to compute the discrete component of the spectral measure μ_{jk} underlying the integral representations for the effective diffusivity tensor D^* shown in equations (15) and (A-71). For notational simplicity, we set $\boldsymbol{u} \to -\boldsymbol{u}$ and $(\tau, \boldsymbol{\xi}) \to (t, \boldsymbol{x})$ and write the operator A in (12) as $A = (-\Delta)^{-1}(\partial_t + \boldsymbol{u}\cdot\boldsymbol{\nabla})$. We will focus on the fluid velocity field $\boldsymbol{u}$ in equation (1), although the methods discussed here extend to a large class of fluid velocity fields, namely, those expressible as a finite sum of Fourier modes.

In Appendix C.1 we showed that $M = -\imath A$ is a self-adjoint operator on an appropriate, dense subset of a Hilbert space $\mathscr{H} \subset L^2(\mathcal{T} \times \mathcal{V})$ given in equation (A-17). Since the space-time periodic velocity field $\boldsymbol{u}$ in (1) is mean-zero, we require that the elements of $\mathscr{H}$ are also mean-zero, i.e., $f \in \mathscr{H}$ implies that $\langle f \rangle = 0$. In Appendix D.1 we discuss how a decomposition of the Hilbert space $\mathscr{H}$ provides a natural decomposition of the spectral measure μ_{jk} into its discrete and continuous components, as shown in equation (A-66). Here, we demonstrate how this mathematical framework can be used to compute the discrete component of μ_{jk}, hence the discrete component of the Stieltjes integral representation for the effective diffusivity tensor D^* shown in equation (A-71).

Toward this goal, rewrite the eigenvalue problem $A\varphi_l = \imath\lambda_l\varphi_l$ as

$$(\partial_t + \boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})\varphi_l = -\imath\lambda_l\Delta\varphi_l. \tag{18}$$

For spatial dimension $d = 2$, the set of functions $\{\exp[\imath(\ell t{+}mx{+}ny)] \mid \ell, m, n \in \mathbb{Z}\}$ are complete in $L^2(\mathcal{T} \times \mathcal{V})$, hence the Hilbert space $\mathscr{H}$. Consequently, the eigenfunction $\varphi_l \in \mathscr{H}$ can be represented as

$$\varphi_l(t, \boldsymbol{x}) = \sum_{\ell,m,n\in\mathbb{Z}} a^l_{\ell,m,n}\, \mathrm{e}^{\imath(\ell t+mx+ny)}, \qquad a^l_{\ell,m,n} = \left\langle \varphi_l, \mathrm{e}^{\imath(\ell t+mx+ny)} \right\rangle, \tag{19}$$

where we have denoted $\boldsymbol{x} = (x, y)$ and $\langle\cdot,\cdot\rangle$ is the $L^2(\mathcal{T} \times \mathcal{V})$–inner-product. Write the fluid velocity field in (1) as $\boldsymbol{u}(t, \boldsymbol{x}) = \boldsymbol{u}_1(\boldsymbol{x}) + \theta \cos t\; \boldsymbol{u}_2(\boldsymbol{x})$, where $\boldsymbol{u}_1 = (\cos y, \cos x)$ and $\boldsymbol{u}_2 = (\sin y, \sin x)$, and write

$$\begin{aligned} \cos t\, \sin y &= \frac{1}{4\imath}(\mathrm{e}^{\imath t} + \mathrm{e}^{-\imath t})(\mathrm{e}^{\imath y} - \mathrm{e}^{-\imath y}) \\ &= \frac{1}{4\imath}(\mathrm{e}^{\imath(t+y)} - \mathrm{e}^{\imath(t-y)} + \mathrm{e}^{\imath(-t+y)} - \mathrm{e}^{\imath(-t-y)}) \end{aligned} \tag{20}$$

Consequently, substituting the formula in (19) for φ_l into equation (18) yields

$$\begin{aligned} 0 = \sum_{\ell,m,n} \mathrm{e}^{\imath(\ell t+mx+ny)} \Big[& \imath\ell\, a^l_{\ell,m,n} - \imath\lambda_l(m^2+n^2)a^l_{\ell,m,n} \\ & +\imath m\big((a^l_{\ell,m,n-1} + a^l_{\ell,m,n+1})/2 \\ & \quad +\theta(a^l_{\ell-1,m,n-1} - a^l_{\ell-1,m,n+1} + a^l_{\ell+1,m,n-1} - a^l_{\ell+1,m,n+1})/(4\imath)\big) \\ & +\imath n\big((a^l_{\ell,m-1,n} + a^l_{\ell,m+1,n})/2 \\ & \quad +\theta(a^l_{\ell-1,m-1,n} - a^l_{\ell-1,m+1,n} + a^l_{\ell+1,m-1,n} - a^l_{\ell+1,m+1,n})/(4\imath)\big)\Big]. \end{aligned} \tag{21}$$

Since $\{\mathrm{e}^{\imath(\ell t+mx+ny)}\}$ is a complete orthonormal set in the underlying Hilbert space, $\sum_{\ell,m,n} \mathrm{e}^{\imath(\ell t+mx+ny)} c_{\ell,m,n} = 0$ only if $c_{\ell,m,n} = 0$ for all $\ell, m, n \in \mathbb{Z}$ [94].

Consequently, equation (21) yields the following infinite set of algebraic equations for the Fourier coefficients $a^l_{\ell,m,n}$ of the eigenfunction φ_l

(22)

$$\begin{aligned}
&\imath\ell\, a^l_{\ell,m,n} + \frac{\imath}{2}[m(a^l_{\ell,m,n-1} + a^l_{\ell,m,n+1}) + n(a^l_{\ell,m-1,n} + a^l_{\ell,m+1,n})] \\
&\qquad + \frac{\theta}{4}[m(a^l_{\ell-1,m,n-1} - a^l_{\ell-1,m,n+1}) + n(a^l_{\ell-1,m-1,n} - a^l_{\ell-1,m+1,n}) \\
&\qquad\quad + m(a^l_{\ell+1,m,n-1} - a^l_{\ell+1,m,n+1}) + n(a^l_{\ell+1,m-1,n} - a^l_{\ell+1,m+1,n})] \\
&\quad = \imath\lambda_l(m^2+n^2)a^l_{\ell,m,n}, \qquad \ell, m, n \in \mathbb{Z},
\end{aligned}$$

The solution of equation (22) determines both the eigenvalues λ_l and the Fourier coefficients $a^l_{\ell,m,n}$ of the eigenfunction φ_l. Notice that for $m = n = 0$, equation (22) implies that $\imath\ell\, a^l_{\ell,0,0} = 0$ for all $\ell \in \mathbb{Z}$. This is consistent with the requirement that φ_l is mean-zero in space, which implies that $a^l_{\ell,0,0} = 0$ for all $\ell \in \mathbb{Z}$. This will be used in Section 4 to compute the discrete component of the spectral measure μ_{jk}.

Towards this goal, we now determine the spectral weights $\langle g_j, \varphi_l\rangle_{1,2}$ of the measure μ_{jk} shown in (A-71) in terms of the Fourier coefficients $a^l_{\ell,m,n}$ of φ_l. Recall from equation (1) that $\boldsymbol{u} = (u_1, u_2)$ with

(23)

$$\begin{aligned}
u_1(t,x,y) &= \cos y + \theta \cos t \sin y \\
&= \frac{1}{2}\left(\mathrm{e}^{\imath y} + \mathrm{e}^{-\imath y}\right) + \frac{\theta}{4\imath}\left(\mathrm{e}^{\imath(t+y)} - \mathrm{e}^{\imath(t-y)} + \mathrm{e}^{\imath(-t+y)} - \mathrm{e}^{\imath(-t-y)}\right),
\end{aligned}$$

and $u_2(t,x,y) = u_1(t,y,x)$. This equation, the formula $\langle g_j, \varphi_l\rangle_{1,2} = \langle u_j, \varphi_l\rangle_2$ in equation (A-72), the Fourier expansion of φ_l in (19), and the orthogonality of the set $\{\mathrm{e}^{\imath(\ell t + mx + ny)}\}$, imply that

(24)

$$\begin{aligned}
\langle \varphi_l, g_1\rangle_{1,2} &= \frac{1}{2}\left(a^l_{0,0,1} + a^l_{0,0,-1}\right) + \frac{\theta}{4\imath}\left(a^l_{1,0,1} - a^l_{1,0,-1} + a^l_{-1,0,1} - a^l_{-1,0,-1}\right) \\
\langle \varphi_l, g_2\rangle_{1,2} &= \frac{1}{2}\left(a^l_{0,1,0} + a^l_{0,-1,0}\right) + \frac{\theta}{4\imath}\left(a^l_{1,1,0} - a^l_{1,-1,0} + a^l_{-1,1,0} - a^l_{-1,-1,0}\right)
\end{aligned}$$

with $\langle g_1, \varphi_l\rangle_{1,2} = \overline{\langle \varphi_l, g_1\rangle}_{1,2}$.

When $\theta = 0$ in the velocity field of equation (1) so that $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$ is time-independent, the operator A no longer involves the time derivative and the associated eigenfunction $\varphi_l = \varphi_l(\boldsymbol{x})$ is time-independent as well. In this case, the system of equations in (22) reduces to

$$\frac{m}{2}(a^l_{m,n-1} + a^l_{m,n+1}) + \frac{n}{2}(a^l_{m-1,n} + a^l_{m+1,n}) = \lambda_l(m^2+n^2)a^l_{m,n}, \quad m,n \in \mathbb{Z}, \tag{25}$$

while equation (24) for the reduces to

$$\langle \varphi_l, g_1\rangle_{1,2} = \frac{1}{2}\left(a^l_{0,1} + a^l_{0,-1}\right), \quad \langle \varphi_l, g_2\rangle_{1,2} = \frac{1}{2}\left(a^l_{1,0} + a^l_{-1,0}\right). \tag{26}$$

4. Numerical Results

In Section 3, we used Fourier methods to transform the eigenvalue problem $A\varphi_l = \imath\lambda_l\varphi_l$ for the operator A in (12), involving the fluid velocity field in (1), into an infinite system of algebraic equations for the Fourier coefficients of the eigenfunctions φ_l. In this section, we truncate the infinite system, convert it to a generalized eigenvalue problem, and numerically compute the discrete component of the spectral measure underlying the integral representations for the symmetric S^* and anti-symmetric A^* parts of the effective diffusivity tensor D^*, shown in equations (15) and (A-71).

By restricting the indices, $-M \leq \ell, m, n \leq M$, and imposing the boundary conditions

$$a^l_{\ell,m,n} = 0 \quad \text{if} \quad \max(|\ell|, |m|, |n|) > M, \tag{27}$$

the infinite systems of equations in (22) and (25) become finite sets of equations. Consider the fluid velocity field in (1) with parameter $\theta \in [0,1]$. In the dynamic ($\theta > 0$) and steady ($\theta = 0$) cases, the bijective mappings $\Theta_d(\ell, m, n)$ and $\Theta_s(m,n)$ defined by

$$\begin{aligned} \Theta_d(\ell,m,n) &= (M+m+1) + (M+n)(2M+1) + (M+\ell)(2M+1)^2, \\ \Theta_s(m,n) &= (M+m+1) + (M+n)(2M+1), \end{aligned} \tag{28}$$

map the corresponding finite set of equations to matrix equations. In particular, they become generalized eigenvalue problems

$$\mathsf{B}\boldsymbol{a}_l = \lambda_l \mathsf{C}\boldsymbol{a}_l. \tag{29}$$

Here B is a symmetric matrix and C is a diagonal matrix of size $(2M+1)^3 \times (2M+1)^3$ for the dynamic case and of size $(2M+1)^2 \times (2M+1)^2$ for the steady case. More specifically, B is Hermitian in the dynamic case and is real-symmetric in the steady case, while the matrix C is real-symmetric and diagonal in both cases. Since B and C are symmetric matrices, the generalized eigenvalues λ_l are real-valued and the eigen-vectors $\boldsymbol{a}_l$ – consisting of the Fourier coefficients for φ_l – satisfy the orthogonality condition [75]

$$\boldsymbol{a}_j^\dagger \mathsf{C} \boldsymbol{a}_k = \delta_{jk}. \tag{30}$$

The matrix C is positive semidefinite and diagonal. However, in the steady case, in both of the matrices B and C, the row and column associated with the Fourier coefficient $a^l_{0,0}$ is all zeros, as can be seen from equation (25). Since the eigenfunction φ_l is mean-zero, we have that $a^l_{0,0} = 0$. Therefore, we simply remove this row and column in both B and C so that C becomes strictly positive definite.

In the dynamic case, the entries of the matrix C do not depend on ℓ. Consequently, we have that $\mathsf{C} = \mathrm{diag}(\mathsf{C}_s, \dots, \mathsf{C}_s)$, where we have denoted by C_s the matrix C in the steady case. Therefore, there are $2M+1$ diagonal entries in C with the value zero, corresponding to $m = n = 0$. The entries of the corresponding rows and columns of the matrix B are all zero except for the diagonal entry, which has the value ℓ, as can be seen from equation (22). This implies that $\ell a^l_{\ell,0,0} = 0$ for all $-M \leq \ell \leq M$. Since the eigenfunction φ_l is mean-zero in time *and* space, we have that $a^l_{\ell,0,0} = 0$ for all $-M \leq \ell \leq M$, which is consistent with the above observation. We therefore simply remove the corresponding rows and columns in both B and C so that C becomes strictly positive definite. This method of removing the null space common to both B and C is called *deflation* [75].

Now that the matrix C is strictly positive definite and diagonal, the matrix C^q is well defined for all $q \in \mathbb{R}$, with entries $(\mathsf{C}^q)_{ij} = \mathsf{C}^q_{ii}\delta_{ij}$, where C^q_{ii} is the ith diagonal of the matrix C raised to the power q, and $\mathsf{C}^q\mathsf{C}^{-q} = \mathsf{I}$. Consequently, the generalized eigenvalue problem in equation (29) can be written as the following standard eigenvalue problem

$$\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}\boldsymbol{v}_l = \lambda_l \boldsymbol{v}_l, \qquad \boldsymbol{v}_l = \mathsf{C}^{1/2}\boldsymbol{a}_l. \tag{31}$$

Since B is a symmetric matrix and C is diagonal, the matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$ is also symmetric with real-valued eigenvalues and orthonormal eigenvectors. From the orthogonality relation $\boldsymbol{v}_j^\dagger \boldsymbol{v}_k = \delta_{jk}$ we recover equation (30) via $\boldsymbol{v}_l = \mathsf{C}^{1/2}\boldsymbol{a}_l$ in (31).

In summary, our numerical method is the following. Create the matrices B and C according to equation (22) or (25) and the corresponding bijective mapping in (28). Remove the rows and columns of the matrices B and C corresponding to $\mathsf{C}_{ii} = 0$. Compute the eigenvalues λ_l and eigenvectors $\boldsymbol{v}_l$ of the symmetric matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$. The computed Fourier coefficients of the eigenfunction φ_l are given by $\boldsymbol{a}_l = \mathsf{C}^{-1/2}\boldsymbol{v}_l$. The eigenvalues associated with the discrete component of the spectral measure shown in equation (A-71) are given by λ_l, while the spectral measure weights $\langle g_1, \varphi_l\rangle_{1,2}$ and $\langle g_2, \varphi_l\rangle_{1,2}$ in (A-71) are determined by the vector $\boldsymbol{a}_l$ via equation (24) or (26).

In our computations, we used for the steady case $M = 150$, yielding matrices of size $(2M+1)^2 - 1 = 90,600$, while in the dynamic case we used $M = 20$, yielding matrices of size $(2M+1)^3 - (2M+1) = 68,880$. The eigenvalues and eigenvectors of the symmetric matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$ were computed using the Matlab function *eig()*. The stability of the computations are measured in terms of the condition numbers $\mathcal{K}_l$ of the eigenvalues λ_l, which are the reciprocals of the cosines of the angles between the left and right eigenvectors. Eigenvalue condition numbers close to 1 indicate a stable computation. Our eigenvalue computations are extremely stable with $\max_l |1 - \mathcal{K}_l| \sim 10^{-14}$, which were computed using the Matlab function *condeig()*.

Displayed in Fig. 1 are our computations of the discrete component of the spectral measure $\mathrm{d}\mu_{11}(\lambda) = \sum_l m_{11}(l)\delta_{\lambda_l}(\mathrm{d}\lambda)$ associated with the fluid velocity field $\boldsymbol{u}$ shown in equation (1), for (a) the steady ($\theta = 0$) and (b) the dynamic ($\theta = 1$) settings. Here, the spectral weights $m_{11}(l) = |\langle g_1, \varphi_l\rangle_{1,2}|^2$ are determined by equations (26) and (24), respectively. Consistent with the symmetries of the flows [16], we have $\mu_{11} = \mu_{22}$, while $\mathrm{Re}\,\mu_{12} = 0$ and $\mathrm{Im}\,\mu_{12} = 0$, up to numerical accuracy and finite size effects.

