Spectral analysis of mixing in chaotic flows via the mapping matrix formalism: Inclusion of molecular diffusion and quantitative eigenvalue estimate in the purely convective limit

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This paper extends the mapping matrix formalism to include the effects of molecular diffusion in the analysis of mixing processes in chaotic flows. The approach followed is Lagrangian, by considering the stochastic formulation of advection-diffusion processes via the Langevin equation for passive fluid particle motion. In addition, the inclusion of diffusional effects in the mapping matrix formalism permits to frame the spectral properties of mapping matrices in the purely convective limit in a quantitative way. Specifically, the effects of coarse graining can be quantified by means of an effective Péclet number that scales as the second power of the linear lattice size. This simple result is sufficient to predict the scaling exponents characterizing the behavior of the eigenvalue spectrum of the advection-diffusion operator in chaotic flows as a function of the Péclet number, exclusively from purely kinematic data, by varying the grid resolution. Simple but representative model systems and realistic physically realizable flows are considered under a wealth of different kinematic conditions—from the presence of large quasi-periodic islands intertwined by chaotic regions, to almost globally chaotic conditions, to flows possessing “sticky islands”—providing a fairly comprehensive characterization of the different numerical phenomenologies that may occur in the quantitative analysis of mapping matrices, and ultimately of chaotic mixing processes. © 2012 American Institute of Physics.

I. INTRODUCTION

Mixing of liquids is a widespread process that is ubiquitously observed in daily life. The transfer of chemical substances within oceans and in the atmosphere is controlled by mixing and dispersion: Mixing plays also an important role in the maintenance of flora and fauna. Laminar mixing also has a wide application in many industrial processes, including polymer processing and food and pharmaceutical industries. In compounding of various substances, it is necessary to obtain fluid mixtures with specified homogeneity properties, such as necessary for solutions, emulsions, and suspensions. The general mechanisms that make laminar mixing effective, independently of the process being either natural or artificial, are the repetitive stretching and folding of material elements leading to a reduction of the characteristic length scales, and to an increase of the intermaterial contact area. At smaller length scales the effect of molecular diffusion becomes more significant, under the influence of which homogenization of the mixture occurs.

The development of efficient numerical algorithms for simulating chaotic mixing, especially in the presence of diffusion is a central issue in fluid mixing theory. The mapping method, based on...
the distribution matrices, offers an efficient way to investigate the kinematics of mixing for generic mixing protocols in time and spatial periodic flows. A variety of two- and three-dimensional prototypes of flows and mixers has been studied by using the mapping method, including the Kenics and other static mixers, the rotated arc mixer, micromixers, various extruders, etc. In these works, the evolution of a scalar field is studied by the iterative application of the mapping matrix to the vector describing the initial/inlet concentration distribution. From the mathematical point of view, the mapping matrix can be viewed as the coarse-grained representation of the Frobenius-Perron operator associated with the linear advection-diffusion operator. A theoretical investigation of the properties of the Koopman operator, which is the adjoint of the Frobenius-Perron operator, is developed in Mezić et al.

Spectral analysis provides the proper formal setting to explain the asymptotic exponential decay of mixing norms (controlled by the eigenvalues of the transport operators), and the occurrence of repeated mixing patterns (controlled by the corresponding eigenfunctions/eigenvectors) in the linear advection-diffusion operator. Strange eigenmodes were observed experimentally by Rothstein, and further analyzed by Liu and Haller. Both Pierrehumbert and Rothstein supported the observation that strange eigenmodes appear also in aperiodic flows as well.

In a physical perspective, fluid mixing is the result of the interaction of a stirring flow with diffusion. Therefore, it is physically relevant to frame spectral analysis in this perspective, by considering the interaction between an advecting field and molecular diffusion, i.e., by analyzing the eigenvalue/eigenfunction properties of the advection-diffusion operator. Toussaint et al.
were amongst the first that considered spectral properties of the advection-diffusion operator in a three-dimensional chaotic flow. Gleeson applied spectral methods to study simple and chaotic flows in an annular geometry. The group of Giona at "La Sapienza" performed detailed analyses of the strange eigenmodes of the advection-diffusion operator in two-dimensional laminar chaotic mixing flows. In particular, the dependence of the dominant decay exponent $\Lambda$ (see Sec. II for definition and further details) as a function of the Péclet number $Pe$ was investigated for different stirring protocols. The authors found a power dependence of $\Lambda$ on $Pe$, i.e., $\Lambda(Pe) \sim Pe^{-\beta}$, with a corresponding exponent $\beta \in [0; 1]$ that depends on the kinematic features of the stirring flow. Simple non-chaotic autonomous flows were thoroughly investigated, using a Schrödinger-like formulation of the advection-diffusion equation.

The above-mentioned studies investigate the advective-diffusive transport in terms of eigenmode decompositions of the advection-diffusion operator. The connection between the spectral properties of the mapping matrix and the advection-diffusion operator has been revealed in Singh et al. in the purely advective limit of the scalar transport. In this paper, Singh et al. apply the mapping matrix formalism to address the spectral properties of purely advected scalar fields, by focusing mainly on the eigenfunctions of the purely convective mapping matrix and on their symmetries that are related to the structure of the quasi-periodic regions of the Poincaré maps associated with the flow.

The present paper is the natural extension of the Singh paper, and is essentially aimed at a quantitative analysis of the eigenvalues of mapping matrices in order to relate the spectral structure of the mapping matrix to the corresponding properties of the advection-diffusion operator. For this reason, we consider the same flow systems analyzed by Singh et al., namely, the time-periodic sine flow (TPSF), as a paradigmatic example of a bounded chaotic flow, and the partitioned pipe mixer (PPM), as a model system for a realistic and physically realizable open mixing system.

Essentially, the scope of this paper is twofold: (i) to generalize the mapping matrix formalism in order to include the effects of molecular diffusion; (ii) to analyze the numerical diffusion effects induced by the coarse-graining in the discretized representation of the mapping matrix in the purely.
II. STATEMENT OF THE PROBLEM

As a relevant byproduct of this analysis, we show that even when the purely convective case is considered, numerical diffusion can be exploited to mimic molecular diffusion, and a strict quantitative correspondence between the spectral structure of the purely convective mapping matrix and that of the continuum advection-diffusion operator can be defined, at least as regards the first dominant eigenvalues/eigenfunctions. More precisely, for any coarse-graining resolution, i.e., for any size of the discretized grid defining a given mapping-matrix approximation, an effective Péclet number can be defined (that is associated with numerical diffusion), so that the mapping matrix at this level of resolution can be viewed as a quantitatively reliable approximation of the continuum advection-diffusion operator for a value of the Péclet number equal to the effective Péclet number of the grid resolution. In addition, we show that the effective Péclet number is proportional to the square of the linear grid size (see Sec. III for details).

The paper is organized as follows. Section II defines the problem setting, and briefly reviews the spectral characterization of mixing, the mapping matrix formalism as a way to addressing the Ulam’s approximation of the Frobenius-Perron operator. Section III describes how molecular diffusion can be accounted for in the mapping-matrix formalism. Specifically, a stochastic formulation of advection-diffusion processes is considered. Moreover, the quantitative characterization of numerical diffusion induced by coarse-graining is introduced via the concept of effective Péclet number. Section IV analyzes the spectral properties of the purely kinematic mapping matrix, and relates the effective Péclet number to the lattice size. By enforcing this relation, we show that it is possible to predict the scaling of the spectral branches (specifically of the dominant decay exponent) from purely convective simulations. Section V analyzes the spectral properties of the diffusive mapping matrix and discusses several approaches to estimate the prefactor of the relation connecting the effective Péclet number to the square of the linear lattice size. Section VI analyzes further the purely kinematic case, and shows that a quantitative estimate of the first eigenvalues of the continuum advection-diffusion operator can be obtained from the purely convective mapping matrix. Moreover, the case of a flow protocol possessing long-range correlations near “sticky” quasi-periodic regions and parabolic fixed points is addressed, since it gives apparently “pathological” results. Finally, Sec. VII discusses the potentialities of the present analysis. Specifically, the mapping formalism (with or without diffusion) can be applied without difficulty to generic physically realizable mixing systems, be them closed or open, and can be exploited to simulate advection-diffusion processes for very high Péclet number, never reached so far in the numerical simulation of chaotic mixing flows. Moreover, the algorithm proposed can be applied to generic random perturbations of a deterministic chaotic dynamics, including the case of stochastic perturbations possessing long-range correlations.