For the 2D steady cell flow in (1) with $\theta = 0$, it is known [31, 71] that $\mathsf{S}^*_{11} \sim \varepsilon^{1/2}$ for $\varepsilon \ll 1$. Our computation of S^*_{11} displayed in Fig. 1(c) is in excellent agreement with this result, with a computed critical exponent of ≈ 0.52 having an error of only 4% relative to its true value 0.5. Reducing M from 150 to 100 changes the value of the critical exponent by less than 0.0015, indicating that the value of $M = 150$ is sufficiently large. In this steady setting, the underlying operator $(-\Delta)^{-1}[\boldsymbol{u}_1\cdot\boldsymbol{\nabla}]$ is compact [14] and therefore has bounded, discrete spectrum away from the spectral origin, with a limit point at $\lambda = 0$ [92]. The limit point behavior of the measure μ_{11} can be seen in the rightmost panel of Fig. 1(a). The decay of S^*_{11} for vanishing ε is due to the magnitude of the measure masses $m_{11}(l) \lesssim 10^{-30}$ for $|\lambda_l| \ll 1$, with a significant *spectral gap* near the limit point. The rigorous result [31, 71] $\mathsf{S}^*_{11} \sim \varepsilon^{1/2}$ as $\varepsilon \to 0$ reveals that the spectrum of the operator

$(-\Delta)^{-1}[\boldsymbol{u}_1\boldsymbol{\cdot}\boldsymbol{\nabla}]$ at $\lambda = 0$ is either continuous or it is discrete with zero mass, otherwise S^*_{11} would diverge as $\varepsilon \to 0$.

In contrast, as shown in Fig. 1(b), the spectral measure μ_{11} associated with the time-dependent fluid velocity field in (1), with $\theta = 1$, has significant values of $m_{11}(l)$ near the spectral origin, with $m_{11}(l) \gtrsim 10^{-10}$ more than *20 orders* of magnitude greater than that of the steady flow. A limit point behavior in the measure μ_{11} near $\lambda = 0$ can be seen in the rightmost panel of Fig. 1(b). It is interesting to note that the support of the measure *supp* μ_{11} satisfies *supp* $\mu_{11} \subset [-M, M]$ for all values of M investigated. Due to the significant mass of the measure near the spectral origin and its uniform nature, as shown in the center panel of Fig. 1(b), the effective diffusivity has an $O(1)$ behavior, $\mathsf{S}^*_{11} \sim 1$ for $\varepsilon \ll 1$, as shown in Fig. 1(d). This is consistent with numerical computations of S^*_{11} using alternate methods [16]. This $O(1)$ behavior of S^*_{11} has been attributed to the Lagrangian chaotic behavior of the flow [16, 106].

Appendix A. Spectral theory of unbounded self-adjoint operators in Hilbert space

The theory of *unbounded* operators in Hilbert space was developed largely by John von Neumann and Marshall H. Stone. It is considerably more technical and challenging than the theory of bounded operators, as unbounded operators do not form an algebra, nor even a linear space, because each one is defined on its own domain. In this section, we review the spectral theory for such operators and, in particular, the celebrated *spectral theorem* for self-adjoint operators [85, 94].

An operator is not determined unless its domain is known. Let Φ_1 and Φ_2 be operators acting on a Hilbert space $\mathscr{H}$ with domains $D(\Phi_1)$ and $D(\Phi_2)$, respectively, $D(\Phi_i) \subset \mathscr{H}$, $i = 1, 2$. They are said to be *identical*, in symbols $\Phi_1 \equiv \Phi_2$, if and only if $D(\Phi_1) = D(\Phi_2)$ and $\Phi_1 f = \Phi_2 f$ for every f of their common domain. They are said to be *equal* in the set $\mathscr{S}$, in symbols $\Phi_1 = \Phi_2$, if and only if $\mathscr{S} \subseteq D(\Phi_1) \cap D(\Phi_2)$ and $\Phi_1 f = \Phi_2 f$ for every $f \in \mathscr{S}$. The operator Φ_2 is said to be an *extension* (*proper extension*) of the operator Φ_1 if $D(\Phi_1) \subseteq D(\Phi_2)$ ($D(\Phi_1) \subset D(\Phi_2)$) and the operators Φ_2 and Φ_1 are equal in $D(\Phi_1)$ [94].

Consider the sesquilinear inner-product $\langle\cdot,\cdot\rangle$ associated with $\mathscr{H}$ satisfying $\langle a\psi, b\varphi\rangle = a\,\overline{b}\,\langle\psi,\varphi\rangle$ and $\langle\psi,\varphi\rangle = \overline{\langle\varphi,\psi\rangle}$ for all $\psi,\varphi \in \mathscr{H}$ and $a, b \in \mathbb{C}$, where $\overline{z}$ denotes complex conjugation of $z \in \mathbb{C}$. The $\mathscr{H}$–inner-product induces a norm $\|\cdot\|$ defined by $\|\psi\| = \langle\psi,\psi\rangle^{1/2}$. A linear operator Φ is said to be *closed* if for every pair of sequences $\{f_n\}$ and $\{\Phi f_n\}$ (with

$f_n \in D(\Phi)$) that converge in the norm $\|\cdot\|$ to the limits f and h, then $f \in D(\Phi)$ and $\Phi f = h$ [94]. The (Hilbert space) adjoint Φ^* of Φ is defined by $\langle \Phi\psi, \varphi\rangle = \langle \psi, \Phi^*\varphi\rangle$ for every $\psi \in D(\Phi)$ and $\varphi \in D(\Phi^*)$. The adjoint Φ^* of Φ is uniquely determined when the domain $D(\Phi)$ *determines* $\mathscr{H}$, i.e., the smallest closed linear manifold containing $D(\Phi)$ is the Hilbert space $\mathscr{H}$ [94]. In this case, $D(\Phi) \subseteq D(\Phi^*)$ and Φ^* is a closed linear operator [94]. The operator Φ is said to be *symmetric* if $\Phi = \Phi^*$. The operator Φ is said to be *self-adjoint* if $\Phi \equiv \Phi^*$. A symmetric operator is said to be *maximal* if it has no proper symmetric extension. A self-adjoint operator is a maximal symmetric operator [94].

The operator Φ is said to be *bounded* (in operator norm) if $\|\Phi\| = \sup_{\{\psi\in\mathscr{H}\,:\,\|\psi\|=1\}} \|\Phi\psi\| < \infty$. A bounded linear symmetric operator is self-adjoint if and only if its domain is $\mathscr{H}$ [94]. Conversely, the Hellinger–Toeplitz theorem states that, if the operator Φ satisfies $\langle \Phi\psi, \varphi\rangle = \langle \psi, \Phi\varphi\rangle$ for *every* $\psi, \varphi \in \mathscr{H}$, then Φ is bounded on $\mathscr{H}$ [85, 94]. This indicates that, if Φ is an *unbounded* symmetric operator on $\mathscr{H}$, then it is self-adjoint only on a *proper subset* of $\mathscr{H}$ that is dense in $\mathscr{H}$.

The spectrum Σ of a self-adjoint operator Φ on a Hilbert space $\mathscr{H}$ is real-valued [85, 94]. If Φ is also bounded, then its spectral radius equal to its operator norm $\|\Phi\|$ [85], i.e.,

$$\Sigma \subseteq [-\|\Phi\|, \|\Phi\|\,]. \tag{A-2}$$

If Φ is instead unbounded, its spectrum Σ can be an unbounded subset of, or can even coincide with the set of real numbers $\mathbb{R}$ [94].

We now summarize the spectral theorem for self-adjoint operators (see Theorems 5.9 and 6.1 in [94]). Let Φ be a fixed self-adjoint operator with spectrum $\Sigma \subseteq \mathbb{R}$ and domain $D(\Phi)$ that is dense in $\mathscr{H}$. If Φ is bounded then we simply take $D(\Phi) \equiv \mathscr{H}$. The spectral theorem states that there is a one-to-one correspondence between the self-adjoint operator Φ and a family of self-adjoint projection operators $\{Q(\lambda)\}_{\lambda\in\Sigma}$ — the resolution of the identity — that satisfies [94]

$$\lim_{\lambda\to\inf\Sigma} Q(\lambda) = 0, \qquad \lim_{\lambda\to\sup\Sigma} Q(\lambda) = I, \tag{A-3}$$

where 0 and I denote the null and identity operators on $\mathscr{H}$, respectively. Furthermore, the *complex-valued* function of the spectral variable λ defined by $\mu_{\psi\varphi}(\lambda) = \langle Q(\lambda)\psi, \varphi\,\rangle$ has real, $\operatorname{Re}\mu_{\psi\varphi}(\lambda)$, and imaginary, $\operatorname{Im}\mu_{\psi\varphi}(\lambda)$, parts that are strictly increasing for $\lambda \in \Sigma$ and of bounded variation for all

$\psi, \varphi \in D(\Phi)$ [94], where

$$\text{(A-4)} \qquad \begin{aligned} \operatorname{Re} \mu_{\psi\varphi}(\lambda) &= \frac{1}{2}\left(\mu_{\psi\varphi}(\lambda) + \overline{\mu}_{\psi\varphi}(\lambda)\right), \\ \operatorname{Im} \mu_{\psi\varphi}(\lambda) &= \frac{1}{2\,\imath}\left(\mu_{\psi\varphi}(\lambda) - \overline{\mu}_{\psi\varphi}(\lambda)\right), \end{aligned}$$

$\imath = \sqrt{-1}$, and $\lambda \in \Sigma$.

By the sesquilinearity of the inner-product and the fact that the projection operator $Q(\lambda)$ is self-adjoint, the function $\mu_{\psi\varphi}(\lambda)$ satisfies $\mu_{\varphi\psi}(\lambda) = \overline{\mu}_{\psi\varphi}(\lambda)$. Moreover, the function $\mu_{\psi\psi}(\lambda)$ is real-valued and positive, $\mu_{\psi\psi}(\lambda) = \langle Q(\lambda)\psi, \psi\rangle = \langle Q(\lambda)\psi, Q(\lambda)\psi\rangle = \|Q(\lambda)\psi\|^2 \geq 0$, hence $\operatorname{Re} \mu_{\psi\psi}(\lambda) = \mu_{\psi\psi}(\lambda)$ and $\operatorname{Im} \mu_{\psi\psi}(\lambda) = 0$. With each of these strictly increasing functions of bounded variation, we associate Stieltjes measures [93, 94, 36]

$$\text{(A-5)} \qquad \begin{aligned} \mathrm{d}\mu_{\psi\varphi}(\lambda) &= \mathrm{d}\langle Q(\lambda)\psi, \varphi\rangle, & \mathrm{dRe}\, \mu_{\psi\varphi}(\lambda) &= \mathrm{dRe}\, \langle Q(\lambda)\psi, \varphi\rangle, \\ \mathrm{d}\mu_{\psi\psi}(\lambda) &= \mathrm{d}\|Q(\lambda)\psi\|^2, & \mathrm{dIm}\, \mu_{\psi\varphi}(\lambda) &= \mathrm{dIm}\, \langle Q(\lambda)\psi, \varphi\rangle, \end{aligned}$$

which we will denote by $\mu_{\psi\psi}$, $\mu_{\psi\varphi}$, $\operatorname{Re} \mu_{\psi\varphi}$, and $\operatorname{Im} \mu_{\psi\varphi}$. We stress that $\mu_{\psi\psi}$ is a positive measure, $\mu_{\psi\varphi}$ is a complex measure, while $\operatorname{Re} \mu_{\psi\varphi}$ and $\operatorname{Im} \mu_{\psi\varphi}$ are signed measures [93, 94].

The spectral theorem also provides an operational calculus in Hilbert space which yields powerful integral representations involving the Stieltjes measures shown in equation (A-5). A summary of the relevant details are as follows. Let $F(\lambda)$ and $G(\lambda)$ be arbitrary complex-valued functions and denote by $\mathscr{D}(F)$ the set of all $\psi \in D(\Phi)$ such that $F \in L^2(\mu_{\psi\psi})$, i.e., F is square integrable on the set Σ with respect to the *positive* measure $\mu_{\psi\psi}$, and similarly define $\mathscr{D}(G)$. Then $\mathscr{D}(F)$ and $\mathscr{D}(G)$ are linear manifolds and there exist linear operators denoted by $F(\Phi)$ and $G(\Phi)$ with domains $\mathscr{D}(F)$ and $\mathscr{D}(G)$, respectively, which are defined in terms of the following Radon–Stieltjes integrals [94]

$$\text{(A-6)} \qquad \begin{aligned} \langle F(\Phi)\psi, \varphi\rangle &= \int_{-\infty}^{\infty} F(\lambda)\, \mathrm{d}\mu_{\psi\varphi}(\lambda), && \forall\, \psi \in \mathscr{D}(F),\ \varphi \in D(\Phi), \\ \langle F(\Phi)\psi, G(\Phi)\varphi\rangle &= \int_{-\infty}^{\infty} F(\lambda)\overline{G}(\lambda)\, \mathrm{d}\mu_{\psi\varphi}(\lambda), && \forall\, \psi \in \mathscr{D}(F),\ \varphi \in \mathscr{D}(G), \end{aligned}$$

where the integration in (A-6) is over the spectrum Σ of Φ [85, 94].

The mass $\mu^0_{\psi\varphi} = \int_{-\infty}^{\infty} \mathrm{d}\mu_{\psi\varphi}(\lambda)$ of the Stieltjes measure $\mu_{\psi\varphi}$ satisfies [94] $\mu^0_{\psi\varphi} = \lim_{\lambda\to\sup\Sigma} \mu_{\psi\varphi}(\lambda) - \lim_{\lambda\to\inf\Sigma} \mu_{\psi\varphi}(\lambda)$. Consequently, equation (A-3) and the Cauchy-Schwartz inequality yield

$$\mu^0_{\psi\varphi} = \int_{-\infty}^{\infty} \mathrm{d}\langle Q(\lambda)\psi, \varphi\,\rangle = \langle\psi, \varphi\rangle, \qquad |\mu^0_{\psi\varphi}| \le \|\psi\|\,\|\varphi\| < \infty. \tag{A-7}$$

Equation (A-7) demonstrates that the measures in (A-5) are *finite measures*, i.e., they have bounded mass [94].

Equation (A-6) can be generalized, holding with suitable notational changes, for *maximal normal operators* [94]. Such a normal operator $\mathbf{N}$ with domain $D(\mathbf{N})$ dense in $\mathscr{H}$ commutes with its adjoint $\mathbf{N}^*$, i.e., $\mathbf{N}\mathbf{N}^* = \mathbf{N}^*\mathbf{N}$, and can be decomposed as $\mathbf{N} = \Phi_1 + \imath\Phi_2$, where Φ_1 and Φ_2 are self-adjoint and commute. The spectrum of the normal operator $\mathbf{N}$ is a (possibly unbounded) subset of $\mathbb{C}$ [94]. A special case of a normal operator is a *skew-adjoint* operator satisfying $\mathbf{N}^* = -\mathbf{N}$. It can be decomposed as $\mathbf{N} = \imath\Phi_2$ and since Φ_2 is self-adjoint having purely real spectrum, the skew-adjoint operator $\mathbf{N} = \imath\Phi_2$ has purely imaginary spectrum [94]. Consequently, given such a maximal skew-adjoint operator, one can focus attention on the self-adjoint operator $\Phi_2 = -\imath\mathbf{N}$ without having to resort to the more notationally complicated spectral theory of normal operators.

The signed measures $\mathrm{Re}\,\mu_{\psi\varphi}$ and $\mathrm{Im}\,\mu_{\psi\varphi}$ shown in equation (A-5) arise naturally when considering a maximal skew-adjoint operator $\mathbf{N} = \imath\Phi$, where Φ is self-adjoint. This can be illustrated by considering some special cases. Consider the functional $\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle$ involving *real-valued* Hilbert space members $F(\mathbf{N})\psi$ and $G(\mathbf{N})\varphi$, so that $\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle = \langle G(\mathbf{N})\varphi, F(\mathbf{N})\psi\rangle \in \mathbb{R}$ and, in particular,

$$\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle = \frac{1}{2}(\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle + \langle G(\mathbf{N})\varphi, F(\mathbf{N})\psi\rangle). \tag{A-8}$$

Now consider the special cases $F(\mathbf{N}) = G(\mathbf{N})$ and $F(\mathbf{N}) = \mathbf{N}G(\mathbf{N})$, i.e., $F(\imath\lambda) = G(\imath\lambda)$ and $F(\imath\lambda) = \imath\lambda G(\imath\lambda)$ in equation (A-6), respectively. It follows from equations (A-6) and (A-8), the identities $\mathrm{Re}\,z = (z + \overline{z})/2$ and $\mathrm{Im}\,z = (z - \overline{z})/(2\imath)$, and the linearity properties [94] of Stieltjes-Radon integrals with respect to the functions $\mu_{\psi\varphi}(\lambda)$ and $\overline{\mu}_{\psi\varphi}(\lambda)$ that

$$\begin{aligned} \langle G(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle &= \int_{-\infty}^{\infty} |G(\imath\lambda)|^2\, \mathrm{d}\mathrm{Re}\,\mu_{\psi\varphi}(\lambda), \\ \langle \mathbf{N}G(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle &= -\int_{-\infty}^{\infty} \lambda\, |G(\imath\lambda)|^2\, \mathrm{d}\mathrm{Im}\,\mu_{\psi\varphi}(\lambda). \end{aligned} \tag{A-9}$$

An important property of a self-adjoint operator Φ which will be used later is that its domain $D(\Phi)$ comprises those and only those elements $\psi \in$

$\mathscr{H}$ such that the Stieltjes integral $\int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\mu_{\psi\psi}(\lambda)$ is convergent. When $\psi \in D(\Phi)$ the element $\Phi\psi$ is determined by the relations [94]

$$\langle \Phi\psi, \varphi \rangle = \int_{-\infty}^{\infty} \lambda \, \mathrm{d}\mu_{\psi\varphi}(\lambda), \qquad \|\Phi\psi\|^2 = \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\mu_{\psi\psi}(\lambda), \tag{A-10}$$

where φ is an arbitrary element in $D(\Phi)$ [94]. In fact, this determines the one-to-one correspondence between the self-adjoint operator Φ and its resolution of the identity $Q(\lambda)$ [94].

Appendix B. Time derivative as a maximal normal operator

A key example of an unbounded operator is the time derivative ∂_t acting on the space $L^2(\mathcal{T})$ of Lebesgue measurable functions that are also square integrable on the interval $\mathcal{T} = [0, T]$, say. The unboundedness of ∂_t as an operator on $L^2(\mathcal{T})$ can be understood by considering the orthonormal set of functions $\{\varphi_n\} \subset L^2(\mathcal{T})$ defined by

$$\varphi_n(t) = \beta \sin(n\pi t/T), \quad \beta = \sqrt{2/T}, \qquad \langle \varphi_n, \varphi_m \rangle_2 = \delta_{nm}, \tag{A-11}$$

where $n, m \in \mathbb{N}$ and $\langle \cdot, \cdot \rangle_2$ denotes the sesquilinear $L^2(\mathcal{T})$–inner-product. It follows from $\partial_t \varphi_n = (n\pi\beta/T)\cos(n\pi t/T)$ and $\|\partial_t \varphi_n\|^2 = (n\pi/T)^2$, that the norm of the members of the set $\{\partial_t \varphi_n\}$ grows arbitrarily large as $n \to \infty$. This clearly demonstrates the unboundedness of the operator ∂_t with domain $L^2(\mathcal{T})$.

When one also imposes periodic or Dirichlet boundary conditions, simple integration by parts demonstrates that the operator ∂_t is *skew-symmetric* on $L^2(\mathcal{T})$ so that $-\imath\partial_t$ is symmetric with respect to the sesquilinear inner-product $\langle \cdot, \cdot \rangle_2$. We now identify an everywhere dense subset of $L^2(\mathcal{T})$ on which $-\imath\partial_t$ is a bounded linear self-adjoint operator [85, 94]. Consider the class $\mathscr{A}_\mathcal{T}$ of all functions $\psi \in L^2(\mathcal{T})$ such that $\psi(t)$ is *absolutely continuous* [86] on the interval $\mathcal{T}$ and has a derivative $\psi'(t)$ belonging to $L^2(\mathcal{T})$, i.e., [94, 86]

$$\mathscr{A}_\mathcal{T} = \left\{ \psi \in L^2(\mathcal{T}) \;\middle|\; \psi(t) = c + \int_0^t g(s) ds, \quad g \in L^2(\mathcal{T}) \right\}, \tag{A-12}$$

where the constant c and function $g(s)$ are arbitrary. Now, consider the set $\tilde{\mathscr{A}}_\mathcal{T}$ of all functions $\psi \in \mathscr{A}_\mathcal{T}$ that satisfy the periodic boundary condition $\psi(0) = \psi(T)$, i.e. functions ψ satisfying the properties of equation (A-12) with c arbitrary and $\int_0^T g(s) ds = 0$. In order to help clarify the ideas that were discussed in Appendix A in terms of an abstract Hilbert space $\mathscr{H}$, we also consider the set $\hat{\mathscr{A}_T}$ of all functions $\psi \in \mathscr{A}_T$ that satisfy the Dirichlet boundary condition $\psi(0) = \psi(T) = 0$, i.e. functions ψ satisfying the properties of equation (A-12) with $c = 0$ and $\int_0^T g(s)ds = 0$. More concisely,

$$\text{(A-13)} \qquad \begin{aligned} \tilde{\mathscr{A}_T} &= \{\psi \in \mathscr{A}_T \mid \psi(0) = \psi(T)\}, \\ \hat{\mathscr{A}_T} &= \{\psi \in \mathscr{A}_T \mid \psi(0) = \psi(T) = 0\}. \end{aligned}$$

These function spaces satisfy $\hat{\mathscr{A}_T} \subset \tilde{\mathscr{A}_T} \subset \mathscr{A}_T$ and are each everywhere dense in $L^2(\mathcal{T})$ [94]. Let the operators B, $\tilde{B}$, and $\hat{B}$ be identified as $-\imath\partial_t$ with domains $\mathscr{A}_T$, $\tilde{\mathscr{A}_T}$, and $\hat{\mathscr{A}_T}$, respectively. Then, $\hat{B}$ is a closed linear symmetric operator with the adjoint $\hat{B}^* \equiv B$, and the operator $\tilde{B}$ is a *self-adjoint* extension of $\hat{B}$ [94]. In symbols, this means that $\tilde{B} = \tilde{B}^*$ on $\tilde{\mathscr{A}_T}$ and $D(\tilde{B}) = D(\tilde{B}^*) = \tilde{\mathscr{A}_T}$, i.e., $\tilde{B} \equiv \tilde{B}^*$ on $\tilde{\mathscr{A}_T}$. This establishes that the operator $-\imath\partial_t$ with domain $\tilde{\mathscr{A}_T}$ is self-adjoint, hence ∂_t is a maximal skew-symmetric (normal) operator on $\tilde{\mathscr{A}_T}$. The operator $\imath\partial_t$ on $\tilde{\mathscr{A}_T}$ has a simple point spectrum, consisting of eigenvalues $\lambda = 2n\pi/T$, $n \in \mathbb{Z}$, with corresponding eigenfunctions $\exp(\imath\, 2n\pi t/T)$ [94].

Appendix C. Hilbert spaces, resolvents, and integral representations of the effective diffusivity

In this section we provide a spectral theory of effective diffusivities for space-time periodic flows. In particular, two different approaches to the effective parameter problem for advection-diffusion were proposed in [76] and [4, 5] for *time-independent* flows. We generalize these results to the setting of *time-dependent*, chaotic flows. Specifically, we formulate rigorous mathematical frameworks for each approach which provide Stieltjes integral representations for both the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* for space-time periodic flows, involving a spectral measure of an *unbounded* self-adjoint operator. In Appendix C.1 we generalize the approach proposed in [76], while in Appendix C.2 we generalize the approach proposed in [4, 5]. In Appendix D we establish that the two approaches are equivalent, using the one-to-one correspondence between a self-adjoint operator and its resolution of the identity [94], discussed in the paragraph containing equation (A-10).

C.1. Scalar fields and effective diffusivity

In this section we provide an abstract Hilbert space formulation of the effective parameter problem for advection enhanced diffusion by a space-time periodic fluid velocity field $\boldsymbol{u}(t,\boldsymbol{x})$. To fix ideas, consider the following sets $\mathcal{T}=[0,T]$ and $\mathcal{V}=\otimes_{j=1}^{d}[0,\ell]$ which define the space-time period cell $\mathcal{T}\times\mathcal{V}$ for $\boldsymbol{u}(t,\boldsymbol{x})$. Now consider the Hilbert spaces $L^2(\mathcal{T})$ and $L^2(\mathcal{V})$ of Lebesgue measurable scalar functions over the complex field $\mathbb{C}$ that are also square integrable on $\mathcal{T}$ and $\mathcal{V}$, respectively [36]. Define the associated Hilbert spaces $\mathscr{H}_{\mathcal{T}}$ and $\mathscr{H}_{\mathcal{V}}$ of mean-zero periodic functions

$$\text{(A-14)}\qquad \begin{aligned}\mathscr{H}_{\mathcal{T}}&=\big\{\psi\in L^2(\mathcal{T})\,|\,\psi(t)=\psi(t+T),\ \langle\psi\rangle_{\mathcal{T}}=0\big\},\\ \mathscr{H}_{\mathcal{V}}&=\big\{\psi\in L^2(\mathcal{V})\,|\,\psi(\boldsymbol{x})=\psi(\boldsymbol{x}+\ell\boldsymbol{e}_j),\ \langle\psi\rangle_{\mathcal{V}}=0\big\},\end{aligned}$$

for all $j=1,\dots,d$, where the $\boldsymbol{e}_j$ are standard basis vectors. Here, $\langle\cdot\rangle_{\mathcal{T}}$ and $\langle\cdot\rangle_{\mathcal{V}}$ denote temporal average over $\mathcal{T}$ and spatial average over $\mathcal{V}$, respectively. Associated with these averaging operations are sesquilinear inner-products, $\langle\cdot,\cdot\rangle_{\mathcal{T}}$ and $\langle\cdot,\cdot\rangle_{\mathcal{V}}$, which induce norms, $\|\cdot\|_{\mathcal{T}}$ and $\|\cdot\|_{\mathcal{V}}$ [36]. Denote by $\langle\cdot\rangle$ space-time averaging over $\mathcal{T}\times\mathcal{V}$. Now define the Hilbert space $\mathscr{H}_{\mathcal{T}\mathcal{V}}=\mathscr{H}_{\mathcal{T}}\otimes\mathscr{H}_{\mathcal{V}}$ with sesquilinear inner-product $\langle\cdot,\cdot\rangle$ given by $\langle\psi,\varphi\rangle=\langle\psi\,\overline{\varphi}\rangle$, with $\langle\varphi,\psi\rangle=\overline{\langle\psi,\varphi\rangle}$. The $\mathscr{H}_{\mathcal{T}\mathcal{V}}$–inner-product induces a norm $\|\cdot\|$ given by $\|\psi\|=\langle\psi,\psi\rangle^{1/2}$ [36].

In equation (A-13) we defined the space $\tilde{\mathscr{A}}_{\mathcal{T}}$ of absolutely continuous $\mathcal{T}$–periodic functions with time derivatives belonging to $L^2(\mathcal{T})$. We now define its mean-zero subspace

$$\text{(A-15)}\qquad \tilde{\mathscr{A}}_{\mathcal{T}}^{0}=\tilde{\mathscr{A}}_{\mathcal{T}}\cap\mathscr{H}_{\mathcal{T}}=\{\psi\in\tilde{\mathscr{A}}_{\mathcal{T}}\,|\,\langle\psi\rangle_{\mathcal{T}}=0\},$$

which is *not* a Hilbert space but is instead an everywhere dense subset of the Hilbert space $\mathscr{H}_{\mathcal{T}}$ [94]. To treat spatial dependence, we define the Sobolev space $\mathscr{H}_{\mathcal{V}}^{1,2}$ which is itself a Hilbert space [14, 35, 63],

$$\text{(A-16)}\qquad \mathscr{H}_{\mathcal{V}}^{1,2}=\big\{\psi\in\mathscr{H}_{\mathcal{V}}\ |\ \|\boldsymbol{\nabla}\psi\|_{\mathcal{V}}<\infty\big\}.$$

The $\mathscr{H}_{\mathcal{V}}^{1,2}$–norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$ is induced by the $\mathscr{H}_{\mathcal{V}}^{1,2}$–inner-product: $\|\boldsymbol{\nabla}\psi\|_{\mathcal{V}}=\langle\boldsymbol{\nabla}\psi\boldsymbol{\cdot}\boldsymbol{\nabla}\psi\rangle_{\mathcal{V}}^{1/2}$. The Sobolev space $\mathscr{H}_{\mathcal{V}}^{1,2}$ is the closure in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$ of the space $C^2(\mathcal{V})$ of all twice continuously differentiable periodic functions in $\mathscr{H}_{\mathcal{V}}$, and all the elements of $\mathscr{H}_{\mathcal{V}}^{1,2}$ are those elements of $\mathscr{H}_{\mathcal{V}}$ which have square integrable gradients on the set $\mathcal{V}$ [14]. Functions in $\mathscr{H}_{\mathcal{V}}^{1,2}$ need not be differentiable in the classical sense. Instead, $f\in\mathscr{H}_{\mathcal{V}}^{1,2}$ has derivatives $\partial f/\partial x_j\in L^2(\mathcal{V})$ defined by $\partial f/\partial x_j=\lim_{n\to\infty}\partial f_n/\partial x_j$, where $f_n\in C^2(\mathcal{V})$ are Cauchy in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$ and converge to f in $L^2(\mathcal{V})$ [63].

Finally, consider the Hilbert space $\mathscr{H}$ and its everywhere dense subset $\mathscr{F}$ defined by

$$\text{(A-17)} \qquad \begin{aligned} \mathscr{H} &= \{\psi \in \mathscr{H}_T \otimes \mathscr{H}_{\mathcal{V}}^{1,2} \mid \langle\psi\rangle = 0\}, \\ \mathscr{F} &= \{\psi \in \tilde{\mathscr{A}}_T^0 \otimes \mathscr{H}_{\mathcal{V}}^{1,2} \mid \langle\psi\rangle = 0\}. \end{aligned}$$

We stress that $\tilde{\mathscr{A}}_T^0$ and $\mathscr{F}$ are *not* a complete Hilbert spaces. Instead, they are everywhere dense subsets of the complete Hilbert spaces $\mathscr{H}_T$ and $\mathscr{H}$, respectively. Recalling that $\boldsymbol{\psi}\cdot\boldsymbol{\varphi} = \boldsymbol{\psi}^T\overline{\boldsymbol{\varphi}}$, the sesquilinear $\mathscr{H}$–inner-product is given by $\langle\psi,\varphi\rangle_{1,2} = \langle\boldsymbol{\nabla}\psi\cdot\boldsymbol{\nabla}\varphi\rangle$ with associated norm $\|\cdot\|_{1,2}$ given by $\|\psi\|_{1,2} = \langle|\boldsymbol{\nabla}\psi|^2\rangle^{1/2}$. In the case of a time-independent fluid velocity field $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$ we set $\mathscr{H} \equiv \mathscr{F} \equiv \mathscr{H}_{\mathcal{V}}^{1,2}$.

We now use properties of the Hilbert space $\mathscr{H}$ to obtain functional formulas for the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* defined in equations (10) and (11), involving the solution χ_j of the cell problem in equation (9) and a maximal skew-symmetric operator A on $\mathscr{F}$. We then derive from the cell problem a resolvent formula for χ_j involving the operator A. The spectral theorem discussed in Appendix A then yields Stieltjes integral representations for S^* and A^*, which are established in Theorem 1 below.

Applying the linear operator $(-\Delta)^{-1}$ to both sides of the cell problem in equation (9) yields, with appropriate notational changes,

$$\text{(A-18)} \qquad (-\Delta)^{-1}u_j = (\varepsilon + A)\chi_j, \qquad A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla}).$$

We will discuss the key properties of the operators $(-\Delta)^{-1}$ and A in more detail below. Now write the functional $\langle u_j\chi_k\rangle$ in equation (10) as [76]

$$\text{(A-19)} \quad \langle[(-\Delta)(-\Delta)^{-1}u_j]\,\chi_k\rangle = \langle\boldsymbol{\nabla}(-\Delta)^{-1}u_j\cdot\boldsymbol{\nabla}\chi_k\rangle = \langle(-\Delta)^{-1}u_j, \chi_k\rangle_{1,2}.$$

This calculation will be justified below. Substituting the formula in (A-18) for $(-\Delta)^{-1}u_j$ into equation (A-19) yields equation (12), with appropriate notational changes, which provides functional formulas for the components S^*_{jk} and A^*_{jk}, $j,k = 1,\ldots,d$, of S^* and A^*. Equation (A-18) leads to the resolvent formula shown in (13). From equations (12) and (13) we have the functional formulas for S^*_{jk} and A^*_{jk} shown in equation (14) involving the resolvent of the operator A. The following theorem establishes the Stieltjes integral representations in (15) for these functional formulas of S^*_{jk} and A^*_{jk}.

Theorem 1 *The operator $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$ is a maximal (skew-symmetric) normal operator on the function space $\mathscr{F}$, hence $M = -\imath A$*

is self-adjoint on $\mathscr{F}$. Let $Q(\lambda)$ be the resolution of the identity in one-to-one correspondence with M. Define the complex valued function $\mu_{jk}(\lambda) = \langle Q(\lambda)g_j, g_k\rangle_{1,2}$, $j,k = 1,\ldots,d$, where $g_j = (-\Delta)^{-1}u_j$. Consider the positive measure μ_{kk} and the signed measures $Re\,\mu_{jk}$ and $Im\,\mu_{jk}$ associated with $\mu_{jk}(\lambda)$, introduced in (A-4)*. Then, for $u_j \in \tilde{\mathscr{A}}_T^0 \otimes (\mathscr{H}_{\mathcal{V}} \cap L^r(\mathcal{V}))$, $2 < r \leq \infty$, $\chi_j \in \mathscr{F}$, and all $0 < \varepsilon < \infty$, the functional formulas for S^*_{jk} and A^*_{jk} in* (14) *have the Radon–Stieltjes integral representations shown in equation* (15).