II. STATEMENT OF THE PROBLEM

This section defines the problem setting and provides a concise overview of the technical tools applied throughout the paper.

Let \( c(x, t) \) be a scalar field associated with a physical transported entity \( C \) (a diffusing solute, temperature, etc.) defined in a mixing domain \( \mathcal{M} \), and advected by a deterministic laminar incompressible flow \( \mathbf{V}(x, t), \nabla \cdot \mathbf{V} = 0 \). Let \( D \) be the diffusivity of \( C \) in the fluid continuum. The evolution of \( C \) within \( \mathcal{M} \) is described by the advection-diffusion equation

\[
\frac{\partial c}{\partial t} = -\nabla \cdot (\mathbf{V} c) + D \nabla^2 c
\]  

(1)

for its concentration field \( c \). Let \( c_{\text{ref}} \) be a reference value for \( c \), \( L_0 \) a characteristic length-scale of the mixing domain \( \mathcal{M} \), and \( V_0 \) a characteristic velocity scale. Introducing \( u(x, t) = c(x, t)/c_{\text{ref}} \), \( \mathbf{v}(x, t) = \mathbf{V}(x, t)/V_0 \), and \( t V_0/L_0 \mapsto t, x/L_0 \mapsto x \), the non-dimensional form of Eq. (1) reads as

\[
\frac{\partial u}{\partial t} = -\nabla \cdot (\mathbf{v} u) + \frac{1}{P_e} \nabla^2 u,
\]  

(2)
where \( Pe = V_0 L_0 / D \) is the Péclet number. For notational simplicity, we still use the symbols \( t \) and \( \mathbf{x} \) to indicate the non-dimensional time and position vector, as well as the nabla operator \( \nabla \) entering Eq. (2) refers to the non-dimensional coordinates. Henceforth, unless otherwise specified, solely non-dimensional variables are used.

The mixing domain \( \mathcal{M} \) can be bounded or unbounded. In closed bounded systems, mixing quantification can be obtained by considering the norm decay of the field \( u(\mathbf{x}, t) \) as a function of time \( t \). Since in a closed and bounded system the normal component of the flux vanishes at the boundary of \( \mathcal{M} \), the mean value \( \bar{u}(t) = (1 / \text{meas}(\mathcal{M})) \int_{\mathcal{M}} u(\mathbf{x}, t) \, d\mathbf{x} \) where \( \text{meas}(\mathcal{M}) \) is the Lebesgue measure of \( \mathcal{M} \) is conserved, i.e., \( \bar{u}(t) = \bar{u}(t = 0) = \bar{u}_0 \) for any \( t > 0 \). It is convenient to define a new field \( \phi(\mathbf{x}, t) = u(\mathbf{x}, t) - \bar{u}_0 \) possessing zero mean. The field \( \phi(\mathbf{x}, t) \) satisfies the same advection-diffusion equation defining the evolution of \( u(\mathbf{x}, t) \), i.e., Eq. (2) and its \( L^2 \)-norm,

\[
||\phi||_{L^2}^2(t) = \left[ \int_{\mathcal{M}} \phi^2(\mathbf{x}, t) \, d\mathbf{x} \right]^{1/2},
\]

decays to zero as

\[
dt ||\phi||_{L^2}^2(t) = - \frac{2}{Pe} ||\nabla \phi||_{L^2}^2(t),
\]

where \( ||\nabla \phi||_{L^2}^2(t) \) is the \( L^2 \)-norm of the gradient of \( \phi \).

In open mixing systems, such as a mixing tube (in Sec. VII we consider the case of the partitioned pipe mixer), the mixing domain \( \mathcal{M} \) can be represented as the Cartesian product \( \mathcal{M} = (0, 1) \times \Sigma_\perp \), where \( \Sigma_\perp \) is the transverse cross section of the tube. In many applications of open flow devices steady-state mixing properties are particularly relevant.

Let \( \mathbf{x} = (x_\perp, z) \) where \( x_\perp \) and \( z \) are, respectively, the transverse and axial coordinates, and \( \mathbf{v} = (v_\perp, w) \) be the analogous decomposition for the non-dimensional velocity field. For high Péclet numbers, if the characteristic transverse length is much smaller than the axial length of the device, axial diffusion is negligible. Therefore, at steady state Eq. (2) simplifies as

\[
w \frac{\partial u}{\partial z} = - v_\perp \cdot \nabla_\perp u + \frac{1}{Pe} \nabla_\perp^2 u,
\]

where \( \nabla_\perp \) is the transverse nabla operator and incompressibility has been enforced. Let us assume \( w = w(x_\perp) \) to be independent of the axial coordinate (as for Stokes flows in straight channels), and that \( w(x_\perp) \geq 0 \). Under these assumptions, Eq. (5) is similar to the time-evolution of a scalar field in a two-dimensional mixing domain (in this analogy represented by the cross section \( \Sigma_\perp \)), where the time variable is replaced by the axial coordinate weighted with respect to the axial velocity component.

Let \( \phi = u - \bar{u}_w \), where \( \bar{u}_w(z) = (1 / \text{meas}(\Sigma_\perp)) \int_{\Sigma_\perp} w(x_\perp) u(x_\perp, z) \, dx_\perp \) is the weighted average of \( u \) with respect to the axial coordinate. Since the flux vanishes at the boundary of the cross section, from Eq. (5) it follows that \( \bar{u}_w(z) = \bar{u}_{w,0} = \text{constant}. \) Therefore, \( \phi(x, z) \) satisfies Eq. (5) and

\[
dt ||\phi||_{L^2}^2(z) = - \frac{2}{Pe} ||\nabla_\perp \phi||_{L^2}^2(z),
\]

where

\[
||\phi||_{L^2}(z) = \left[ \int_{\Sigma_\perp} w(x_\perp) \phi^2(x_\perp, z) \, dx_\perp \right]^{1/2}, \quad ||\nabla_\perp \phi||_{L^2}^2(z) = \left[ \int_{\Sigma_\perp} |\nabla \phi(x_\perp, z)|^2 \, dx_\perp \right]^{1/2}
\]

that represents the analogous of Eq. (4) for open mixing system at steady state possessing axial symmetry.

### A. Spectral analysis of mixing

Consider a closed mixing system in the presence of a time-periodic velocity field \( \mathbf{v}(\mathbf{x}, t + T_p) = \mathbf{v}(\mathbf{x}, t) \), where \( T_p > 0 \) is the flow period. For time-periodic flows, Eq. (2) is a parabolic equation with time-periodic coefficients. In order to obtain an autonomous (time-independent) representation
of the concentration field dynamics, it is convenient to sample its evolution at time instants multiple of the flow period. Therefore, the stroboscopic operator \( \mathcal{P} : L^2(\mathcal{M}) \to L^2(\mathcal{M}) \) can be defined from Eq. (2)–\( L^2(\mathcal{M}) \) is the space of square summable functions defined in \( \mathcal{M} \)–in such way that it transforms the scalar field \( u_n(x) = u(x, nT_p) \) solution of Eq. (2) at time \( t = nT_p \) into the field \( u_{n+1}(x) = u(x, (n+1)T_p) \) after one flow period,

\[
u_{n+1}(x) = \mathcal{P}[u_n(x)]. \tag{8}\]

The operator \( \mathcal{P} \) is essentially the Floquet (or Poincaré) operator associated with an advection-diffusion equation with time-periodic coefficients.\(^{29}\)

In the case \( Pe^{-1} = 0 \) (i.e., in the purely convective limit), it is more convenient\(^{17}\) to regard \( \mathcal{P} : L^1(\mathcal{M}) \to L^1(\mathcal{M}) \) as an operator acting on \( L^1(\mathcal{M}) \) since \( L^2(\mathcal{M}) \subseteq L^1(\mathcal{M}) \) if \( \mathcal{M} \) is bounded and possesses finite measure. Let \( \Phi : \mathcal{M} \to \mathcal{M} \) be the Poincaré map of the time-periodic incompressible kinematics, \( dx/dt = \mathbf{v}(\mathbf{x}, t) \). The map \( \Phi \) is measure-preserving and at \( Pe^{-1} = 0 \) the Floquet operator can be expressed as

\[
\mathcal{P}[u_n(x)] = u_n(\Phi^{-1}(x)), \tag{9}\]

i.e., if \( \mathcal{P} \) reduces to the Frobenius-Perron operator associated with the fluid kinematics.