Before proving Theorem 1, we discuss key properties of the linear operator $(-\Delta)^{-1}$. The Laplacian Δ maps constants to the null element 0 and periodic functions to periodic functions. In (9) the operator Δ is acting on a periodic function. Hence, in the present context, the domain of the operator $(-\Delta)^{-1}$ is mean-zero periodic functions. The operator is $(-\Delta)^{-1}$ based on convolution with respect to the Green's function for the Laplacian, i.e., $(-\Delta)^{-1}f(\boldsymbol{x}) = \int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})\, f(\boldsymbol{y})\mathrm{d}\boldsymbol{y}$. The Green's function G is determined by the sum of a fundamental solution for the Laplacian that vanishes on the boundary $\partial\mathcal{V}$ of the spatial region $\mathcal{V}$ and a harmonic function with, in the present context, periodic boundary conditions on $\mathcal{V}$ [35, 63, 92]. It is positive, $G > 0$, symmetric, $G(\boldsymbol{x}-\boldsymbol{y}) = G(\boldsymbol{y}-\boldsymbol{x})$, and integrable [35, 92] (see Proposition 0.5 in [35])

$$\sup_{\boldsymbol{x}\in\mathcal{V}} \int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})\mathrm{d}\boldsymbol{y} \leq C < \infty. \tag{A-20}$$

Consequently, by Young's inequality [35, 36], $(-\Delta)^{-1}$ is a bounded operator on $L^p(\mathcal{V})$ for $1 \leq p \leq \infty$: if $\psi \in L^p(\mathcal{V})$ then $(-\Delta)^{-1}\psi \in L^p(\mathcal{V})$ and

$$\|(-\Delta)^{-1}\psi\|_p \leq C\|\psi\|_p, \quad 1 \leq p \leq \infty, \tag{A-21}$$

where $\|\cdot\|_p$ denotes the $L^p(\mathcal{V})$–norm and C is defined in (A-20). Since $\mathcal{V}$ is bounded, it has finite Lebesgue measure $|\mathcal{V}| < \infty$. Consequently, we have [36] $L^p(\mathcal{V}) \supset L^q(\mathcal{V})$ for all $0 < p < q \leq \infty$ with $\|\psi\|_p \leq \|\psi\|_q\,|\mathcal{V}|^{(1/p)-(1/q)}$. In particular, $L^r(\mathcal{V}) \subset L^2(\mathcal{V})$ for all $2 < r \leq \infty$.

The operator $(-\Delta)^{-1}$ satisfies $\langle(-\Delta)(-\Delta)^{-1}f, h\rangle = \langle f, h\rangle$ in the following weak, distributional sense [63, 35]. Let $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ and let $\{f_n\}$ be a sequence of $\mathcal{V}$–periodic functions with $f_n \in C^2(\mathcal{V})$ that is Cauchy the norm $\langle|\boldsymbol{\nabla}\cdot|^2\rangle_{\mathcal{V}}$ and converges to f in $L^2(\mathcal{V})$. Then, for all $h \in \mathscr{H}^{1,2}_{\mathcal{V}}$ we have (see

Theorem 1 in Section 4.2 of [63])

$$
\text{(A-22)}\qquad \begin{aligned} \langle(-\Delta)(-\Delta)^{-1}f,h\rangle_{\mathcal{V}} &\equiv \lim_{n\to\infty}\left\langle\int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})(-\Delta_y)f_n(\boldsymbol{y})\mathrm{d}\boldsymbol{y},h\right\rangle_{\mathcal{V}}\\ &=\lim_{n\to\infty}\langle f_n,h\rangle_{\mathcal{V}}\\ &=\langle f,h\rangle_{\mathcal{V}}, \end{aligned}
$$

by the continuity of the inner-product [36] and since the boundary terms [63] $\int_{\partial\mathcal{V}}[f_n(\boldsymbol{y})\,\partial G(\boldsymbol{x}-\boldsymbol{y})/\partial\mathbf{n}_y - G(\boldsymbol{x}-\boldsymbol{y})\,\partial f_n(\boldsymbol{y})/\partial\mathbf{n}_y]\,\mathrm{d}S_y$ vanish by periodicity. Here $\mathrm{d}S_y$ denotes the surface measure on $\partial\mathcal{V}$ and $\partial/\partial\mathbf{n}_y$ is the outward normal derivative on the boundary. Consequently, recalling that $\|\cdot\|_{\mathcal{V}}$ is the norm induced by the $\mathscr{H}_{\mathcal{V}}$–inner product $\langle\cdot,\cdot\rangle_{\mathcal{V}}$, integration by parts, the Cauchy-Schwartz inequality, $|\langle f,g\rangle_{\mathcal{V}}|\le\|f\|_{\mathcal{V}}\,\|g\|_{\mathcal{V}}$, and Young's inequality for $p=2$ imply for $\psi\in\mathscr{H}_{\mathcal{V}}$ that

(A-23)
$$
\|\boldsymbol{\nabla}(-\Delta)^{-1}\psi\|_{\mathcal{V}}^2=\langle\boldsymbol{\nabla}(-\Delta)^{-1}\psi\boldsymbol{\cdot}\boldsymbol{\nabla}(-\Delta)^{-1}\psi\rangle_{\mathcal{V}}=|\langle(-\Delta)^{-1}\psi,\psi\rangle_{\mathcal{V}}|\le C\|\psi\|_{\mathcal{V}}^2 .
$$

Therefore, the operator $(-\Delta)^{-1}$ maps $\mathscr{H}_{\mathcal{V}}$ to $\mathscr{H}_{\mathcal{V}}^{1,2}\cup\mathbb{C}$ (since $(-\Delta)^{-1}\psi$ is not necessarily mean-zero for $\psi\in\mathscr{H}_{\mathcal{V}}$).

The following lemma will be used in the proof of Theorem 1 below.

Lemma 2 *Assume that the components u_j, $j=1,\dots,d$ of the fluid velocity field $\boldsymbol{u}$ satisfy $u_j\in\tilde{\mathscr{A}}_{\mathcal{T}}^0\otimes(\mathscr{H}_{\mathcal{V}}\cap L^r(\mathcal{V}))$ for $2<r\le\infty$. Then the operator $(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$ is bounded on $\mathscr{H}$. Moreover, the following are upper bounds for its operator norm $\|(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})\|_{1,2}$*

$$
\text{(A-24)}\qquad \|(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})\|_{1,2}\le\sqrt{C}\sqrt{\sup_{(t,\boldsymbol{x})\in\mathcal{T}\times\mathcal{V}}|\boldsymbol{u}|^2},\quad \textit{when}\quad r=\infty,
$$

$$
\text{(A-25)}\qquad \|(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})\|_{1,2}\le\sqrt{Cd}\,\left[\sum_{j=1}^{d}\langle|u_j|^r\rangle\right]^{1/r}\quad \textit{when}\quad 2<r<\infty,
$$

where the constant C is defined in equation (A-20) *and satisfies* $0<C<\infty$.

Proof of Lemma 2. Denote by $\|\cdot\|_p$ the $L^p(\mathcal{T}\times\mathcal{V})$–norm and let $f\in\mathscr{H}$. A calculation similar to the one given in (A-23) and Hölder's inequality [36], $\|f\,g\|_1\le\|f\|_{p_1}\,\|g\|_{q_1}$, with conjugate exponents satisfying $(1/p_1)+(1/q_1)=1$ and $1\le p_1,\,q_1\le\infty$, yield

$$
\text{(A-26)}\qquad \|(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})f\|_{1,2}^2\le C\|\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}f\|_{p_1}\,\|\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}f\|_{q_1} .
$$

When $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes \mathscr{H}_{\mathcal{V}}$, for each fixed $\boldsymbol{x} \in \mathcal{V}$ the function $u_j(\cdot, \boldsymbol{x})$ is absolutely continuous on the closed bounded set $\mathcal{T}$ and is therefore uniformly bounded [86, 87]. If we also have that $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes L^r(\mathcal{V})$ for $r = \infty$ then $u_j(t, \cdot)$ is also uniformly bounded for each $t \in \mathcal{T}$ and the fluid velocity field satisfies $\sup_{(t,\boldsymbol{x})\in\mathcal{T}\times\mathcal{V}} |\boldsymbol{u}|^2 < \infty$. An example of such a fluid velocity field is in equation (1). In this case, equation (A-26) with $p_1 = q_1 = 2$ and the Cauchy-Schwartz inequality, $|\boldsymbol{u}\cdot\boldsymbol{\nabla} f| \le |\boldsymbol{u}|\,|\boldsymbol{\nabla} f|$, yield the bound given in equation (A-24).

We now use equation (A-26) to establish the bound in (A-25), assuming that $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes (\mathscr{H}_{\mathcal{V}} \cap L^r(\mathcal{V}))$ for $r \ge 2$ and $r \ne \infty$. Let's first focus on the term $\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{p_1}$. Since $1 \le p_1 \le \infty$, the function x^{p_1} is convex for $x > 0$. Therefore, by Jensen's inequality, $(\sum_{i=1}^d |a_i|/d\,)^{p_1} \le \sum_{i=1}^d |a_i|^{p_1}/d$, and the triangle inequality we have

$$\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{p_1}^{p_1} = \left\langle \left| \sum_{i=1}^d u_i\, \partial_i f \right|^{p_1} \right\rangle \le d^{\,p_1-1} \sum_{i=1}^d \langle |u_i|^{p_1}\, |\partial_i f|^{p_1} \rangle\,, \tag{A-27}$$

where ∂_i denotes partial differentiation in the ith direction. Hölder's inequality, implies that $\langle |u_i|^{p_1}\, |\partial_i f|^{p_1} \rangle \le \langle |u_i|^{p_1 p_2} \rangle^{1/p_2} \langle |\partial_i f|^{p_1 q_2} \rangle^{1/q_2}$, with conjugate exponents satisfying $(1/p_2) + (1/q_2) = 1$ and $1 \le p_2, q_2 \le \infty$. Another application of Hölder's inequality finally yields

$$\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{p_1}^{p_1} \le d^{\,p_1-1} \left[\sum_{i=1}^d \langle |u_i|^{p_1 p_2} \rangle^{p_3/p_2} \right]^{1/p_3} \left[\sum_{i=1}^d \langle |\partial_i f|^{p_1 q_2} \rangle^{q_3/q_2} \right]^{1/q_3}, \tag{A-28}$$

with conjugate exponents satisfying $(1/p_3) + (1/q_3) = 1$ and $1 \le p_3, q_3 \le \infty$. An analogous calculation shows that equation (A-28) holds for the term $\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_1}^{q_1}$ in (A-26) with p_1 substituted by q_1 and the p_j and q_j, $j = 2, 3$, substituted by $\hat{p}_j$ and $\hat{q}_j$, say, respectively.

Note that

$$\|f\|_{1,2}^2 = \left\langle |\boldsymbol{\nabla} f|^2 \right\rangle = \sum_{i=1}^d \left\langle |\partial_i f|^2 \right\rangle. \tag{A-29}$$

Therefore, in light of equation (A-28) and its analogue for $\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_1}^{q_1}$, in order to obtain a bound for $\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{p_1} \|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_1}$ in terms of $\|f\|_{1,2}^2$, we require that

$$p_1 q_2 = 2, \quad q_2 = q_3, \qquad q_1 \hat{q}_2 = 2, \quad \hat{q}_2 = \hat{q}_3. \tag{A-30}$$

Since $(1/p_2) + (1/q_2) = 1$ and $(1/p_3) + (1/q_3) = 1$, we have $q_2 = q_3$ if and only if $p_2 = p_3$. Similarly, we have $\hat{q}_2 = \hat{q}_3$ if and only if $\hat{p}_2 = \hat{p}_3$. Note that $(1/p_2) + (1/q_2) = 1$ implies $q_2 = p_2/(p_2 - 1)$. This and $p_1 q_2 = 2$ imply that $p_1 p_2/(p_2 - 1) = 2$. Similarly, we have that $q_1 \hat{p}_2/(\hat{p}_2 - 1) = 2$. Consequently, we have the following bounds on p_2 and $\hat{p}_2$

$$p_2 > 1, \qquad \hat{p}_2 > 1. \tag{A-31}$$

In summary, taking p_1th roots in equation (A-28) and q_1th roots of its analogue for the term $\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_1}^{q_1}$, equations (A-30) and (A-26) yield

$$\|(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})f\|_{1,2}^2 \leq Cd \left[\sum_{j=1}^{d}\langle|u_j|^r\rangle\right]^{1/r} \left[\sum_{j=1}^{d}\langle|u_j|^{\hat{r}}\rangle\right]^{1/\hat{r}} \|f\|_{1,2}^2. \tag{A-32}$$

Here, we used that $d^{1-1/p_1}\, d^{1-1/q_1} = d$, as $(1/p_1) + (1/q_1) = 1$. We also used equation (A-30) to show $1/(p_1 q_3) + 1/(q_1 \hat{q}_3) = 1/2 + 1/2 = 1$, and we have denoted $r = p_1 p_2$ and $\hat{r} = q_1 \hat{p}_2$. If we set $r = \hat{r}$ in equation (A-32), this establishes the bound in (A-25). However, we first need to establish the range of values of r and $\hat{r}$ for which the bound holds. We do so by establishing a relation between the exponents r and $\hat{r}$.

Since $q_1 = p_1/(p_1 - 1)$, we have $\hat{r} = p_1 \hat{p}_2/(p_1 - 1)$. A little algebra shows that $p_1(\hat{r} - \hat{p}_2) = \hat{r}$. Consequently, the strict positivity $\hat{r} > 0$ and the inequality $\hat{p}_2 > 1$ in (A-31) imply that $\hat{r} > \hat{p}_2 > 1$. We may therefore write $p_1 = \hat{r}/(\hat{r} - \hat{p}_2)$. This, $r = p_1 p_2$, and a little algebra shows that the exponents r and $\hat{r}$ are related by $r\hat{r} = \hat{r} p_2 + r \hat{p}_2$. Equation (A-31) then implies that

$$r\hat{r} > \hat{r} + r. \tag{A-33}$$

This inequality can be used to find bounds on quantities such as $\max(r, \hat{r})$ etc. However, recognizing that the values of r and $\hat{r}$ both determine the regularity of just one function, namely the fluid velocity field $\boldsymbol{u}$, we set $r = \hat{r}$ in equation (A-33), which implies that $r > 2$. Since we assumed that $r \neq \infty$. This restricts the value of r to the interval $2 < r < \infty$. This completes the proof of Lemma 2 □.

Proof of Theorem 1. We first establish that the operator $M = -\imath A$ with domain $\mathscr{F}$ is self-adjoint, where $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$. We have already established in Appendix B that the operator $-\imath\partial_t$ with domain $\tilde{\mathscr{A}}_T^0$ is self-adjoint [94]. A bounded linear symmetric operator is self-adjoint on a Hilbert space if and only its domain is the Hilbert space itself (Theorem

2.24 in [94]). By Young's inequality in (A-21), the linear operator $(-\Delta)^{-1}$ is bounded on the Hilbert space $\mathscr{H}_{\mathcal{V}}$. It is also symmetric on $\mathscr{H}_{\mathcal{V}}$ [92, 35]. Consequently, the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}_{\mathcal{V}}$ is self-adjoint. It is also self-adjoint on $\mathscr{H}_{\mathcal{V}}^{1,2}$. Indeed, recalling that $\mathcal{V}=[0,\ell]^d$, the calculation in equation (A-23) and the Poincaré inequality [63], $\|f\|_{\mathcal{V}}\leq 2\ell\|\boldsymbol{\nabla} f\|_{\mathcal{V}}$, show that the operator $(-\Delta)^{-1}$ is bounded on $\mathscr{H}_{\mathcal{V}}^{1,2}$ with operator norm bounded by the quantity $2\ell\sqrt{C}$. It is also symmetric on the Hilbert space $\mathscr{H}_{\mathcal{V}}^{1,2}$, as the following calculation shows, which establishes that the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}_{\mathcal{V}}^{1,2}$ is self-adjoint. Similar to (A-23) for $f,h\in\mathscr{H}_{\mathcal{V}}^{1,2}$ we have

(A-34)
$$\langle\boldsymbol{\nabla}(-\Delta)^{-1}f\boldsymbol{\cdot}\boldsymbol{\nabla} h\rangle_{\mathcal{V}}=\langle f,h\rangle_{\mathcal{V}}=\langle f,(-\Delta)(-\Delta)^{-1}h\rangle_{\mathcal{V}}=\langle\boldsymbol{\nabla} f\boldsymbol{\cdot}\boldsymbol{\nabla}(-\Delta)^{-1}h\rangle_{\mathcal{V}}.$$

By Young's inequality in (A-21), the operators $-\imath\partial_t$ and $(-\Delta)^{-1}$ commute on $\tilde{\mathscr{A}}_T^0\otimes\mathscr{H}_{\mathcal{V}}$ (Theorem 2.27 in [36]). Since $\mathscr{H}_{\mathcal{V}}^{1,2}\subset\mathscr{H}_{\mathcal{V}}$, it follows that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint [94].

In Lemma 2 we established that the linear operator $(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is bounded when $u_j\in\tilde{\mathscr{A}}_T^0\otimes(\mathscr{H}_{\mathcal{V}}\cap L^r(\mathcal{V}))$ for $2<r\leq\infty$. We now establish that it is antisymmetric on the Hilbert space $\mathscr{H}$ which, in turn, establishes that the symmetric operator $-\imath(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is self-adjoint. The antisymmetry of $(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ on $\mathscr{H}$ depends on the incompressibility, $\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{u}=0$, of the fluid velocity field [14]. Consequently, for $f,h\in\mathscr{H}$ we have [14]

(A-35)
$$\begin{aligned}\langle(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})f,h\rangle_{1,2}&=\langle[\boldsymbol{\nabla}((-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla} f)]\boldsymbol{\cdot}\boldsymbol{\nabla} h\rangle\\&=\langle[(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla} f)]\,,h\rangle\\&=\langle[\boldsymbol{\nabla}\boldsymbol{\cdot}(\boldsymbol{u}f)]\,,h\rangle\\&=-\langle f\,,[(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})h]\rangle\\&=-\langle f\,,[(-\Delta)(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})h]\rangle\\&=-\langle\boldsymbol{\nabla} f\boldsymbol{\cdot}[\boldsymbol{\nabla}(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})h]\rangle\\&=-\langle f,(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})h\rangle_{1,2}.\end{aligned}$$

This establishes that the bounded linear operator $-\imath(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$ is symmetric on $\mathscr{H}$, hence self-adjoint on $\mathscr{H}$.

We now summarize our findings. We have established that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint and the operator $-\imath(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is self-adjoint when the components u_j, $j=1,\ldots,d$, of $\boldsymbol{u}$ satisfy $u_j\in\tilde{\mathscr{A}}_T^0\otimes(\mathscr{H}_{\mathcal{V}}\cap L^r(\mathcal{V}))$ for $2<r\leq\infty$. Consequently, the difference of these two operators $M=-\imath A$, with $A=(-\Delta)^{-1}(\partial_t-\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$, with domain

$D(M) = \mathscr{F} \cap \mathscr{H} = \mathscr{F}$ [94] is self-adjoint when $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes (\mathscr{H}_{\mathcal{V}} \cap L^r(\mathcal{V}))$ for $2 < r \leq \infty$. Thus $A = \imath M$ is a maximal (skew-symmetric) normal operator on $\mathscr{F}$ [94].

The complex-valued functions involved in the functional formulas for S_{jk}^* and A_{jk}^* in equation (14) are $F(\lambda) = (\varepsilon + \imath\lambda)^{-1}$ and $G(\lambda) = \imath\lambda(\varepsilon + \imath\lambda)^{-1}$. For all $0 < \varepsilon < \infty$, we have $|F(\lambda)|^2 = (\varepsilon^2 + \lambda^2)^{-1} \leq \varepsilon^{-2} < \infty$ and $|G(\lambda)|^2 = \lambda^2(\varepsilon^2 + \lambda^2)^{-1} \leq 1$. Since μ_{kk} is a finite measure for all $k = 1, \ldots, d$, as shown in equation (A-7), we therefore have that $f \in \mathscr{D}(F)$ and $f \in \mathscr{D}(G)$ for all $f \in D(M)$ when $0 < \varepsilon < \infty$. For $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes \mathscr{H}_{\mathcal{V}}$ the function $g_j = (-\Delta)^{-1} u_j$ satisfies $\langle g_j(\cdot, \boldsymbol{x})\rangle_{\mathcal{T}} = 0$ for all $\boldsymbol{x} \in \mathcal{V}$, hence by the Fubini theorem [36] we have $\langle g_j \rangle = 0$. Moreover, since the operator $(-\Delta)^{-1}$ maps $\mathscr{H}_{\mathcal{V}}$ to $\mathscr{H}_{\mathcal{V}}^{1,2} \cup \mathbb{C}$ we have $g_j = (-\Delta)^{-1} u_j \in \mathscr{F}$. Since $\mathscr{F} \subseteq D(M)$, the conditions of the spectral theorem are satisfied. Consequently, the integral representations in equation (A-6) hold for the functions $F(\lambda)$ and $G(\lambda)$ defined above, involving the complex measure μ_{jk}. The discussion leading to equation (A-9) then establishes the integral representations for S_{jk}^* and A_{jk}^* shown in equation (15).

It is worth noting that from equations (A-7) and (A-23), the mass μ_{jk}^0 of the measure μ_{jk} is given by $\mu_{jk}^0 = \langle g_j, g_k \rangle_{1,2} = \langle (-\Delta)^{-1} u_j, u_k \rangle$. Since $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes \mathscr{H}_{\mathcal{V}}$ and $(-\Delta)^{-1}$ is a self-adjoint operator on $\mathscr{H}_{\mathcal{V}}$, hence $\tilde{\mathscr{A}}_{\mathcal{T}}^0 \otimes \mathscr{H}_{\mathcal{V}}$, the spectral theorem demonstrates that

$$\mu_{jk}^0 = \langle (-\Delta)^{-1} u_j, u_k \rangle = \int \lambda \, \mathrm{d}\langle \tilde{Q}(\lambda) u_j, u_k \rangle. \tag{A-36}$$

In other words, the mass μ_{jk}^0 of the measure μ_{jk} is the first moment of the spectral measure $\mathrm{d}\langle \tilde{Q}(\lambda) u_j, u_k \rangle$ for the negative inverse Laplacian $(-\Delta)^{-1}$, where $\tilde{Q}(\lambda)$ is the resolution of the identity in one-to-one correspondence with the self-adjoint operator $(-\Delta)^{-1}$. This completes the proof of Theorem 1 $\square$.

C.2. Curl-free vector fields and effective diffusivity

In this section we consider an alternate formulation of the effective parameter problem for advection-diffusion that was first proposed [4, 5] for *time-independent* flows. In particular, we provide a rigorous mathematical framework which generalizes this formulation to include space-time periodic fluid velocity fields, with possibly chaotic dynamics. This approach provides analogous formulas to those shown in equations (12)–(15) involving the *curl-free* vector field $\boldsymbol{\nabla}\chi_j$ shown in equation (9), with suitable notational changes, and a maximal (skew-symmetric) normal operator $\mathbf{A}$ acting on a Hilbert space of vector-valued functions.

Towards this goal, recall the Hilbert spaces $\mathscr{H}_\mathcal{T}$ and $\mathscr{H}_\mathcal{V}$ of scalar functions given in equation (A-14) and the function space $\tilde{\mathscr{A}}_\mathcal{T}^0$ given in (A-15). Now define their d-dimensional analogues over the complex field $\mathbb{C}$,

$$\text{(A-37)} \qquad \mathcal{H}_\mathcal{T} = \otimes_{j=1}^d \mathscr{H}_\mathcal{T}, \qquad \mathcal{H}_\mathcal{V} = \otimes_{j=1}^d \mathscr{H}_\mathcal{V}, \qquad \tilde{\mathcal{A}}_\mathcal{T}^0 = \otimes_{j=1}^d \tilde{\mathscr{A}}_\mathcal{T}^0.$$

By the Helmholtz theorem [56, 12, 31], the Hilbert space $\otimes_{j=1}^d L^2(\mathcal{V})$ can be decomposed into mutually orthogonal subspaces of (weakly) curl-free $\mathcal{H}_\times$, divergence-free $\mathcal{H}_\bullet$, and constant $\mathcal{H}_0$ vector fields, $\otimes_{j=1}^d L^2(\mathcal{V}) = \mathcal{H}_\times \oplus \mathcal{H}_\bullet \oplus \mathcal{H}_0$. The orthogonal projectors associated with this decomposition are given by $\boldsymbol{\Gamma}_\times = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}$, $\boldsymbol{\Gamma}_\bullet = \boldsymbol{\nabla}\times(-\boldsymbol{\Delta})^{-1}\boldsymbol{\nabla}\times$, and $\boldsymbol{\Gamma}_0 = \langle\cdot\rangle$, respectively, satisfying $\mathbf{I} = \boldsymbol{\Gamma}_\times + \boldsymbol{\Gamma}_\bullet + \boldsymbol{\Gamma}_0$ [31, 71, 66]. Here, $\boldsymbol{\Delta} = \mathrm{diag}(\Delta, \ldots, \Delta)$ is the vector Laplacian with inverse $\boldsymbol{\Delta}^{-1} = \mathrm{diag}(\Delta^{-1}, \ldots, \Delta^{-1})$, $\langle\cdot\rangle$ denotes space-time averaging over the period cell $\mathcal{T}\times\mathcal{V}$, and $\mathbf{I}$ is the identity operator on $\otimes_{j=1}^d L^2(\mathcal{V})$. Since the Hilbert space $\mathcal{H}_\mathcal{V} \subset \otimes_{j=1}^d L^2(\mathcal{V})$ is comprised of mean-zero functions, we have $\mathcal{H}_\mathcal{V} = \mathcal{H}_\times \oplus \mathcal{H}_\bullet$.

Due to the *curl-free* vector field $\boldsymbol{\nabla}\chi_j$ at the heart of the cell problem in equation (9), we will find particular use of the Hilbert space $\mathcal{H}_\times$, which we define as

$$\text{(A-38)} \qquad \mathcal{H}_\times = \{\boldsymbol{\psi} \in \mathcal{H}_\mathcal{V} \mid \boldsymbol{\Gamma}\boldsymbol{\psi} = \boldsymbol{\psi} \text{ weakly}\}, \quad \boldsymbol{\Gamma} = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot},$$

where we have denoted $\boldsymbol{\Gamma}_\times$ by $\boldsymbol{\Gamma}$ for notational simplicity. We also denote by $\langle\cdot,\cdot\rangle_\times$ the sesquilinear inner-product associated with the Hilbert space $\mathcal{H}_\times$, where $\langle\boldsymbol{\psi},\boldsymbol{\varphi}\rangle_\times = \langle\boldsymbol{\psi}\boldsymbol{\cdot}\boldsymbol{\varphi}\rangle_\mathcal{V}$, which induces a norm $\|\cdot\|_\times$ given by $\|\boldsymbol{\psi}\|_\times = \langle\boldsymbol{\psi},\boldsymbol{\psi}\rangle_\times^{1/2}$. Recall that the Sobolev space $\mathscr{H}_\mathcal{V}^{1,2}$ in (A-16) is the closure in the norm $\|\boldsymbol{\nabla}\cdot\|_\mathcal{V}$ of the space $C^2(\mathcal{V})$ of all twice continuously differentiable periodic functions in $\mathscr{H}_\mathcal{V}$ [14]. If $f \in \mathscr{H}_\mathcal{V}^{1,2}$, equation (A-22) shows that $\boldsymbol{\nabla}f$ is curl free, $\boldsymbol{\nabla}f \in \mathcal{H}_\times$, in the following weak sense. Let $\{f_n\}$ be a sequence of $\mathcal{V}$–periodic functions with $f_n \in C^2(\mathcal{V})$ that is Cauchy in the norm $\langle|\boldsymbol{\nabla}\cdot|^2\rangle_\mathcal{V}$ and converges to f in $L^2(\mathcal{V})$. Then, for all $\boldsymbol{\psi} \in \mathcal{H}_\times$ we have

$$\text{(A-39)} \quad \langle\boldsymbol{\Gamma}\boldsymbol{\nabla}f,\boldsymbol{\psi}\rangle_\times \equiv \lim_{n\to\infty}\langle\boldsymbol{\nabla}(-\Delta)^{-1}(-\Delta)f_n,\boldsymbol{\psi}\rangle_\times = \lim_{n\to\infty}\langle\boldsymbol{\nabla}f_n,\boldsymbol{\psi}\rangle_\times = \langle\boldsymbol{\nabla}f,\boldsymbol{\psi}\rangle_\times.$$

Consequently, since the differential operator $\boldsymbol{\nabla}$ maps $\mathscr{H}_\mathcal{V}^{1,2}$ to $\mathcal{H}_\mathcal{V}$ we have $\{\boldsymbol{\nabla}f \in \mathcal{H}_\mathcal{V} \,|\, f \in \mathscr{H}_\mathcal{V}^{1,2}\} \subseteq \mathcal{H}_\times$. It is therefore clear that on the function space $\{\boldsymbol{\nabla}f \in \mathcal{H}_\mathcal{V} \,|\, f \in \mathscr{H}_\mathcal{V}^{1,2}\}$ the operator $\boldsymbol{\Gamma}$ is a projection, hence bounded by unity in operator norm and trivially symmetric (since it acts as the identity operator on $\mathcal{H}_\times$). This establishes a direct link between the Hilbert spaces $\mathscr{H}^{1,2}_{\mathcal{V}}$ and $\mathcal{H}_\times$. The following lemma shows that these Hilbert spaces are in one-to-one isometric correspondence. This establishes that $\mathcal{H}_\times \equiv \{\boldsymbol{\nabla} f \in \mathcal{H}_{\mathcal{V}} \mid f \in \mathscr{H}^{1,2}_{\mathcal{V}}\}$ which, in turn, establishes that the linear symmetric bounded operator $\boldsymbol{\Gamma}$ with domain $\mathcal{H}_\times$ is self-adjoint.

Lemma 3 *The Hilbert spaces $\mathscr{H}^{1,2}_{\mathcal{V}}$ and $\mathcal{H}_\times$ are in one-to-one isometric correspondence, which we denote by $\mathscr{H}^{1,2}_{\mathcal{V}} \sim \mathcal{H}_\times$. More specifically, temporarily denote the inner-product induced norm of the Hilbert space $\mathscr{H}^{1,2}_{\mathcal{V}}$ by $\|f\|_{1,2} = \langle \boldsymbol{\nabla} f \boldsymbol{\cdot} \boldsymbol{\nabla} f\rangle^{1/2}_{\mathcal{V}}$. Then, for every $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ we have $\boldsymbol{\nabla} f \in \mathcal{H}_\times$ and $\|\boldsymbol{\nabla} f\|_\times = \|f\|_{1,2}$. Conversely, for every $\boldsymbol{\psi} \in \mathcal{H}_\times$ there exists unique $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ (up to equivalence class) such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|f\|_{1,2} = \|\boldsymbol{\psi}\|_\times$.*

Proof of Lemma 3. The discussion involving equation (A-39) shows that if $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$, then the vector field $\boldsymbol{\nabla} f \in \mathcal{H}_{\mathcal{V}}$ satisfies $\boldsymbol{\Gamma}\boldsymbol{\nabla} f = \boldsymbol{\nabla} f$ weakly so that $\boldsymbol{\nabla} f \in \mathcal{H}_\times$. Moreover, $\|\boldsymbol{\nabla} f\|^2_\times = \langle \boldsymbol{\nabla} f \boldsymbol{\cdot} \boldsymbol{\nabla} f\rangle_{\mathcal{V}} = \|f\|^2_{1,2} < \infty$. Consequently, for every $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ we have $\boldsymbol{\nabla} f \in \mathcal{H}_\times$ and $\|\boldsymbol{\nabla} f\|^2_\times = \|f\|^2_{1,2}$. Conversely, $\boldsymbol{\psi} \in \mathcal{H}_\times$ implies $\boldsymbol{\psi} = \boldsymbol{\Gamma}\boldsymbol{\psi} = \boldsymbol{\nabla} f$ weakly, where we have defined the scalar-valued function $f = \Delta^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{\psi}$. Since $\boldsymbol{\psi} = \boldsymbol{\nabla} f$, the $\mathscr{H}^{1,2}_{\mathcal{V}}$ norm of f satisfies $\|f\|^2_{1,2} = \langle \boldsymbol{\psi}\boldsymbol{\cdot}\boldsymbol{\psi}\rangle_{\mathcal{V}} = \|\boldsymbol{\psi}\|^2_\times < \infty$ so that $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$. Moreover, f is uniquely determined by $\boldsymbol{\psi}$ (up to a zero Lebesgue measure equivalence class), since if $f_1 = \Delta^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{\psi}$ and $f_2 = \Delta^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{\psi}$ then $\boldsymbol{\Gamma}\boldsymbol{\psi} = \boldsymbol{\psi}$ implies that $\|f_1 - f_2\|_{1,2} = \|\boldsymbol{\psi} - \boldsymbol{\psi}\|_\times = 0$. Consequently, for every $\boldsymbol{\psi} \in \mathcal{H}_\times$ there exists unique $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|f\|_{1,2} = \|\boldsymbol{\psi}\|_\times$. In summary, the Hilbert spaces $\mathscr{H}^{1,2}_{\mathcal{V}}$ and $\mathcal{H}_\times$ are in one-to-one isometric correspondence, which we denote by $\mathscr{H}^{1,2}_{\mathcal{V}} \sim \mathcal{H}_\times$. This concludes our proof of Lemma 3 $\square$.

Analogous to equation (A-17), we define the Hilbert space $\mathcal{H}$ and its everywhere dense subset $\mathcal{F}$,

$$\begin{aligned} \mathcal{H} &= \big\{\boldsymbol{\psi} \in \mathcal{H}_{\mathcal{T}} \otimes \mathcal{H}_\times \mid \langle \boldsymbol{\psi}\rangle = 0\big\}, \\ \mathcal{F} &= \big\{\boldsymbol{\psi} \in \tilde{\mathcal{A}}^0_{\mathcal{T}} \otimes \mathcal{H}_\times \mid \langle \boldsymbol{\psi}\rangle = 0\big\}. \end{aligned} \tag{A-40}$$

Recall that $\langle\cdot\rangle$ denotes space-time averaging over $\mathcal{T} \times \mathcal{V}$. Denote by $\langle\cdot,\cdot\rangle$ the sesquilinear inner-product associated with the Hilbert space $\mathcal{H}$, given by $\langle \boldsymbol{\psi}, \boldsymbol{\varphi}\rangle = \langle \boldsymbol{\psi}\boldsymbol{\cdot}\boldsymbol{\varphi}\rangle$, with $\langle \boldsymbol{\psi}, \boldsymbol{\varphi}\rangle = \overline{\langle \boldsymbol{\varphi}, \boldsymbol{\psi}\rangle}$. Here, $\boldsymbol{\psi}\boldsymbol{\cdot}\boldsymbol{\varphi} = \boldsymbol{\psi}^T\overline{\boldsymbol{\varphi}}$, transposition of the vector $\boldsymbol{\psi}$ is denoted $\boldsymbol{\psi}^T$, and $\overline{\boldsymbol{\varphi}}$ denotes component-wise complex conjugation, with $\boldsymbol{\psi}\boldsymbol{\cdot}\boldsymbol{\psi} = |\boldsymbol{\psi}|^2$. The norm $\|\cdot\|$ induced by this inner-product is given by $\|\boldsymbol{\psi}\| = \langle \boldsymbol{\psi}, \boldsymbol{\psi}\rangle^{1/2}$. In the case of a steady fluid velocity field $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$, we set $\mathcal{H} \equiv \mathcal{F} \equiv \mathcal{H}_\times$.