For any \( Pe < \infty \), the operator \( \mathcal{P} \) is compact, and therefore it admits a countable spectrum of eigenvalues \( \{\nu_n\}_{n=1}^{\infty} \), lying in the unit circle, that can be ordered in a non-increasing way with respect to the moduli, \( |\nu_1| > |\nu_2| \geq |\nu_3| \geq \cdots |\nu_n| \geq \cdots \). Let \( \{\psi_n(x)\}_{n=1}^{\infty} \) be the corresponding eigenfunctions. Due to conservation of the transported entity, the first eigenvalue equals 1, i.e., \( \nu_1 = 1 \), and the corresponding eigenfunction is uniform, \( \psi_1(x) = const \) for \( x \in \mathcal{M} \). Starting from \( \nu_2 \), all the other eigenvalues admit norms strictly less than 1. Therefore, for generic initial conditions (i.e., apart from a set of initial conditions of zero measure), the norm decay of scalar fields transported according to Eq. (2) is controlled by the second eigenvalue \( \nu_2 \) (referred to, for this reason, as the dominant eigenvalue). Specifically, since \( \phi_0(x) = u_0(x) - \langle u_0 \rangle \) possesses zero mean, then

\[
||\phi_n||_{L^2} \sim |\nu_2|^n e^{-\nu_2 T_p}, \tag{10}\]

where the exponent \( \Lambda \) will be referred to as the dominant decay exponent of the time-periodic advection-diffusion operator,

\[
\Lambda = -\frac{\log |\nu_2|}{T_p}. \tag{11}\]

**B. Mapping matrix formalism**

The mapping method is based on a suggestion due to Spencer and Wiley\(^{34}\) that describes the transport of a conservative quantity from one state to another by means of a discretized mapping represented, on a finite grid, by a matrix. In their 1951 paper, Spencer and Wiley clearly state their idea: "...Now define a distribution matrix \( [D_{ij}] \), where \( D_{ij} \) is the fraction of material originating in the \( i \)th cell which is to be forced in the \( j \)th cell after the mixing operation.”

Several decades after this suggestion, a wealth of papers applied to different prototypical, industrial, and micro-mixers have shown that the mapping approach (i.e., the coarse-grained discretization of the action of a convective mixing field onto a discretized concentration field) provides an efficient computational tool for obtaining an accurate characterization of mixing properties in the purely kinematic case at a feasible computational cost.\(^{15,35,36}\) The entries of this matrix contain the fractions of material from one part of the domain which is transferred to various parts of the domain when specific flow is applied. Figure 1 depicts how the entries of the mapping matrix are approximated.

The flow domain is divided into a number of cells \( N \) which are numbered sequentially and \( N_m \) markers are uniformly placed inside each cell and tracked in time from \( t = t_0 \) to \( t = t_0 + \Delta t \). If the number of markers in the donor cell \( \Omega_j \) is \( N_j = N_m \) at \( t = t_0 \) and the number of markers found after tracking in the recipient cell \( \Omega_i \) is \( N_i \) at \( t = t_0 + \Delta t \), then the entry \( M_{ij} \) of the mapping matrix \( \mathbf{M} \) is
FIG. 1. Illustration of the computation of the entries $M_{ij}$ of the mapping matrix $M$. The cell $\Omega_j$ at $t = t_0$ is covered with a number of markers that are tracked during flow in $\Delta t$ to arrive at the final cross section $t = t_0 + \Delta t$. The ratio of the number of markers received by the recipient cell $\Omega_i$ to the initial number of markers in $\Omega_j$ is determined. In this case $M_{ij} = 3/16$.

The ratio of the number of markers received by the recipient cell $\Omega_i$ to the initial number of markers in $\Omega_j$ is determined. In this case $M_{ij} = 3/16$.

Calculated\(^{36}\) as

$$M_{ij} = \frac{N_{ij}}{N_j}. \quad (12)$$

Once the mapping matrix is evaluated over the period of the flow protocol, the evolution of the concentration field at discrete time intervals multiple of the period can be estimated as

$$C_n = (M(M(...(MC_0)...))^n, \quad (13)$$

where $C_0$ is a $N$-dimensional vector representing the initial concentration inside the flow domain.

The vector $C_n$ provides the coarse-grained description of the volume fraction (dimensionless concentration) of the transported entity in the moving continuum, and its $i$th entry corresponds to the concentration locally averaged at the cell $\Omega_i$.

The mapping matrix approach, envisaged by Spencer and Wiley, corresponds essentially to the same idea proposed by Ulam\(^{18}\) and usually referred to as the Ulam’s method or the Ulam’s approximation of the Frobenius-Perron operator\(^{17}\) associated with a deterministic dynamical system. Let $\Phi : M \rightarrow M$ be a discrete dynamical system on a bounded subset $M \subset R^d$ or on a $d$-dimensional compact (boundaryless) manifold. The Ulam’s method consists in decomposing the domain $M$ into a finite system of disjoint subsets $\{\Omega_i\}_{i=1}^N$, $\bigcup_{i=1}^N \Omega_i = M$, $\Omega_i \cap \Omega_j = \emptyset$ for $i \neq j$, and approximating the Frobenius-Perron operator associated with $\Phi$ by the matrix

$$M_{ij}^U = \frac{\text{meas}(\Omega_i \cap \Phi^{-1}(\Omega_j))}{\text{meas}(\Omega_j)}, \quad (14)$$

where $\text{meas}(\Omega_j)$ is the $d$-dimensional Lebesgue measure of $\Omega_j$, and $M_{ij}^U$ are the entries of the Ulam’s matrix. Equation (12) corresponds to the computation of the Ulam’s matrix using a swarm of $N_m$ particles to approximate the measures entering the numerator and the denominator of Eq. (14).

The suggestion of Spencer and Wiley and the method proposed by Ulam essentially coincide and originated a cascade of highly valuable research contributions both in physical fluid-dynamic oriented literature (accounted above) and in the mathematical/numerical theory of dynamical systems. In the mathematically oriented literature, Ulam’s method and its convergence properties has been first tackled by Li\(^{37}\) that proved the convergence of the Ulam’s method to the unique absolutely continuous invariant measure for a class of interval maps, and subsequently by many other authors\(^{19,20,38-41}\) that extended the analysis to higher dimensional dynamical systems and applied Ulam’s method to determine entropies and other dynamical invariants.\(^{42}\)

It is rather remarkable that these two lines of research essentially focused on the same topic, that can be referred to in a “fair” way as the Spencer-Wiley-Ulam method to study dynamical systems using a coarse-grained representation of the evolution equation for densities has proceeded for
decades along parallel pathways without finding a common intersection/exchange point that could have been fruitful both for the physical understanding and for the mathematical formalization of the problem.

C. Model systems

As a benchmark model we consider the TPSF.\textsuperscript{43, 44} TPSF is defined as periodic sequences of two steady sinusoidal flows, which act during the half of the period $T_p$ inside the unit bi-periodic square $\Omega = (0, 1) \times (0, 1)$,

$$\begin{align*}
v_1 &= (v_x, v_y) = (\sin (2\pi y), 0) \quad \text{for} \quad 0 \leq t < \frac{T_p}{2}, \\
v_2 &= (v_x, v_y) = (0, \sin (2\pi x)) \quad \text{for} \quad \frac{T_p}{2} \leq t < T_p.
\end{align*}$$

The qualitative kinematic properties of TPSF protocols (obtained by varying the period $T_p$) are analogous to those observed in physically realizable flow systems. Therefore, the use of TPSF allows to gather valuable information on fluid mixing under a wealth of different kinematic conditions at moderate computational costs. TPSF admits an analytical expression for fluid particle orbits. Its Poincaré map is given by

$$\begin{align*}
\Phi(x_i, y_i) &= \begin{cases} 
x_i = x_{i-1} + T_p \sin (2\pi y_{i-1})/2 & \text{mod. 1,} \\
y_i = y_{i-1} + T_p \sin (2\pi x_i)/2 & \text{mod. 1,}
\end{cases}
\end{align*}$$ (16)

and the Floquet operator $\mathcal{P}$ associated with TPSF can be expressed as\textsuperscript{29}

$$\mathcal{P} [u_n(x)] = e^{(-v_1 \cdot \nabla + Pe^{-1}\nabla^2)T_p/2} \circ \left( e^{(-v_2 \cdot \nabla + Pe^{-1}\nabla^2)T_p/2} [u_n(x)] \right),$$ (17)

where “$\circ$” indicates operator composition. The eigenvalues of $\mathcal{P}$ obtained by Fourier-series expansion has been studied by Cerbelli,\textsuperscript{29} and we use the data reported in this paper (together with new data obtained using the same Fourier expansion as in Cerbelli et al.\textsuperscript{29}) to compare the spectral properties of the mapping matrix with those of the advection-diffusion operator. Let $v_2$ be the second eigenvalue of $\mathcal{P}$. For a given $Pe$, the dominant decay exponent of the continuum advection-diffusion operator will be indicated with the symbol $\Lambda(\mathcal{P}) = -\log |v_2|/T_p$.

As a model for a realistic, physically realizable chaotic open flow, we consider the PPM, which consists of a tube partitioned into a sequence of semi-circular ducts by means of rectangular plates placed orthogonally to each other. As in Singh et al.,\textsuperscript{33} we use the approximation for the velocity field proposed by Khakhar et al.\textsuperscript{45} The non-dimensional parameter characterizing this mixer is the ratio $\beta_{ppm} = 4v_{tr}L/3\gamma(v_c)R$, where $v_{tr}$ is the characteristic transverse velocity, $\langle v_c \rangle$ is the mean axial velocity, $L$ the length of each pipe element, $R$ the pipe radius, and $\gamma = (11/3)^{1/2} - 1$.

III. MAPPING MATRIX DESCRIPTION OF ADVECTION-DIFFUSION PROCESSES

One of the objectives of the present paper is to generalize the mapping matrix formalism in order to include the effects of molecular diffusion. This can be achieved by considering the Langevin formulation of Brownian motion in the presence of a deterministic bias (i.e., of a convective field), and by enforcing the correspondence between stochastic differential equations and advection-diffusion processes.\textsuperscript{17, 46}

This section describes the stochastic construction of the mapping matrix in the presence of diffusion, its statistical characterization, and some general observations regarding its spectral structure.

A. Langevin construction of the diffusive mapping matrix

Consider a $d$-dimensional flow ($d = 2, 3$) associated with a time-periodic non-dimensional velocity field $\mathbf{v}(\mathbf{x}, t + T_p) = \mathbf{v}(\mathbf{x}, t)$, defined on a bounded domain $\mathcal{M}$.
The motion of a passive fluid particle, initial located at \( x_0 \in \mathcal{M} \), under the influence of the flow and of random fluctuations is described by the non-dimensional stochastic differential equation\(^{17,46,47}\)

\[
\frac{dx(t)}{dt} = v(x(t), t) dt + \sqrt{\frac{2}{Pe}} d\xi(t),
\]

where \( d\xi(t) \) are the infinitesimal increments of a \( d \)-dimensional Wiener process, i.e., of a vector-valued stochastic process \( \xi(t) = (\xi_1(t), \ldots, \xi_d(t)) \) in which \( \xi_h(t) \) and \( \xi_k(t) \) are independent stochastic processes for \( h \neq k \), possessing uncorrelated increments distributed in a normal way, i.e., \( \langle \xi_h(t) \rangle = 0 \) and \( \langle (\xi_h(t + \tau) - \xi_h(t))^2 \rangle = \tau \), for any \( h = 1, \ldots, d \) and \( t, \tau > 0 \), and such that \( \xi(0) = 0 \). Equation (18) is equipped with the initial condition \( x(0) = x_0 \). Let \( P(x, t|x_0) \) be the probability density function (pdf) associated with the stochastic process (18). For notational simplicity, we indicate this random variable. This means that the

\[
\text{P} \text{éclet number.}
\]

\[
\text{Equation (19) corresponds to the classical non-dimensional advection-diffusion equation, where } Pe
\]

\[
\text{is the Péclet number.}
\]

The stochastic differential equation (18) can be solved numerically by means of an Euler-Langevin explicit scheme.\(^{17}\) Let \( h \) be the time step, \( x_n = x(nh) \), and \( v_n(x) = v(x, nh) \). The Euler-Langevin solver for Eq. (18) is simply given by

\[
x_{n+1} = x_n + v_n h + \sqrt{\frac{2}{Pe}} h^{1/2} r_n,
\]

where \( r_n = (r_{1,n}, \ldots, r_{d,n}) \) is the realization of a \( d \)-dimensional normally distributed independent random variable. This means that the \( d \) random variables \( r_{1,n}, \ldots, r_{d,n} \), are independent of each other and distributed according to a Gaussian pdf with zero mean and unit variance. A further discussion on the numerical methods for solving Eq. (18) is addressed in Sec. VII.

By making use of the stochastic formulation of advection-diffusion processes, the mapping matrix can be constructed by following exactly the same computational strategy adopted in the purely convective case. Specifically:

- The mixing domain \( \mathcal{M} \) is discretized with a structured grid of \( N = N_c^d \) boxes (\( N_c \times N_c \) for two-dimensional simulations).
- Within each box, say the \( i \)th, \( N_p^d \) particles are considered, uniformly distributed at internal points. Let \( x_p^{i,j} \) be the position vector of the \( j \)th particle \( j = 1, \ldots, N_p^d \).
- For each \( x_p^{i,j} \), let \( x_0^{i,j} = \Phi^{-1}(x_p^{i,j}) \) the effective initial position, where \( \Phi^{-1} \) is the inverse of the kinematic Poincaré map.
- The Langevin equation (18) is solved according to Eq. (20) for a time interval \( T_p \), equal to the flow period, starting from the initial condition \( x_0^{i,j} \). Let \( X_f = \{ x_f^{i,j} \}_{j=1}^{N_p^d} \) the ensemble of the final positions for the \( (N_c \times N_p)^d \) particles considered.
- The mapping matrix \( \mathbf{M} \in \mathcal{M}_{N_f^d \times N_f^d} \) is evaluated in the usual way, i.e., by means of Eq. (12), where \( N_f^d \) is the number of particles in the ensemble \( X_f \) that fall within the \( i \)th box of the discretization, given that their initial position is within the \( j \)th box, and \( N_f^d \) is the number of initial positions falling within the \( j \)th box.

Therefore, the mapping matrix can be viewed as a coarse-grained representation of the Floquet operator \( \mathcal{P} \) associated with an advection-diffusion process.

For any finite value of \( Pe \), the mapping matrix obtained by means of the method described above will be indicated as the diffusive mapping matrix. The matrix obtained in the limit case of \( Pe \rightarrow \infty \), i.e., when no stochastic contribution is superimposed to deterministic convection induced by the flow, will be referred to as the purely convective mapping matrix.