Since the fluid velocity field $\boldsymbol{u}$ is incompressible, $\boldsymbol{\nabla}\cdot\boldsymbol{u} = 0$, there is a real skew-symmetric matrix $\mathsf{H}(t, \boldsymbol{x})$ satisfying [4, 5]

$$\text{(A-41)} \qquad \boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}, \qquad \mathsf{H}^T = -\mathsf{H}.$$

Note that $\boldsymbol{\nabla}\cdot[\mathsf{H}\boldsymbol{\nabla}\varphi] = [\boldsymbol{\nabla}\cdot\mathsf{H}]\cdot\boldsymbol{\nabla}\varphi + \mathsf{H} : \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi$. Due to the anti-symmetry of the matrix H and the symmetry of the Hessian operator $\boldsymbol{\nabla}\boldsymbol{\nabla}$ when acting on a sufficiently smooth space of functions, we have $\mathsf{H} : \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi = 0$ for all such smooth functions φ, yielding

$$\text{(A-42)} \qquad \boldsymbol{\nabla}\cdot[\mathsf{H}\boldsymbol{\nabla}\varphi] = [\boldsymbol{\nabla}\cdot\mathsf{H}]\cdot\boldsymbol{\nabla}\varphi.$$

Using this identity and the representation of the velocity field $\boldsymbol{u}$ in (A-41), the advection-diffusion equation in (2) can be written as a diffusion equation [31, 71],

$$\text{(A-43)} \qquad \partial_t\phi = \boldsymbol{\nabla}\cdot\mathsf{D}\boldsymbol{\nabla}\phi, \quad \phi(0, \boldsymbol{x}) = \phi_0(\boldsymbol{x}), \qquad \mathsf{D} = \varepsilon\mathsf{I} + \mathsf{H},$$

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

$$\text{(A-44)} \qquad \mathsf{D}_{jk} = \varepsilon\delta_{jk} + \mathsf{H}_{jk}, \quad j, k = 1, \ldots, d.$$

The cell problem in (9) can also be written as the following diffusion equation [31, 71]

$$\text{(A-45)} \qquad \partial_\tau\chi_j = \boldsymbol{\nabla}_\xi\cdot[\mathsf{D}(\boldsymbol{\nabla}_\xi\chi_j + \boldsymbol{e}_j)], \quad \langle\boldsymbol{\nabla}_\xi\chi_k\rangle = 0, \qquad \mathsf{D} = \varepsilon\mathsf{I} + \mathsf{H},$$

where $\langle\boldsymbol{\nabla}_\xi\chi_k\rangle = 0$ follows from the periodicity of χ_k. We stress that equation (A-43) involves the slow $(t, \boldsymbol{x})$ and fast variables $(\tau, \boldsymbol{\xi})$, while equation (A-45) involves only the fast variables. As the remainder of the analysis involves only the fast variables, for notational simplicity, we will drop the subscripts ξ shown in equation (A-45) and use ∂_t to denote ∂_τ.

We now recast the first formula in equation (A-45) in a more suggestive, divergence form. Define the operator $\mathbf{T} : \tilde{\mathcal{A}}^0_{\mathcal{T}} \to \mathcal{H}_{\mathcal{T}}$ by $(\mathbf{T}\boldsymbol{\psi})_j = \partial_t\psi_j$, $j = 1, \ldots, d$. For $f \in \mathscr{F}$ we have [31, 71, 36]

$$\text{(A-46)} \qquad \boldsymbol{\nabla}(-\Delta)^{-1}\partial_t f = (-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\nabla} f,$$

in a weak distributional sense. This allows $\partial_t\chi_k$ in (A-45) to be written in divergence form [31, 71], $\partial_t\chi_k = (-\Delta)(-\Delta)^{-1}\partial_t\chi_k = -\boldsymbol{\nabla}\cdot[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\nabla}\chi_k$.

Define the vector-valued function $\boldsymbol{E}_k = \boldsymbol{\nabla}\chi_k + \boldsymbol{e}_k$ and the operator $\boldsymbol{\sigma} = \varepsilon\mathsf{I} + \mathbf{S}$, where $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$. In the case of a steady fluid velocity field $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$ we define $\mathbf{S} = \mathsf{H}$ and $\boldsymbol{\sigma} = \mathsf{D}$. With these definitions, the cell problem in (A-45) can be written via (A-42) as $\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{E}_k = 0$, $\langle\boldsymbol{E}_k\rangle = \boldsymbol{e}_k$, which is equivalent to

(A-47)

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

The formulas in (A-47) are analogous to the quasi-static limit of Maxwell's equations for a conductive medium [38, 66], where $\boldsymbol{E}_k$ and $\boldsymbol{J}_k$ play the role of the local electric field and current density, respectively, and $\boldsymbol{\sigma}$ plays the role of the local conductivity tensor of the medium. In the analytic continuation method for composites [38, 65, 10], the effective conductivity tensor $\boldsymbol{\sigma}^*$ is defined as

$$\langle\boldsymbol{J}_k\rangle = \boldsymbol{\sigma}^*\langle\boldsymbol{E}_k\rangle, \tag{A-48}$$

which relates the mean intensity and flux. In the setting of a *time-independent* fluid velocity field, where $\mathbf{S} = \mathsf{H}$, the linear constitutive relation $\boldsymbol{J}_k = \boldsymbol{\sigma}\boldsymbol{E}_k$ in (A-47) relates the local intensity and flux. In this case, due to the skew-symmetry of H, the local intensity-flux relationship is similar to that of a Hall medium [45, 31, 71, 66]. However, in the setting of a *time-dependent* fluid velocity field, where $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$, the constitutive relation $\boldsymbol{J}_k = \boldsymbol{\sigma}\boldsymbol{E}_k$ in (A-47) is a non-local integro-differential equation. A natural question to ask is the following. What is the precise relationship between the bulk transport coefficients D^* and $\boldsymbol{\sigma}^*$ for the two effective parameter problems? This question is addressed in Lemma 5 below.

We now derive functional formulas for the components S^*_{jk} and A^*_{jk}, $j, k = 1, \ldots, d$, of the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* that are analogous to those shown in equation (12). Writing the cell problem in (A-47) as $\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j = -\boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{e}_j = -u_j$, and inserting this expression for u_j into the functional $\langle u_j\,\chi_k\rangle$ in (10) yields

$$\begin{aligned}\langle u_j\,\chi_k\rangle &= -\langle[\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j]\,\chi_k\rangle \\ &= \langle\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle \\ &= \varepsilon\langle\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle + \langle\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle.\end{aligned} \tag{A-49}$$

Here, we have used the periodicity of χ_k and H in the second equality and the final equality follows from the property $\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j = \boldsymbol{\nabla}\chi_j$ and the symmetry of $\mathbf{\Gamma}$, together yielding $\langle \mathbf{S}\boldsymbol{\nabla}\chi_j \cdot \boldsymbol{\nabla}\chi_k\rangle = \langle \mathbf{S}\mathbf{\Gamma}\boldsymbol{\nabla}\chi_j \cdot \mathbf{\Gamma}\boldsymbol{\nabla}\chi_k\rangle = \langle \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}\boldsymbol{\nabla}\chi_j \cdot \boldsymbol{\nabla}\chi_k\rangle$. Equations (10), (11), and (A-49) imply that

(A-50)
$$\mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle \boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle), \qquad \mathsf{A}^*_{jk} = \langle \mathbf{A}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle, \qquad \mathbf{A} = \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}.$$

We stress that $\mathbf{\Gamma}$ is a self-adjoint projection on $\mathcal{H}$, implying

(A-51)
$$\langle \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle = \langle \mathbf{\Gamma}\mathbf{S}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle = \langle \mathbf{S}\mathbf{\Gamma}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle = \langle \mathbf{S}\boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle.$$

Since $\boldsymbol{\nabla}\chi_k$ is real-valued we have $\langle \boldsymbol{\nabla}\chi_k, \boldsymbol{\nabla}\chi_j\rangle = \langle \boldsymbol{\nabla}\chi_j, \boldsymbol{\nabla}\chi_k\rangle$, implying that S^*, as defined by (A-50), is a symmetric matrix. By Young's inequality in (A-21), the operators ∂_t and $(-\Delta)^{-1}$ commute on $\tilde{\mathscr{A}}^0_{\mathcal{T}} \otimes \mathscr{H}_{\mathcal{V}}$ (Theorem 2.27 in [36]). Therefore, we have $(-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\psi} = \mathbf{T}(-\boldsymbol{\Delta})^{-1}\boldsymbol{\psi}$, for $\boldsymbol{\psi} \in \mathcal{F}$ [36, 92]. This, the symmetry of $(-\boldsymbol{\Delta})^{-1}$ and the skew-symmetry of the operators $\mathbf{T}$ and H imply that the operator $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$ is skew-symmetric on $\mathcal{F}$. Since $\mathbf{\Gamma}$ is self-adjoint on $\mathcal{F}$, the operator $\mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}$ is also skew-symmetric on $\mathcal{F}$. Just as in the discussion below equation (12), this implies that A^*, as defined by (A-50), is an antisymmetric matrix.

Applying the integro-differential operator $\boldsymbol{\nabla}(-\Delta)^{-1}$ to the cell problem in equation (A-47), written via (A-42) as $\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j = -\boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{e}_j$, yields

$$\mathbf{\Gamma}(\varepsilon\mathsf{I} + \mathbf{S})\boldsymbol{\nabla}\chi_j = -\mathbf{\Gamma}\mathsf{H}\boldsymbol{e}_j. \tag{A-52}$$

This and $\mathbf{\Gamma}\boldsymbol{\nabla}\chi_j = \boldsymbol{\nabla}\chi_j$ provides the following resolvent formula for $\boldsymbol{\nabla}\chi_j$, which is analogous to equation (13),

$$\boldsymbol{\nabla}\chi_j = (\varepsilon\mathsf{I} + \mathbf{A})^{-1}\boldsymbol{g}_j, \qquad \boldsymbol{g}_j = -\mathbf{\Gamma}\mathsf{H}\boldsymbol{e}_j. \tag{A-53}$$

Inserting the resolvent formula for $\boldsymbol{\nabla}\chi_j$ in equation (A-53) into (A-50) yields the following analogue of equation (14)

$$\begin{aligned} \mathsf{S}^*_{jk} &= \varepsilon\left(\delta_{jk} + \langle (\varepsilon\mathsf{I} + \mathbf{A})^{-1}\boldsymbol{g}_j, (\varepsilon\mathsf{I} + \mathbf{A})^{-1}\boldsymbol{g}_k\rangle\right), \\ \mathsf{A}^*_{jk} &= \langle \mathbf{A}(\varepsilon\mathsf{I} + \mathbf{A})^{-1}\boldsymbol{g}_j, (\varepsilon\mathsf{I} + \mathbf{A})^{-1}\boldsymbol{g}_k\rangle, \end{aligned} \tag{A-54}$$

We therefore have the following corollary of Theorem 1.

Corollary 4 *The operator* $\mathbf{A} = \mathbf{\Gamma}\mathbf{S}\mathbf{\Gamma}$ *is a maximal (skew-symmetric) normal operator on the function space* $\mathcal{F}$, *hence* $\mathbf{M} = -\imath\mathbf{A}$ *is self-adjoint* *on $\mathcal{F}$. Let $\mathbf{Q}(\lambda)$ be the resolution of the identity in one-to-one correspondence with $\mathbf{M}$. Define the complex valued function $\mu_{jk}(\lambda) = \langle \mathbf{Q}(\lambda)\boldsymbol{g}_j, \boldsymbol{g}_k\rangle$, $j,k = 1,\ldots,d$, where $\boldsymbol{g}_j = -\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j$. Consider the positive measure μ_{kk} and the signed measures $Re\,\mu_{jk}$ and $Im\,\mu_{jk}$ associated with $\mu_{jk}(\lambda)$, introduced in equation* (A-4)*. Then, for $u_j \in \tilde{\mathscr{A}}_T^0 \otimes (\mathscr{H}_\mathcal{V} \cap L^r(\mathcal{V}))$, $2 < r \le \infty$, $\boldsymbol{\nabla}\chi_j \in \mathcal{F}$, and all $0 < \varepsilon < \infty$, the functional formulas for S^*_{jk} and A^*_{jk} shown in* (A-54) *have the Radon–Stieltjes integral representations shown in equation* (15).

Proof of Corollary 4. We first establish that the operator $\mathbf{M} = -\imath\mathbf{A}$ with domain $\mathcal{F}$ is self-adjoint, where $\mathbf{A} = \boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$ and $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$. Let's focus for now on the operator $-\imath\boldsymbol{\Gamma}[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\Gamma}$ with domain $\mathcal{F}$. Since $\boldsymbol{\Gamma} : \mathcal{H}_\mathcal{V} \to \mathcal{H}_\times$ is a projection, it acts as the identity on $\mathcal{H}_\times$. We can therefore focus on the operator $\imath[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]$. For $\boldsymbol{\psi} \in \mathcal{F}$, the jth component of the vector field $-\imath(-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\psi}$ is given by $-\imath(-\Delta)^{-1}\partial_t\psi_j$, where ψ_j is the jth component of $\boldsymbol{\psi}$. In the proof of Theorem 1 we established that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint. Since the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}_\mathcal{V}$ is self-adjoint [92], an analogous argument establishes that the operator $-\imath(-\boldsymbol{\Delta})^{-1}\mathbf{T}$ with domain $\mathcal{F}$ is self-adjoint.

Now focus on the operator $-\imath\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ with domain $\mathcal{H}$. Since $\boldsymbol{\Gamma}$ is a self-adjoint operator on $\mathcal{H}_\times$ and $-\imath\mathsf{H}$ is a Hermitian matrix, the operator $-\imath\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ is symmetric on $\mathcal{H}$. Recall that equation (A-41) provides the following representation of the fluid velocity field $\boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}$. We now establish that $\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ is bounded on $\mathcal{H}$ when the components u_j, $j = 1,\ldots,d$, of $\boldsymbol{u}$ satisfy $u_j \in \tilde{\mathscr{A}}_T^0 \otimes (\mathscr{H}_\mathcal{V} \cap L^r(\mathcal{V}))$ for $2 < r \le \infty$. This, in turn, establishes that the operator $-\imath\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ with domain $\mathcal{H}$ is self-adjoint. Lemma 2 implies that every $\boldsymbol{\psi} \in \mathcal{H}$ satisfies $\boldsymbol{\psi} = \boldsymbol{\nabla}f$ where $\boldsymbol{\psi}(t,\cdot) \in \mathcal{H}_\times$ and $f(t,\cdot) \in \mathscr{H}^{1,2}_\mathcal{V}$ for all $t \in \mathcal{T}$. Since the operator $\boldsymbol{\Gamma} = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\cdot$ acts as the identity on $\mathcal{H}_\times$ and $\boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}$, equation (A-42) implies

(A-55)
$$\|\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}\boldsymbol{\psi}\| = \|\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\nabla}f\| = \|\boldsymbol{\nabla}(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}f]\| = \|(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}f]\|_{1,2}.$$

This, Lemma 2, and Lemma 3 show that $\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ is bounded on $\mathcal{H}$.

We now summarize our findings. We have established that the operator $-\imath\boldsymbol{\Gamma}[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\Gamma}$ with domain $\mathcal{F}$ is self-adjoint and the operator $-\imath\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{\Gamma}$ with domain $\mathcal{H}$ is self-adjoint when the components u_j, $j = 1,\ldots,d$, of $\boldsymbol{u}$ satisfy $u_j \in \tilde{\mathscr{A}}_T^0 \otimes (\mathscr{H}_\mathcal{V} \cap L^r(\mathcal{V}))$ for $2 < r \le \infty$. Consequently, the sum of these two operators $\mathbf{M} = -\imath\mathbf{A}$, where $\mathbf{A} = \boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$ and $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T}+\mathsf{H}$, with domain $D(\mathbf{M}) = \mathcal{F}\cap\mathcal{H} = \mathcal{F}$ [94] is self-adjoint when $u_j \in \tilde{\mathscr{A}}_T^0\otimes(\mathscr{H}_\mathcal{V}\cap L^r(\mathcal{V}))$ for $2 < r \le \infty$. Thus $\mathbf{A} = \imath\mathbf{M}$ is a maximal (skew-symmetric) normal operator on $\mathcal{F}$ [94].

In the proof of Theorem 1 we established that the functions $F(\lambda) = (\varepsilon + \imath\lambda)^{-1}$ and $G(\lambda) = \imath\lambda(\varepsilon + \imath\lambda)^{-1}$ involved in the functional formulas for S^*_{jk} and A^*_{jk} in (A-54) are bounded for all $0 < \varepsilon < \infty$ so that $\boldsymbol{\varphi} \in \mathscr{D}(F)$ and $\boldsymbol{\varphi} \in \mathscr{D}(G)$ for all $\boldsymbol{\varphi} \in D(\mathbf{M})$ when $0 < \varepsilon < \infty$. By equation (A-41) and the definition of $g_j = (-\Delta)^{-1} u_j$ in (13) we have

$$\text{(A-56)} \qquad \boldsymbol{g}_j = -\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j = \boldsymbol{\nabla}(-\Delta)^{-1} u_j = \boldsymbol{\nabla} g_j.$$

In the proof of Theorem 1 we established that $g_j \in \mathscr{F}$. This, equation (A-56), and Lemma 3 implies that $\boldsymbol{g}_j \in \mathcal{F}$. Since $\mathcal{F} \subseteq D(\mathbf{M})$, the conditions of the spectral theorem are satisfied. Just as in the remainder of the proof of Theorem 1, this establishes the integral representations for S^*_{jk} and A^*_{jk} shown in (15). From equation (A-7), the mass μ^0_{jk} of the measure μ_{jk} is given by

$$\text{(A-57)} \qquad \mu^0_{jk} = \langle \boldsymbol{g}_j, \boldsymbol{g}_k \rangle = \langle \boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j, \boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_k \rangle = \langle \mathsf{H}^T \boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j, \boldsymbol{e}_k \rangle.$$

Moreover, $|\mu^0_{jk}| \le \|\mathsf{H}\|^2 < \infty$ for all $j, k = 1, \ldots, d$. This completes the proof of Corollary 4 $\square$.