Let \( \{ \lambda_j \} \) be the eigenvalues of \( \mathbf{M} \), that lie within the unit circle. The eigenvalues can be ordered in a decreasing way with respect to their moduli. For a closed mixing system, the first eigenvalue
equals 1 and this corresponds to the conservation of the transported entity. The second eigenvalue $\lambda_2$ is less than one, and, within the mapping matrix formalism is related to the dominant decay exponent $\Lambda_{mp}$, via the relation

$$\Lambda_{mp} = -\frac{\log |\lambda_2|}{T_p},$$  \hspace{1cm} (21)$$

where $T_p$ is the flow period. Henceforth, we indicate with the subscript “mp” the dominant decay exponent obtained from the mapping matrices, and with $\Lambda$ the corresponding exponent obtained from the spectral analysis of the continuous advection-diffusion operator, obtained, e.g., by Fourier methods as in Cerbelli et al.\textsuperscript{29}

**B. Statistical analysis of the mapping matrix**

In the presence of diffusion, the mapping matrix obtained via the stochastic Langevin equation (19) is a stochastic process itself. This section investigates its statistical properties as it regards its spectral structure with a focus on its second eigenvalue or equivalently on the associated dominant decay exponent $\Lambda_{mp}$. The analysis of higher-order eigenvalues and of the general spectral structure is discussed in Sec. III C. The eigenvalue convergence of a class of linear stochastic systems has been analyzed by De Doná et al.\textsuperscript{48} by considering one-dimensional models driven by uncorrelated random perturbations as a function of the state discretization.

For the sake of simplicity, we consider in this section two-dimensional systems such as the TPSF on the unit two-dimensional torus. The construction of the mapping matrix depends essentially on the two integers: $N^2$, expressing the total number of boxes in the discretization of the mixing domain, and $N^2_p$ corresponding to the total number of particles per box. In point of fact, the algorithm for generating $M$ based on Eq. (20) depends also on the time step $h$. A preliminary analysis of the effect of the time step $h$ (not reported for the sake of brevity) indicates that the choice $h = 10^{-2}$ provides, for the TPSF, a reliable trade-off between accuracy and computational speed in the simulation of the Langevin equation using the Euler-Langevin algorithm (20).

As $N_p$ increases, a statistical averaging over a larger ensemble of fluid particle trajectories is performed, so that it is to be expected that the effects of stochastic fluctuations on the spectral properties of $M$ become less pronounced as $N_p$ increases. We performed the statistical analysis by considering an ensemble of $N_e = 10^3$ realizations of the diffusive mapping matrix. Let $\langle \Lambda_{mp} \rangle$ and $\sigma^2_\lambda$ be the mean and the variance of the dominant decay exponent $\Lambda_{mp}$, respectively, obtained with respect to the ensemble of $N_e$ realizations of $M$: $\langle \Lambda_{mp} \rangle = N_e^{-1} \sum_{\alpha=1}^{N_e} \Lambda_{mp,\alpha}$, $\sigma^2_\lambda = N_e^{-1} \sum_{\alpha=1}^{N_e} (\Lambda_{mp,\alpha} - \langle \Lambda_{mp} \rangle)^2$, where $\Lambda_{mp,\alpha}$ is the dominant decay exponent of the $\alpha$th realization. Results are limited to $N_e = 65$ as for higher grid resolutions the computational cost in evaluating the spectral structure of $M$ matrices becomes prohibitive.

Figure 2 panel (a) depicts the behavior of $\langle \Lambda_{mp} \rangle$ vs $N_p$ for two values of the flow period at $N_e = 50$, $Pe = 10^3$. The horizontal lines correspond to the values of $\Lambda(Pe)$ obtained by means of Fourier analysis of the advection-diffusion equation. The value of the Péclet number ($Pe = 10^3$) is chosen so that the effect of numerical diffusion for this grid resolution ($N_e = 50$) is negligible, see Figure 9 and the analysis developed in Sec. III C.

Starting from $N_p \geq 10$ the average value of $\Lambda_{mp}$ practically coincides with that of $\Lambda(Pe)$. More interesting is the scaling of $\sigma_\lambda$ (depicted in panel (b)) for $T_p = 0.8$ and $N_e = 50$ for several values of $N_p$. As can be observed

$$\sigma_\lambda \sim N_p^{-1},$$  \hspace{1cm} (22)$$

which can be justified by means of elementary statistical mechanical considerations\textsuperscript{49,50} (we return to this issue below).

In order to provide a direct quantification of the statistical error induced by stochasticity in the construction of $M$, it is convenient to consider the normalized squared root of the
FIG. 2. Panel (a): $\langle \Lambda_{mp} \rangle$ vs the number of particles per unit interval $N_p$ for the TPSF at $Pe = 10^3$ and $N_c = 50$: symbols (●) refers to $T_p = 0.8$, (□) to $T_p = 1.6$. The horizontal lines represent the corresponding values of $\Lambda_{mp}$ obtained by means of Fourier analysis. Panel (b): $\sigma / \Lambda_1$ vs $N_p$ for the TPSF at $T_p = 0.8$, $Pe = 10^3$. The arrow indicates increasing values of $N_c$: (□) $N_c = 15$, (■) $N_c = 30$, (○) $N_c = 50$, (●) $N_c = 65$. Lines (A) and (B) represent the scaling $\sigma / \Lambda_1 \sim N_p^{-1}$. Panel (c): $\sigma^*/\Lambda_1$ vs $N_p$ for several TPSF protocols: (□) $T_p = 0.8$, $Pe = 10^3$, $N_c = 30$, (■) $T_p = 0.8$, $Pe = 10^3$, $N_c = 30$, (○) $T_p = 1.18$, $Pe = 10^3$, $N_c = 30$, (●) $T_p = 1.6$, $Pe = 10^3$, $N_c = 50$. Panel (d): Prefactor $s^*/\Lambda_1$ of the normalized variance vs $N_c$ for the TPSF at $T_p = 0.8$, $Pe = 10^3$. The solid line represents the scaling $s^*/\Lambda_1 \sim N_c^{-1}$.

variance $\sigma^*_\Lambda$, namely,

$$\sigma^*_\Lambda = \frac{\sigma_\Lambda}{\langle \Lambda_{mp} \rangle}, \quad (23)$$

which is depicted in panel (c) for several values of the flow period and of the Péclet number. From Eq. (22), it follows that $\sigma^*_\Lambda \sim N_p^{-1}$ and this power-law scaling sets in starting from low values of $N_p \geq 5$. Regarding the values attained by $\sigma^*_\Lambda$, starting from $N_p = 20$, $\sigma^*_\Lambda < 10^{-2}$ and for $N_p = 100$ its numerical value ranges between $8 \times 10^{-4}$ and $2 \times 10^{-3}$. This means that, for $N_p = 100$, the error in the estimate of $\Lambda_{mp}$ is order of 0.1%–1% of the value of the dominant decay exponent. This indicates that, at least for the TPSF, from a single realization of the diffusive mapping matrix, the statistical error in the estimate of $\Lambda_{mp}$ can be kept well below 1% provided that $N_p$ is taken large enough $N_p \geq 50$. Throughout this paper, all the simulations of the TPSF refers to $N_p = 100$, and this is the reason why in numerical analysis of $\Lambda_{mp}$ presented in the remainder of this paper, the value of the dominant decay exponent has been estimated from a single realization of the mapping matrix without reporting the error bars.

Let $s_\Lambda$ and $s^*_\Lambda$ be the prefactors of the scalings of $\sigma_\Lambda$ and $\sigma^*_\Lambda$, respectively, as a function of $N_p$ ($N_p \geq 5$), i.e., $\sigma_\Lambda = s_\Lambda / N_p$, $\sigma^*_\Lambda = s^*_\Lambda / N_p$. The information on the statistical properties of $\sigma_\Lambda$ and $\sigma^*_\Lambda$ can be visualized in a compact way by depicting the behavior of the prefactor $s^*_\Lambda$ as a function of...
FIG. 3. Invariant rescaling of $\sigma^*_\Lambda$ vs $N_c \times N_p$ for several TSPC protocols at a fixed value of $Pe = 10^3$: (□) $T_p = 0.8, N_c = 15$; (■) $T_p = 0.8, N_c = 30$; (●) $T_p = 0.8, N_c = 65$; (△) $T_p = 1.6, N_c = 30$; (▲) $T_p = 1.6, N_c = 50$; (▽) $T_p = 1.18, N_c = 30$. The solid line represents the scaling $\sigma^*_\Lambda = a/(N_c \times N_p)$ with $a = 5$.