We conclude this section with the following lemma, which provides a precise relationship between the effective parameter $\boldsymbol{\sigma}^*$ defined in equation (A-48) and the effective parameter D^* defined in (10).

Lemma 5 *Let the components D^*_{jk} and σ^*_{jk}, $j, k = 1, \ldots, d$, of the effective tensors D^* and $\boldsymbol{\sigma}^*$ be defined as in equations* (4)–(10) *and* (A-43)–(A-48), *respectively. Then these effective tensors related by*

$$\text{(A-58)} \qquad \boldsymbol{\sigma}^* = [\mathsf{D}^*]^T + \langle \mathsf{H} \rangle.$$

Proof of Lemma 5. Below equation (A-37) we discussed the Helmholtz theorem, i.e., the orthogonal decomposition $\otimes_{j=1}^d L^2(\mathcal{V}) = \mathcal{H}_\times \oplus \mathcal{H}_\bullet \oplus \mathcal{H}_0$. Define the function spaces $\mathcal{F}_\times = \tilde{\mathcal{A}}^0_{\mathcal{T}} \otimes \mathcal{H}_\times$ and $\mathcal{F}_\bullet = \tilde{\mathcal{A}}^0_{\mathcal{T}} \otimes \mathcal{H}_\bullet$. From equation (A-47), the vector-valued functions $\boldsymbol{J}_k = \boldsymbol{\sigma}\boldsymbol{E}_k$ and $\boldsymbol{E}_k = \boldsymbol{\nabla}\chi_k + \boldsymbol{e}_k$ satisfy $\boldsymbol{J}_k \in \mathcal{F}_\bullet$ and $\boldsymbol{E}_k \in \mathcal{F}_\times$ while $\boldsymbol{\nabla}\chi_k \in \{\boldsymbol{\psi} \in \mathcal{F}_\times \mid \langle \boldsymbol{\psi} \rangle = 0\}$, where $\boldsymbol{\sigma} = \varepsilon\mathsf{I} + \mathbf{S}$ and $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$. By the mutual orthogonality of the Hilbert spaces $\mathcal{H}_\times$ and $\mathcal{H}_\bullet \oplus \mathcal{H}_0$ we have $\langle \boldsymbol{J}_j \cdot \boldsymbol{\nabla}\chi_k \rangle = 0$ for all $j, k = 1, \ldots, d$ (which is equivalent to equation (A-49)). Consequently, from equation (A-48) we have $\langle \boldsymbol{J}_j \cdot \boldsymbol{E}_k \rangle = \langle \boldsymbol{J}_j \cdot \boldsymbol{e}_k \rangle = \sigma^*_{jk}$.

By the definition $\boldsymbol{u} = \boldsymbol{\nabla} \cdot \mathsf{H}$ in (A-41) and periodicity, integration by parts yields $\langle \mathsf{H}\boldsymbol{e}_j \cdot \boldsymbol{\nabla}\chi_k \rangle = -\langle u_j \chi_k \rangle$. From $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$ we also have

$\mathbf{S}\boldsymbol{e}_j = \mathsf{H}\boldsymbol{e}_j$. Therefore, by the skew-symmetry of $\mathbf{S}$, $\langle \boldsymbol{\nabla}\chi_j \rangle = 0$, and the formula $\mathsf{D}^*_{jk} = \varepsilon\delta_{jk} + \langle u_j \chi_k \rangle$ in (10), we have

$$
\begin{aligned}
\sigma^*_{jk} &= \langle \boldsymbol{J}_j \cdot \boldsymbol{e}_k \rangle \\
&= \langle (\varepsilon \mathsf{I} + \mathbf{S})\boldsymbol{\nabla}\chi_j \cdot \boldsymbol{e}_k \rangle + \langle (\varepsilon \mathsf{I} + \mathbf{S})\boldsymbol{e}_j \cdot \boldsymbol{e}_k \rangle \\
&= -\langle \boldsymbol{\nabla}\chi_j \cdot \mathsf{H}\boldsymbol{e}_k \rangle + \langle (\varepsilon \mathsf{I} + \mathsf{H})\boldsymbol{e}_j \cdot \boldsymbol{e}_k \rangle \\
&= \langle \chi_j \, u_k \rangle + \varepsilon\delta_{jk} + \langle \mathsf{H}_{jk} \rangle \\
&= \mathsf{D}^*_{kj} + \langle \mathsf{H}_{jk} \rangle,
\end{aligned}
\tag{A-59}
$$

which is equivalent to (A-58). This concludes our proof of Lemma 5 □.

Appendix D. An isometric correspondence

A natural question to ask is the following. Is the formulation of the effective parameter problem described in Theorem 1 equivalent to the effective parameter problem described in Corollary 4? The answer is in the affirmative. The correspondence between the two formulations is one of isometry, and is summarized by the following theorem.

Theorem 6 *The function spaces $\mathscr{F}$ and $\mathcal{F}$ defined in equations* (A-17) *and* (A-40) *are in one-to-one isometric correspondence. This induces a one-to-one isometric correspondence between the domains $D(A)$ and $D(\mathbf{A})$ of the operators A and $\mathbf{A}$ defined in equations* (12) *and* (A-50)*, respectively. Specifically, for every $f \in D(A)\cap\mathscr{F}$ we have $\boldsymbol{\nabla} f \in D(\mathbf{A})\cap\mathcal{F}$ and $\|Af\|_{1,2} = \|\mathbf{A}\boldsymbol{\nabla} f\|$, and conversely, for each $\boldsymbol{\psi} \in D(\mathbf{A}) \cap \mathcal{F}$ there exists unique $f \in D(A) \cap \mathscr{F}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|\mathbf{A}\boldsymbol{\psi}\| = \|Af\|_{1,2}$. The Radon–Stieltjes measures underlying the integral representations of Theorem 1 and Corollary 4 are equal, $\mathrm{d}\langle Q(\lambda)g_j, g_k\rangle_{1,2} = \mathrm{d}\langle \mathbf{Q}(\lambda)\boldsymbol{g}_j, \boldsymbol{g}_k\rangle$, $j,k = 1,\ldots,d$, up to null sets of measure zero, where $\boldsymbol{g}_j = \boldsymbol{\nabla} g_j$. Moreover, the operators $\mathbf{A}$ and A are related by $\mathbf{A}\boldsymbol{\nabla} = \boldsymbol{\nabla} A$, which implies and is implied by the weak equality $\mathbf{Q}(\lambda)\boldsymbol{\nabla} = \boldsymbol{\nabla} Q(\lambda)$.*

Proof of Theorem 6. Recall, we have $\boldsymbol{\nabla} g_j = \boldsymbol{g}_j$ from equation (A-56). We use the formula $\boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}$ in equation (A-41) and the weak identity in (A-42) to write the operator $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$ defined in (12) as $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{\nabla})$. Using the definition $\boldsymbol{\Gamma} = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\cdot$ in (A-38), the formula $\boldsymbol{\nabla}(-\Delta)^{-1}\partial_t = (-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\nabla}$ in (A-46), and the representation $\mathbf{A} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \boldsymbol{\Gamma}\mathsf{H}$, which holds in the weak sense shown in (A-51), the operators A and $\mathbf{A}$ are related by

$$
\boldsymbol{\nabla} A = [(-\boldsymbol{\Delta})^{-1}\mathbf{T} + \boldsymbol{\Gamma}\mathsf{H}]\boldsymbol{\nabla} = \mathbf{A}\boldsymbol{\nabla}, \qquad \boldsymbol{\nabla} g_j = \boldsymbol{g}_j.
\tag{A-60}
$$

Consequently, by applying the differential operator $\boldsymbol{\nabla}$ to both sides of the formula $(\varepsilon + A)\chi_j = g_j$ of (13), we obtain the formula $(\varepsilon \mathsf{I} + \mathbf{A})\boldsymbol{\nabla}\chi_j = \boldsymbol{g}_j$ of equation (A-53).

Since the function spaces $\mathscr{F}$ and $\mathcal{F}$ differ only in the characterization of the spatial variable, the one-to-one isometry $\mathscr{H}_{\mathcal{V}}^{1,2} \sim \mathcal{H}_\times$ established in Lemma 3 induces the one-to-one isometry $\mathscr{F} \sim \mathcal{F}$. We now demonstrate that the one-to-one isometry between $\mathscr{F}$ and $\mathcal{F}$ induces a one-to-one isometry between the domains $D(A)$ and $D(\mathbf{A})$ of the operators A and $\mathbf{A}$. This, in turn, follows from the one-to-one correspondence between a self-adjoint operator and its resolution of the identity discussed in Appendix A, leading to equation (A-10). More specifically, the domain $D(M)$ of the self-adjoint operator M, for example, comprises those and only those elements f of $\mathscr{H}$ such that the Stieltjes integral $\int \lambda^2 \, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2$ is convergent, and when $f \in D(M)$ the element Mf is determined by the relations in equation (A-10). Since $A = \imath M$ it is clear that $D(A) = D(M)$. We established in Appendix C.1 that $\mathscr{F} \subseteq D(A)$ and in Appendix C.2 that $\mathcal{F} \subseteq \mathbf{A}$.

Let $f \in D(A) \cap \mathscr{F}$. From the relation $\mathscr{F} \sim \mathcal{F}$, we have that $\boldsymbol{\nabla} f \in \mathcal{F}$, so from equation (A-60)

(A-61)
$$\|Af\|_{1,2}^2 = \langle Af, Af\rangle_{1,2} = \langle \boldsymbol{\nabla} Af \boldsymbol{\cdot} \boldsymbol{\nabla} Af\rangle = \langle \mathbf{A}\boldsymbol{\nabla} f \boldsymbol{\cdot} \mathbf{A}\boldsymbol{\nabla} f\rangle = \|\mathbf{A}\boldsymbol{\nabla} f\|^2.$$

Consequently, from equation (A-10) we have that

$$\int \lambda^2 \, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 = \int \lambda^2 \, \mathrm{d}\|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|^2, \tag{A-62}$$

and the convergence of the integral on the left-hand-side of (A-62) implies the convergence of the integral on the right-hand-side which, in turn, implies that $\boldsymbol{\nabla} f \in D(\mathbf{A})$.

Conversely, let $\boldsymbol{\psi} \in D(\mathbf{A}) \cap \mathcal{F}$. From the relation $\mathscr{F} \sim \mathcal{F}$, there exists unique $f \in \mathscr{F}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$. Equation (A-60) then implies that

$$\|\mathbf{A}\boldsymbol{\psi}\|^2 = \langle \mathbf{A}\boldsymbol{\nabla} f, \mathbf{A}\boldsymbol{\nabla} f\rangle = \langle \boldsymbol{\nabla} Af, \boldsymbol{\nabla} Af\rangle = \langle Af, Af\rangle_{1,2} = \|Af\|_{1,2}^2. \tag{A-63}$$

Again, equation (A-10) implies that (A-62) holds, and the convergence of the integral on the right-hand-side of (A-62) implies the convergence of the integral on the left-hand-side which, in turn, implies that $f \in D(A)$.

In summary, for every $f \in D(A) \cap \mathscr{F}$ we have $\boldsymbol{\nabla} f \in D(\mathbf{A})$ and $\|Af\|_{1,2}^2 = \|\mathbf{A}\boldsymbol{\nabla} f\|^2$. Conversely, for every $\boldsymbol{\psi} \in D(\mathbf{A}) \cap \mathcal{F}$, there exists unique $f \in D(A)$

such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|\mathbf{A}\boldsymbol{\psi}\|^2 = \|Af\|_{1,2}^2$. This generates a one-to-one isometric correspondence between the domains $D(\mathbf{A})$ and $D(A)$.

We now show that this result implies, and is implied by the weak equality $\boldsymbol{\nabla} Q(\lambda) = \mathbf{Q}(\lambda)\boldsymbol{\nabla}$, where $Q(\lambda)$ and $\mathbf{Q}(\lambda)$ are the *self-adjoint projection operators* in one-to-one correspondence with the operators A and $\mathbf{A}$, respectively. From equation (A-62) and the linearity properties of Radon–Stieltjes integrals [94], we have that

$$\begin{aligned} 0 &= \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}(\|Q(\lambda)f\|_{1,2}^2 - \|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|^2) \\ &= \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}(\langle [\boldsymbol{\nabla} Q(\lambda) - \mathbf{Q}(\lambda)\boldsymbol{\nabla}] f \boldsymbol{\cdot} \boldsymbol{\nabla} f\rangle). \end{aligned} \tag{A-64}$$

Equation (A-64) implies that for all $f \in D(A) \cap \mathscr{F} \iff \boldsymbol{\nabla} f \in D(\mathbf{A}) \cap \mathcal{F}$ we have $\mathrm{d}\|Q(\lambda)f\|_{1,2}^2 = \mathrm{d}\|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|^2$, up to sets of measure zero. Moreover, the equality $\boldsymbol{\nabla} Q(\lambda) = \mathbf{Q}(\lambda)\boldsymbol{\nabla}$ holds in this weak sense. Conversely, assume that $Q(\lambda)$ and $\mathbf{Q}(\lambda)$ are the resolutions of the identity in one-to-one correspondence with the operators A and $\mathbf{A}$ and that $\boldsymbol{\nabla} Q(\lambda) f = \mathbf{Q}(\lambda)\boldsymbol{\nabla} f$ for every $f \in D(A) \cap \mathscr{F} \iff \boldsymbol{\nabla} f \in D(\mathbf{A}) \cap \mathcal{F}$. Then equation (A-64) holds and implies equation (A-62). Equation (A-10) then implies that $\|\mathbf{A}\boldsymbol{\nabla} f\|^2 = \|Af\|_{1,2}^2 = \|\boldsymbol{\nabla} Af\|^2$, which implies that $\mathbf{A}\boldsymbol{\nabla} = \boldsymbol{\nabla} A$ in this weak sense. Since $g_k \in D(A)$ and $\boldsymbol{g}_k \in D(\mathbf{A})$ with $\boldsymbol{g}_k = \boldsymbol{\nabla} g_k$, this result implies that the Radon–Stieltjes measures underlying the integral representations of Theorem 1 are equal to that of Corollary 4, $\mathrm{d}\|Q(\lambda)g_k\|_{1,2} = \mathrm{d}\|\mathbf{Q}(\lambda)\boldsymbol{g}_k\|$, up to null sets of measure zero, for all $j, k = 1, \ldots, d$. This concludes our proof of Theorem 6 $\square$.

D.1. Discrete integral representations by eigenfunction expansion

The integral representations of Theorem 1 and Corollary 4 shown in equation (15), involve a spectral measure μ_{jk}, $j, k = 1, \ldots, d$, which has discrete and continuous components [85, 94]. In this section, we review these properties of μ_{jk} and provide an explicit formula for its discrete component. Towards this goal, we summarize some general spectral properties of the self-adjoint operators $M = -\imath A$ and $\mathbf{M} = -\imath\mathbf{A}$ on the function spaces $\mathscr{F}$ and $\mathcal{F}$, which are dense subsets of the associated Hilbert spaces $\mathscr{H}$ and $\mathcal{H}$, given in equations (A-17) and (A-40), respectively. We will focus on the operator M and the Hilbert space $\mathscr{H}$, as the discussion regarding $\mathbf{M}$ and $\mathcal{H}$ is analogous.

Recall from equation (A-10) that the domain $D(M)$ of the self-adjoint operator M comprises those and only those elements $f \in \mathscr{H}$ such that

$\|Mf\|_{1,2}^2 = \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 < \infty$, where $Q(\lambda)$ is the resolution of the identity in one-to-one correspondence with M [94]. The integration is over the spectrum Σ of M, which has continuous Σ_{cont} and discrete (pure-point) Σ_{pp} components, $\Sigma = \Sigma_{\text{cont}} \cup \Sigma_{\text{pp}}$ [85, 94]. We first focus on the discrete spectrum Σ_{pp}.