The dependence of the variance $\sigma_\Lambda$ on the Péclet number is depicted in Figure 4 by considering a typical situation ($T_p = 0.8, N_c = 30$), reporting the behavior of the two prefactors $s_\Lambda$ and $s^*_\Lambda$. While, as expected, $s_\Lambda$ is a decreasing function of $Pe$, since the effect of the stochastic diffusive contribution on the fluctuations is more pronounced at low Péclet values. Conversely, $s^*_\Lambda$ can increase or decrease with $Pe$ depending on the scaling of $\langle \Lambda_{mp} \rangle$ as a function of $Pe$. Up to $Pe < 10^3$, anticipating a result thoroughly addressed in Sec. III C, the effect of numerical diffusion induced by coarse-graining is more pronounced at low Péclet values.
practically negligible. In this region of Pe-values, the scaling $s_{\Lambda} \sim Pe^{-1/2}$ can be observed (line (c) in Figure 4). A deviation from this scaling occurs at higher Pe values, i.e., when the effect of numerical diffusion controls the spectral structure of $\mathbf{M}$.

To complete the statistical analysis of the dominant spectral properties, consider the probability density function $p_{\Lambda}(\xi)$ of the rescaled dominant decay exponent $\xi = (\Lambda_{mp} - \langle \Lambda_{mp} \rangle) / \sigma_{\Lambda}$. The numerical results for $p_{\Lambda}(\xi)$ obtained from an ensemble of $10^{3}$ realizations of $\mathbf{M}$ are depicted in Figure 5 for several values of $N_{p}$. As can be observed, the numerical data are distributed in a normalized Gaussian way, i.e., $\sigma_{\Lambda} p_{\Lambda}(\xi) = e^{-\xi^{2}/2}/\sqrt{2\pi}$.

Although the construction process of the purely convective mapping matrix is strictly deterministic, its numerical value depends on $N_{p}$, i.e., on the number of particles per box used in the simulations. Figure 6 depicts the behavior of the dominant decay exponent (the superscript “c” indicates the purely convective case) obtained for the purely convective mapping matrix for several typical values of the flow period, and grid resolutions.

The typical situation, occurring for flow protocols the Poincaré maps of which possess either large islands of quasi-periodicity ($T_{p} = 0.8$ corresponding to symbols □ and ■, and lines (a) and (b))
FIG. 7. Panel (a): $\langle \Lambda_{mp} \rangle$ vs $N_p$ for the TPSF at $T_p = 0.8$, $Pe = 10^3$. Line (A) and (□) dominant (second), line (B) and (○) third, line (C) and (●) fourth decay exponent. The solid horizontal lines correspond to the values obtained by Fourier analysis. Panel (b): $\sigma_{\Lambda}^*$ vs $N_p$ for the second third and fourth decay exponent (symbols as in panel (a)). Lines (A) and (B) correspond to the scaling $\sigma_{\Lambda}^* \sim N_p^{-1}$.

or seemingly globally chaotic conditions ($T_p = 1.6$ corresponding to symbols ○ and ●, and lines (c) and (d)), is that it is sufficient to take $N_p = 50$ (i.e., 2500 particles per discretization box) to achieve an accurate estimate of the mapping matrix. However, situations occur in which this may not be sufficient. This is, for example, the case of the TPSF at $T_p = 2.0$ (symbols ▽, ▲, △, and lines (e) to (g)), for which at least $N_p = 100$ is required for convergence. This case corresponds to a Poincaré map possessing small islands of quasi-periodicity or parabolic periodic points, characterized by long-range correlations in the particle kinematics.

C. Further observations on spectral properties

This section addresses the higher-order spectral structure of the diffusive mapping matrix and the generic behavior of $\Lambda_{mp}$ as a function of the Péclet number.

Figure 7 shows the average value and the square root of the rescaled variance associated to the first three non-zero dominant decay exponents (that are related to the second, third, and fourth eigenvalues of the Floquet operator) as a function of the Péclet number at $T_p = 0.8$, $N_c = 50$, and $Pe = 10^3$, compared with the results obtained from Fourier analysis (solid horizontal lines). The convergence properties of the higher-order eigenvalues as a function of $N_p$ are qualitatively identical to that characterizing the second eigenvalue. Therefore, by taking $N_p$ large enough ($N_p \approx 10^2$) the statistical fluctuations in the estimate of the diffusive mapping matrix can be averaged out, and a reliable overall spectral structure can be inferred just from a single realization of $M$.

A comprehensive qualitative picture of the spectrum of the diffusive mapping matrix is reported in Figure 8. Panels (a)–(c) of this figure show the eigenvalue spectrum of the diffusive mapping matrix for different values of the Péclet number, for the TPSF at $T_p = 0.8$, obtained by considering $N_p = 50$. As can be observed, the eigenvalue spectrum converges (for fixed $N_c$) towards an invariant structure as $Pe \to \infty$ which corresponds to the eigenvalue spectrum of the purely convective mapping matrix (see panel (d)).

Once the statistical and numerical properties of the diffusive mapping matrix have been characterized, consider the behavior of the dominant decay exponent as a function of the Péclet number. Figure 9 depicts the typical behavior of $\Lambda_{mp}$ vs the Péclet number for the diffusive mapping matrix. Specifically, the case of $T_p = 0.8$ and $N_c = 50$ is considered, and compared with the results for $A$ obtained by Cerbelli et al. by considering a Fourier-series expansion of the advection-diffusion operator. Up to a given critical Péclet number, that depends on the discretization, i.e., on $N_c$ (this critical $Pe$ value is order of $10^3$ for the case depicted in Figure 9), the values of $\Lambda_{mp}$ practically coincides with those obtained by calculating the dominant decay exponent of the Floquet operator associated with the advection-diffusion Eq. (19) (line (a) in the figure). As the Péclet number...
FIG. 8. Panels (a)–(c): Eigenvalue spectrum of the diffusive mapping matrix ($N_c = 50$) of the sine flow at $T_p = 0.8$ for several values of the Péclet number: (a) $Pe = 10^4$, (b) $Pe = 10^6$, (c) $Pe = 10^8$. Panel (d) depicts the eigenvalue spectrum of the purely convective mapping matrix.

increases, $\Lambda_{mp}$ saturates towards a constant value $\Lambda_{mp}^c$ as a consequence of numerical diffusion induced by discretization. The value $\Lambda_{mp}^c$ corresponds to the dominant decay exponent obtained from the analysis of the purely convective mapping matrix.

Two main conclusions can be drawn from the analysis of the data depicted in Figure 9. (i) Up to a given critical Péclet number, the eigenvalues of the diffusive mapping matrix (obtained by applying

FIG. 9. Dominant decay exponent of the mapping matrix $\Lambda_{mp}$ (dots •) vs $Pe$ at $T_p = 0.8$, $N_c = 50$. Line (a) represents the scaling of the dominant decay exponent $\Lambda$ of the advection-diffusion equation obtained by Cerbelli et al.,\textsuperscript{29} dotted horizontal line (b) is the limit value $\Lambda_{mp}^c$ of $\Lambda_{mp}$ in the purely convective case.
the simple stochastic approach discussed above) provide a reliable and accurate approximation for the eigenvalues of the propagator of the advection-diffusion equation; (ii) coarse-graining introduces an additional diffusive contribution (numerical diffusion) that superimposes to the diffusive term associated with random fluctuations. Therefore, from the analysis of $\Lambda^{c}_{mp}$ it would be possible to infer quantitative information on the effect of numerical diffusion influencing the properties of the mapping matrix in the purely convective setting (diffusionless limit).

The latter observation indicates that the mapping-matrix for a given value of $N_c$ (i.e., for a given coarse-grained resolution), can be regarded as a coarse-grained approximation of an advection-diffusion process,

$$\frac{\partial c}{\partial t} = -\nabla \cdot (V c) + D_{\text{tot}} \nabla^2 c,$$

(26)

associated with a cumulative diffusion coefficient $D_{\text{tot}}$ expressing the superposition of the molecular diffusivity $D$ and of the numerical diffusivity $D_{\text{num}}$ induced by coarse graining,

$$D_{\text{tot}} = D + D_{\text{num}},$$

(27)

where $D_{\text{num}} = D_{\text{num}}(N_c)$ depends on the grid resolution. It is important to observe that Eq. (26) is just a leading-order approximation of the effect of coarse-graining as numerical diffusion may possess highly non-trivial correlation properties that contribute in Eq. (26) as higher-order spatial derivatives of the concentration field $c$. Therefore, Eq. (26) can be regarded as a first starting point to tackle and quantify the action of numerical diffusion on the spectral properties of the matrix $M$.