The $f \in \mathscr{H}$, $f \neq 0$, satisfying $Mf = \lambda f$ with $\lambda \in \Sigma_{\text{pp}}$ are called eigenfunctions and λ is the corresponding eigenvalue. Since M is self-adjoint, λ is real-valued [94]. The span of all eigenfunctions is a *countable* subspace of $\mathscr{H}$ [94]. Accordingly, we will denote the eigenfunctions by φ_l, $l = 1, 2, 3, \ldots$, with corresponding eigenvalues λ_l. Eigenfunctions corresponding to distinct eigenvalues are orthogonal and can be normalized to be orthonormal [94], i.e. if $M\varphi_l = \lambda_l \varphi_l$ and $M\varphi_m = \lambda_m \varphi_m$ for $\lambda_l \neq \lambda_m$, then $\langle \varphi_m, \varphi_n \rangle_{1,2} = \delta_{mn}$. There can be more than one eigenfunction associated with a particular eigenvalue. However, they are linearly independent and, without loss of generality, can be taken to be orthonormal [94]. Consequently, associated with each eigenfunction φ_l is a closed linear manifold, which we denote by $\mathcal{M}(\varphi_l)$. When $l \neq m$, $\mathcal{M}(\varphi_l)$ and $\mathcal{M}(\varphi_m)$ are mutually orthogonal. Set $\mathcal{E} = \oplus_{l=1}^{\infty} \mathcal{M}(\varphi_l)$, $\mathcal{M} = \mathcal{E} \oplus \{0\}$, and let $\mathcal{N} = \mathcal{M}^{\perp}$ be the orthogonal complement of $\mathcal{M}$ in $\mathscr{H}$. All the properties of $\mathcal{M}$ and $\mathcal{N}$ that are relevant here have been collected in the following theorem [94], which provides a natural decomposition of the Hilbert space $\mathscr{H}$ in terms of the mutually orthogonal, closed linear manifolds $\mathcal{M}$ and $\mathcal{N}$, and leads to a decomposition of the measure μ_{kk} into its discrete and continuous components.

Theorem 7 ([94] pages 189 and 247) *One of the three cases must occur:*

1. $\mathcal{E} = \emptyset$ *and* $\mathcal{M} = \{0\}$ *has dimension zero;* $\mathcal{N} = \mathscr{H}$ *has countably infinite dimension. There exists an orthonormal set* $\{\psi_m\}$, $m = 1, 2, 3, \ldots$, *and mutually orthogonal, closed linear manifolds* $\mathcal{N}(\psi_m)$ *which determine* $\mathcal{N}$ *according to* $\mathcal{N} = \oplus_{m=1}^{\infty} \mathcal{N}(\psi_m)$.
2. $\mathcal{E}$ *contains an incomplete orthonormal set* $\{\varphi_l\}$ *so that both* $\mathcal{M}$ *and* $\mathcal{N}$ *are proper subsets of* $\mathscr{H}$, $\mathcal{N}$ *having countably infinite dimension and* $\mathcal{M}$ *having finite or countably infinite dimension. There exists an orthonormal set* $\{\psi_m\}$ *in* $\mathcal{N}$. *The closed linear manifolds* $\mathcal{M}(\varphi_l)$ *and* $\mathcal{N}(\psi_m)$ *are mutually orthogonal and together determine* $\mathscr{H}$ *according to*

$$\mathcal{M} = \oplus_{l=1}^{\infty} \mathcal{M}(\varphi_l), \qquad \mathcal{N} = \oplus_{m=1}^{\infty} \mathcal{N}(\psi_m), \qquad \mathscr{H} = \mathcal{M} \oplus \mathcal{N}.$$

3. $\mathcal{E}$ *contains a complete orthonormal set* $\{\varphi_l\}$*;* $\mathcal{M} = \mathscr{H}$ *has countably infinite dimension;* $\mathcal{N} = \{0\}$ *has zero dimension. In this case, the*

closed linear manifolds $\mathcal{M}(\varphi_l)$ are mutually orthogonal and together determine $\mathcal{M}$ according to $\mathcal{M} = \oplus_{l=1}^{\infty}\mathcal{M}(\varphi_l)$.

In each of these three cases, the closed linear manifolds $\mathcal{M}$ and $\mathcal{N}$ reduce M, i.e., M leaves both $\mathcal{M}$ and $\mathcal{N}$ invariant in the sense that if $f \in D(M)$ and $f \in \mathcal{N}$ then $Mf \in \mathcal{N}$, and similarly for $\mathcal{M}$. In cases (2) and (3), a necessary and sufficient condition that an element $\varphi_l \in \mathscr{H}$ be an eigenfunction with eigenvalue λ_l, is that the function $\|Q(\lambda)\varphi_l\|_{1,2}^2$ is constant on each of the intervals $-\infty < \lambda < \lambda_l$ and $\lambda_l < \lambda < \infty$ [94]. Moreover, a necessary and sufficient condition that $f \in \mathcal{M}$, $f \neq 0$, is

$$f = \sum_{l=1}^{\infty} \langle f, \varphi_l \rangle_{1,2}\, \varphi_l, \qquad \|f\|_{1,2}^2 = \sum_{l=1}^{\infty} |\langle f, \varphi_l \rangle_{1,2}|^2 \neq 0, \tag{A-65}$$

and similarly for $f \in \mathcal{N}$ with orthonormal set $\{\psi_m\}$. In cases (1) and (2), a necessary and sufficient condition that $\psi \neq 0$ be an element of $\mathcal{N}$ is that $\|Q(\lambda)\psi\|_{1,2}^2$ be a continuous function of λ not identically zero [94].

Let f be an arbitrary element of $\mathscr{H}$, and g and h be its (unique) projections on $\mathcal{M}$ and $\mathcal{N}$, respectively, then the equation

$$\begin{aligned} \|Q(\lambda)f\|_{1,2}^2 &= \|Q(\lambda)g\|_{1,2}^2 + \|Q(\lambda)h\|_{1,2}^2, \\ \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 &= \mathrm{d}\|Q(\lambda)g\|_{1,2}^2 + \mathrm{d}\|Q(\lambda)h\|_{1,2}^2 \end{aligned} \tag{A-66}$$

is valid and provides the standard resolution of the monotone function $\|Q(\lambda)f\|_{1,2}^2$ into its discontinuous and continuous monotone components, as well as the decomposition of the measure $\mathrm{d}\|Q(\lambda)f\|_{1,2}^2$ into its discrete and continuous components.

We now use the mathematical framework summarized in Theorem 7 to provide explicit formulas for the discrete parts of the integral representations for S_{jk}^* and A_{jk}^*, shown in equation (15). Recall the cell problem in equation (9) written as in (A-18), $(\varepsilon + A)\chi_j = g_j$. Here $A = \imath M$ is defined in (12), $g_j = (-\Delta)^{-1}u_j$, and u_j is the j^{th} component of the velocity field $\boldsymbol{u}$, $j = 1, \ldots, d$. Moreover, we have $\chi_j, g_j \in \mathscr{F} \subset \mathscr{H}$ and $\mathscr{F} \subset D(A)$. We stress that the arguments presented here are more subtle than those typically used for *bounded* operators in Hilbert space. The reason is a bounded linear operator commutes with all the infinite sums encountered here, by the dominated convergence theorem [36]. However, for the operator A, we must instead rely on general principles of unbounded linear operators in Hilbert space.

Let $\tilde{\chi}_j$ and $\chi_j^\perp$ be the (unique) projections of χ_j on $\mathcal{M}$ and $\mathcal{N}$, respectively, with $\chi_j = \tilde{\chi}_j + \chi_j^\perp$ and similarly for g_j. Since $A = \imath M$ is a linear operator, we have $A\chi_j = A\tilde{\chi}_j + A\chi_j^\perp$. From Theorem 7, the linear manifolds $\mathcal{M}$ and $\mathcal{N}$ both reduce A, which implies $A\tilde{\chi}_j \in \mathcal{M}$ and $A\chi_j^\perp \in \mathcal{N}$. From equation (A-65) we then have $A\tilde{\chi}_j = \sum_l \langle A\tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l$ and

$$\text{(A-67)} \qquad \chi_j = \sum_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + \chi_j^\perp, \quad A\chi_j = \sum_l \imath\lambda_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + A\chi_j^\perp$$

where we have used $\langle A\tilde{\chi}_j, \varphi_l\rangle_{1,2} = -\langle \tilde{\chi}_j, A\varphi_l\rangle_{1,2} = -\langle \tilde{\chi}_j, \imath\lambda_l\varphi_l\rangle_{1,2} = \imath\lambda_l\langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}$. From the cell problem $(\varepsilon + A)\chi_j = g_j$ we therefore have

$$\text{(A-68)} \qquad \varepsilon \sum_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\varphi_l + \sum_l \imath\lambda_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\varphi_l + (\varepsilon + A)\chi_j^\perp = \tilde{g}_j + g_j^\perp,$$

where $(\varepsilon + A)\chi_j^\perp, g_j^\perp \in \mathcal{N}$. Of course, each $f \in \mathcal{N}$ can be represented [94] as $f = \sum_m \langle f, \psi_m\rangle_{1,2}\, \psi_m$, where $\{\psi_m\}$ is the orthonormal set defined in Theorem 7, though we have suppressed this notation in the above equations for simplicity. By the mutual orthogonality of the linear manifolds $\mathcal{M}$ and $\mathcal{N}$, the completeness of the set $\{\varphi_l\} \cup \{\psi_m\}$, and the Parseval identity, taking the inner-product of both sides of equation (A-68) with φ_n yields

$$\text{(A-69)} \qquad \langle \tilde{\chi}_j, \varphi_n\rangle_{1,2} = \frac{\langle \tilde{g}_j, \varphi_n\rangle_{1,2}}{\varepsilon + \imath\lambda_n}\,, \qquad 0 < \varepsilon < \infty.$$

Recall the representations $\mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle \chi_j, \chi_k\rangle_{1,2})$ and $\mathsf{A}^*_{jk} = \langle A\chi_j, \chi_k\rangle_{1,2}$, $j, k = 1, \ldots, d$, shown in equation (12). Writing $\chi_j = \tilde{\chi}_j + \chi_j^\perp$ and $A\chi_j = A\tilde{\chi}_j + A\chi_j^\perp$, the mutual orthogonality of the linear manifolds $\mathcal{M}$ and $\mathcal{N}$, which both reduce A, implies $\langle \chi_j, \chi_k\rangle_{1,2} = \langle \tilde{\chi}_j, \tilde{\chi}_k\rangle_{1,2} + \langle \chi_j^\perp, \chi_k^\perp\rangle_{1,2}$ and $\langle A\chi_j, \chi_k\rangle_{1,2} = \langle A\tilde{\chi}_j, \tilde{\chi}_k\rangle_{1,2} + \langle A\chi_j^\perp, \chi_k^\perp\rangle_{1,2}$. Consequently, from equations (A-67) and (A-69), the completeness of the set $\{\varphi_l\} \cup \{\psi_m\}$, and the Parseval identity, we have

$$
\begin{aligned}
\text{(A-70)} \qquad \langle \chi_j, \chi_k \rangle_{1,2} - \langle \chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2} &= \sum_l \langle \tilde{\chi}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{\chi}_k, \varphi_l \rangle}_1 \\
&= \sum_l \frac{\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{g}_k, \varphi_l \rangle}_1}{\varepsilon^2 + \lambda_l^2} \\
\langle A\chi_j, \chi_k \rangle_{1,2} - \langle A\chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2} &= \sum_l \imath\lambda_l \, \langle \tilde{\chi}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{\chi}_k, \varphi_l \rangle}_1 \\
&= \sum_l \frac{\imath\lambda_l \, \langle \tilde{g}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{g}_k, \varphi_l \rangle}_1}{\varepsilon^2 + \lambda_l^2} \, .
\end{aligned}
$$

Since χ_j and $A\chi_j$ are real-valued, just as in equation (A-9), we have

$$
\begin{aligned}
\text{(A-71)} \qquad \langle \chi_j, \chi_k \rangle_{1,2} - \langle \chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2} &= \sum_l \frac{\mathrm{Re}\,[\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{g}_k, \varphi_l \rangle}_1]}{\varepsilon^2 + \lambda_l^2} \\
\langle A\chi_j, \chi_k \rangle_{1,2} - \langle A\chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2} &= -\sum_l \frac{\lambda_l \, \mathrm{Im}\,[\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \overline{\langle \tilde{g}_k, \varphi_l \rangle}_1]}{\varepsilon^2 + \lambda_l^2} \, .
\end{aligned}
$$

The right hand sides of the formulas in equation (A-71) are Radon–Stieltjes integrals associated with a *discrete* measure. The terms $\langle \chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2}$ and $\langle A\chi_j^{\perp}, \chi_k^{\perp} \rangle_{1,2}$ also have Radon–Stieltjes integral representations involving the *continuous* measure $\mathrm{d}\langle Q(\lambda) g_j^{\perp}, g_k^{\perp} \rangle_{1,2}$ via equation (15). We note that from the decomposition $g_j = \tilde{g}_j + g_j^{\perp}$, we have $\langle \tilde{g}_j, \varphi_l \rangle_{1,2} = \langle g_j, \varphi_l \rangle_{1,2}$. A useful property of the inner-product $\langle g_j, \varphi_l \rangle_{1,2}$ and the form of $g_j = (-\Delta)^{-1} u_j$ is that (see equation (A-19))

$$
\text{(A-72)} \qquad \langle g_j, \varphi_l \rangle_{1,2} = \langle u_j, \varphi_l \rangle_2 .
$$

This property is used in Section 3 to calculate S_{jk}^* and A_{jk}^* for a large class of velocity fields.

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N. Benjamin Murphy,
University of California at Irvine, Department of Mathematics, 340 Rowland Hall, Irvine, CA 92697-3875, USA *E-mail address:* `nbmurphy@math.uci.edu`

Elena Cherkaev,
University of Utah, Department of Mathematics, 155 South 1400 East Room 233, Salt Lake City, UT 84112-0090, USA *E-mail address:* `elena@math.utah.edu`

Jack Xin,
University of California at Irvine, Department of Mathematics, 340 Rowland Hall, Irvine, CA 92697-3875, USA *E-mail address:* `jxin@math.uci.edu`

Jingyi Zhu,
University of Utah, Department of Mathematics, 155 South 1400 East Room 233, Salt Lake City, UT 84112-0090, USA *E-mail address:* `zhu@math.utah.edu`

Kenneth M. Golden
University of Utah, Department of Mathematics, 155 South 1400 East Room 233, Salt Lake City, UT 84112-0090, USA *E-mail address:* `golden@math.utah.edu`

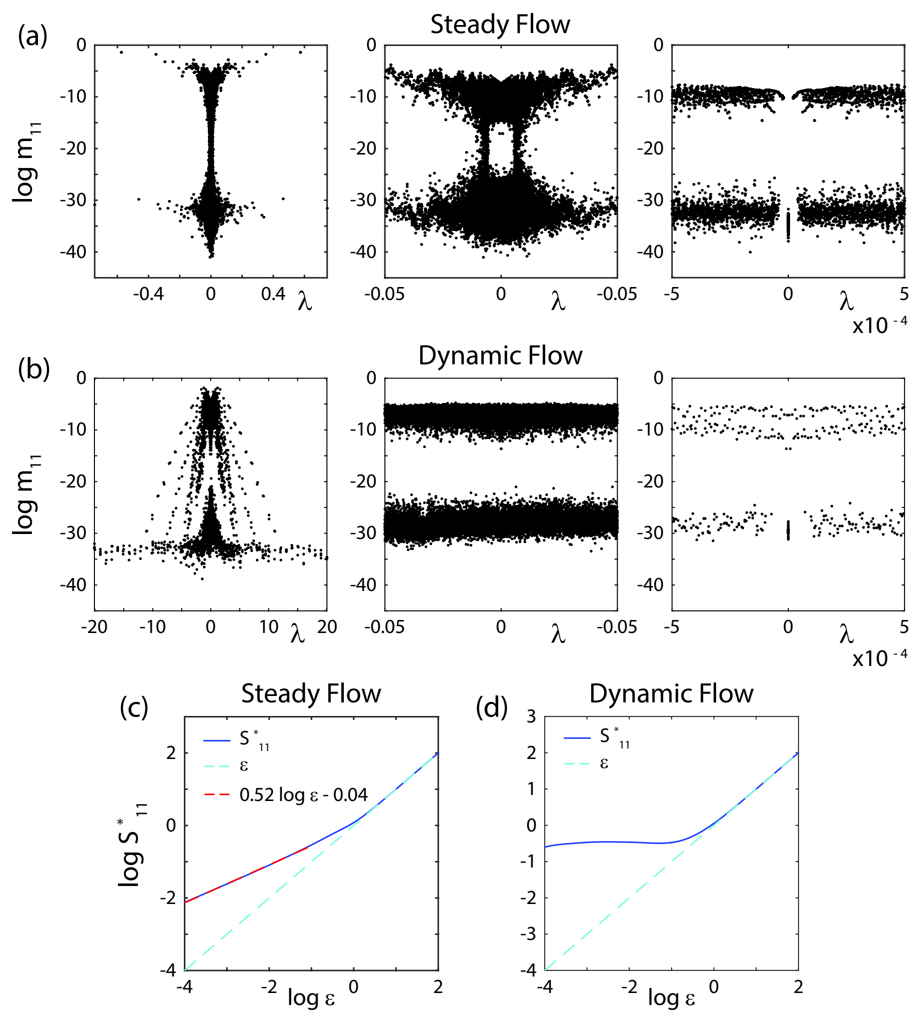

Figure 1: Computations of spectral measures and effective diffusivities for steady and dynamic flows. The spectral measure μ_{11} associated with the flow in (1) are displayed for (a) the steady setting and (b) the dynamic setting with the associated effective diffusivity S^*_{11} displayed in (c) and (d), respectively. In the steady case (a), the limit point of the measure near $\lambda = 0$ has small measure mass with $m_{11} \lesssim 10^{-30}$, leading to the asymptotic behavior $\mathsf{S}^*_{11} \sim \varepsilon^{1/2}$ for $\varepsilon \ll 1$, displayed in (c). In the dynamic case (b), the significant measure mass $m_{11} \gtrsim 10^{-10}$ near $\lambda = 0$ leads to the asymptotic behavior $\mathsf{S}^*_{11} \sim 1$ for $\varepsilon \ll 1$, displayed in (d).