In dimensionless term, the resulting Péclet number $P e_{\text{tot}}$ is therefore given by

$$\frac{1}{P e_{\text{tot}}} = \frac{1}{P e} + \frac{1}{P e_{\text{eff}}},$$

(28)

where $P e_{\text{eff}} = V_0 L_0 / D_{\text{num}}$ is the effective Péclet number associated with numerical diffusion, which equals the value of the Péclet number pertaining to the purely convective mapping matrix for a given grid resolution $N_c$.

These two issues are carefully addressed in the remainder of this paper by considering a detailed analysis of the TPSF for different, and characteristic values, of the flow period $T_p$.

Equations (27) and (28) represent a leading-order description of the impact of coarse-graining (numerical diffusion) in the construction of the mapping matrices on a discrete grid. Although several studies address the effect of numerical diffusion on advection-diffusion algorithms,\textsuperscript{33,34} mostly with reference to finite-difference schemes, very little is known on the influence of numerical diffusion on spectral properties of advection-diffusion operators, especially for generic mixing (partially or globally chaotic) flows. Consistently with the main motivation of the present analysis, which is rooted on the spectral characterization of advection-diffusion dynamics, we develop a heuristic spectral description of numerical diffusion effects, which is based on the matching of the dominant decay exponent, i.e., of the second eigenvalue of the Floquet operator. This approach that is thoroughly described in Secs. IV A and IV B, provides a reliable quantification of coarse-graining effects in the construction of the mapping matrices, that is also quantitatively consistent with the short-term decay of the concentration field, as discussed in Sec. VI B.

Finally, it is important to observe that the effect of coarse-graining via an effective Péclet number as described in this paper is \textit{generic} with respect to the grid structure, in the meaning that it applies to all of the grid for which no finite proper subset of grid elements represents an invariant subset for the kinematic equations of motion. In practice, it applies to almost all of the grids, apart from those that are explicitly tailored on an ergodic partition of the particle kinematics.

**IV. THE CONVECTIVE LIMIT: INTRODUCTORY ANALYSIS**

This section discusses the spectral properties of purely convective mapping matrices. The main result is that, due to numerical diffusion, the purely convective limit can be applied to obtain reliable quantitative spectral information on the advection-diffusion operator by simply varying the grid resolution.
A. Effective Péclet number

We have discussed in Sec. III C that coarse-graining acts as a superimposed diffusive contribution corresponding to a dimensionless Péclet number equal to $Pe_{\text{eff}}$, and $Pe_{\text{eff}}$ is a function of the grid resolution. The central issue in the analysis of the purely kinematic limit (i.e., in the absence of molecular diffusion) is therefore the understanding of the dependence of $Pe_{\text{eff}}$ on $N_c$.

Consider first the kinematic case of flows possessing significant islands of quasi-periodicity (TPSF protocols at $T_p = 0.56, 0.8, 1.18$). Let $\Lambda(Pe)$ be the dominant decay exponent obtained from the analysis of the continuum advection-diffusion operator as a function of the Péclet number. In general, the dominant decay exponent follows a power-law dependence on $Pe$ for sufficiently large $Pe$ values,

$$\Lambda(Pe) \simeq \Lambda_0 Pe^{-\beta},$$  \hspace{1cm} (29)

where the exponent $\beta$ is bounded by $0 < \beta \leq 1$.

Given $N_c$ and $\Lambda^c_{\text{mp}}$, the effective Péclet number can be defined from the equation

$$\Lambda(Pe) \equiv \Lambda_0 Pe^{-\beta} = \Lambda^c_{\text{mp}}.$$  \hspace{1cm} (30)

In the graph of $\Lambda(Pe)$ vs $Pe$, $Pe_{\text{eff}}$ corresponds to the abscissa of the intersection point between the $\Lambda(Pe)$-curve and the horizontal line at $\Lambda^c_{\text{mp}}$. The determination of $Pe_{\text{eff}}$ is graphically depicted in Figure 10 for several flow conditions (different flow periods). The data for $\Lambda(Pe)$ (□) and the correlation lines (red solid lines) representing Eq. (29) have been taken from Cerbelli et al. The blue horizontal lines in Figure 10 correspond to the values of $\Lambda^c_{\text{mp}}$ for different grid resolutions $N_c$ (the arrows indicate increasing values of $N_c$). The intersection points, whose abscissas provide $Pe_{\text{eff}}$ are marked with a black dot. Using the values for $Pe_{\text{eff}}$ so obtained, it is possible to infer the dependence of the effective Péclet number on the grid resolution. These data are reported in Figure 11 for the three flow protocols considered. Numerical results suggest that the effective Péclet number is proportional to the square of the line lattice size $N_c$,

$$Pe_{\text{eff}} \simeq p_0 N_c^2,$$  \hspace{1cm} (31)

where the prefactor $p_0$ depends on the flow protocol.

The result expressed by Eq. (31) finds a justification by enforcing Einstein’s diffusion law, that in dimensionless form reads as

$$\langle \Delta x^2 \rangle = \frac{2}{Pe} t.$$  \hspace{1cm} (32)

Substituting $Pe_{\text{eff}}$ to $Pe$, a characteristic time $\alpha T_p$ proportional to $T_p$ for the time $t$, and the lattice spacing $N_c^{-1}$ to $\sqrt{\langle \Delta x^2 \rangle}$ one obtains

$$Pe_{\text{eff}} \sim 2 \alpha T_p N_c^2$$  \hspace{1cm} (33)

as observed in Figure 11. Observe that, under the assumption that the factor $\alpha$ is independent of the flow protocol, Equation (33) predicts that the effective Péclet number increases with $T_p$, as observed in the data reported in Figure 11.

B. Scaling analysis

The result expressed by Eq. (31) (or equivalently Eq. (33)), and confirmed numerically by the data depicted in Figure 11, is relevant in the application of purely convective mapping matrices to transport problems. It indicates that a “nominal” or lattice Péclet number $Pe_c$ equal to the square of $N_c$,

$$Pe_c = N_c^2,$$  \hspace{1cm} (34)

can be defined, and that the lattice Péclet number is proportional to the effective Péclet number $Pe_{\text{eff}}$, through a prefactor $p_0^{-1}$ that depends on the flow protocol, $Pe_c = Pe_{\text{eff}}/p_0$. 

FIG. 10. Dominant scaling exponent $\Lambda_{cm}$ of the purely convective mapping matrix (horizontal lines), and its intersection with the corresponding exponent $\Lambda(Pe)$ of the advection-diffusion operator (solid lines (colored red) and symbols (□)). The arrows indicate increasing values of $N_c = 30, 50, 80, 150, 200$. Symbols (●) correspond to the intersection points between $\Lambda(Pe)$ and $\Lambda_{cm}$, defining the effective Péclet number $Pe_{eff}$. Panel (a): $T_p = 0.56$. Panel (b): $T_p = 0.8$. Panel (c): $T_p = 1.18$.

Therefore, given a set of values $\Lambda_{cm,h}, h = 1, \ldots, m$, for $\Lambda_{cm}(N_c)$ for increasing grid resolutions $N_{c,h}$, the graph of $\Lambda_{cm,h}$ vs the corresponding lattice Péclet numbers $Pe_{c,h}$ provides quantitative information about the scaling of the dominant decay exponent $\Lambda(Pe)$ vs $Pe$, apart from a prefactor in the definition of the abscissa. Specifically, it provides information about the scaling exponent $\beta$ entering Eq. (29) exclusively from purely convective data.

Such a graph is depicted in Figure 12 for several protocols of the time-periodic sine flow. This graph should be compared with the corresponding result for the advection-diffusion operator (see Figure 22 and Table II in Cerbelli et al.\cite{Cerbelli29}).
FIG. 11. Effective Péclet number $\text{Pe}_{\text{eff}}$ vs the square of linear lattice size $N_c^2$. Symbols (□) refer to $T_p = 0.56$, (○) to $T_p = 0.8$, (●) to $T_p = 1.18$. The solid lines correspond to the scaling $\text{Pe}_{\text{eff}} \sim N_{c}^{2}$.

FIG. 12. Dominant scaling exponent $\Lambda_{\text{mp}}$ of the purely convective mapping matrix vs the lattice Péclet number $\text{Pe}_c = N_{c}^{2}$ for different periods of the sine flow. Line (a) and (□) $T_p = 0.56$, line (b) and (■) $T_p = 0.8$, line (c) and (○) $T_p = 1.18$, lines (d) and (e) and (●) $T_p = 1.6$.

As discussed above, it is possible to infer from this graph the value of the exponent $\beta$ characterizing the asymptotic decay of the dominant decay exponent for large Péclet values. The values of $\beta$ so obtained from the analysis of the purely convective mapping matrix are reported in Table I, and are in excellent agreement with the values obtained in Cerbelli et al. The case $T_p = 1.18$ shows the more significant discrepancy, but this is due to the fact that the exponent $\beta = 0.55$ found in Cerbelli et al. is just an intermediate behavior that as $\text{Pe}$ increases evolves into an higher value of the exponent $\beta$. This is confirmed by the analysis developed further (see Figure 18, line (c)).

Insofar, we have analyzed flow protocols characterized by medium/large islands of quasi-periodic motion. Before completing the analysis of the kinematic limit of the mapping approach (that is further discussed in Sec. VI), it is useful to address the properties of the diffusive mapping matrix, for which the effects of molecular diffusion are accounted for via the stochastic formulation described in Sec. III. This choice in the order of presentation permits us to address the estimate of the prefactor $p_0$ entering Eq. (31), which is relevant in the analysis of the spectral properties of the advection-diffusion operator via purely mapping techniques.
TABLE I. Scaling exponent $\beta$ characterizing the behavior of the dominant decay exponent with $Pe$ for different values of the period of the TPSF. The second column, indicated with (mp) refers to the exponent derived from the data of the purely convective mapping matrix, the third column refers to the values obtained in Cerbelli et al.\textsuperscript{29} by considering the continuum advection-diffusion operator.

<table>
<thead>
<tr>
<th>$T_p$</th>
<th>$\beta$ (mp)</th>
<th>$\beta$ (Ref. 29)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.56</td>
<td>0.87</td>
<td>0.865</td>
</tr>
<tr>
<td>0.8</td>
<td>0.745</td>
<td>0.745</td>
</tr>
<tr>
<td>1.18</td>
<td>0.6</td>
<td>0.55</td>
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<tr>
<td>1.6</td>
<td>0</td>
<td>0\textsuperscript{a}</td>
</tr>
<tr>
<td>2.0</td>
<td>0.38</td>
<td>0.37\textsuperscript{b}</td>
</tr>
</tbody>
</table>

\textsuperscript{a}This scaling exponent occurs up to $Pe = 2.4 \times 10^4$, as can be observed from Figure 12 lines (d) and (e), and a crossover occurs for higher $Pe$.

\textsuperscript{b}This value does not correspond to the asymptotic exponent $\beta$, since for $Pe \simeq 10^5$ a crossover phenomenon occurs, as discussed in Sec. VI.

V. DIFFUSIVE MAPPING MATRICES

This section analyzes the spectral properties of the mapping matrix when molecular diffusion is accounted for. First, consider those flow protocols discussed in Sec. IV B which possess an asymptotic power-law scaling of $\Lambda (Pe)$ Eq. (29) with $\beta > 0$.

Figures 13 and 14 depict the behavior of the dominant decay exponent $\Lambda_{mp}$ as a function of the Péclet number for the three different flow protocols analyzed in Sec. IV B, namely, $T_p = 0.56, 0.8, 1.18$.

Since the coarse-graining effect superimposes to molecular diffusion, $\Lambda_{mp}(Pe)$ for sufficiently large $Pe$ (such that Eq. (29) applies) can be expressed as

$$\Lambda_{mp}(Pe) \simeq \Lambda_0 \left( Pe^{-1} + Pe_{eff}^{-1} \right)^{\beta} .$$

From Eq. (35), it follows that

$$\Lambda_{mp}(Pe) \simeq \begin{cases} 
\Lambda_0 Pe^{-\beta} & \text{for } Pe \ll Pe_{eff}, \\
\Lambda_0 Pe_{eff}^{-\beta} & \text{for } Pe \gg Pe_{eff} .
\end{cases}$$

For $Pe \ll Pe_{eff}$, the effects of coarse-graining are immaterial, and the graph of $\Lambda_{mp}(Pe)$ vs $Pe$ reproduces accurately that of $\Lambda(Pe)$ associated with the advection-diffusion operator in the continuum.

FIG. 13. Dominant decay exponent of the mapping matrix with diffusion as a function of the Péclet number. Data refer to $N_c = 50$. Line (a) and (□) refer $T_p = 0.56$, line (b) and (○) to $T_p = 0.8$, line (c) and (●) $T_p = 1.18$. The solid lines are the results of Eq. (39).
FIG. 14. Dominant decay exponent of the mapping matrix with diffusion $\Lambda_{mp}$ (symbols) for the TPSF at a function of the Péclet number: (□) $N_c = 50$, (●) $N_c = 100$, (●) $N_c = 200$. The solid lines represent the function equation (39). Panel (a): $T_p = 0.56$. The dotted line represents the scaling $\Lambda_{mp}(Pe) \sim Pe^{-\beta}$, with $\beta = 0.87$. Panel (b): $T_p = 0.8$. The dotted line represents the scaling $\Lambda_{mp}(Pe) \sim Pe^{-\beta}$, with $\beta = 0.745$. Panel (c): $T_p = 1.18$. The dotted line represents the scaling $\Lambda_{mp}(Pe) \sim Pe^{-\beta}$, with $\beta = 0.6$.

As the Péclet number increases, $\Lambda_{mp}(Pe)$ smoothly converges towards a saturation value that corresponds to the dominant decay exponent $\Lambda_{mp}^c$ of the purely convective mapping matrix. Therefore, $Pe_{eff}$ and $\Lambda_{mp}^c$ are related to each other via the equation

$$\Lambda_0 Pe_{eff}^{-\beta} = \Lambda_{mp}^c.$$  

Observe that the quantities $\Lambda_0$ and $\beta$ entering Eq. (35) are intrinsic properties of the advection-diffusion operator in the continuum limit and do not depend on the mapping matrix approximation (although the exponent $\beta$ can be estimated for the purely convective mapping matrices as discussed in Sec. IV B). Conversely, $Pe_{eff}$ and $\Lambda_{mp}^c$ are parameters that characterize the mapping matrix approximation and depend on the coarse-graining resolution, i.e., on $N_c$. 
Substituting Eq. (37) into Eq. (35), it follows that

\[ \Lambda_{mp} = \Lambda_{mp}^c \left( \frac{Pe_{eff}}{Pe} + 1 \right)^\beta . \]  

(38)

Since \( Pe_{eff} \) depends on \( N_c \), see Eq. (31), it is possible to include explicitly the effects of grid resolution into Eq. (38), thus obtaining

\[ \Lambda_{mp} = \Lambda_{mp}^c(N_c) \left( \frac{p_0 N_c^2}{Pe} + 1 \right)^\beta . \]  

(39)

Since \( \Lambda_{mp}^c \) and \( \beta \) can be obtained from the analysis of purely convective mapping matrices, Eq. (39) indicates that the behavior of \( \Lambda_{mp}(Pe) \) can be described by a single-parameter curve, in which the only unknown parameter is \( p_0 \), i.e., the prefactor relating the effective Péclet number to the square of \( N_c^2 \). This observation suggests a simple way to estimate \( p_0 \). In point of fact, by considering a low-resolution grid, Eq. (39) can be used to estimate \( p_0 \) from the graph of \( \Lambda_{mp}(Pe) \) vs \( Pe \).

As an example, Figure 12 shows the data of the dominant decay exponent for the diffusive mapping matrix at \( N_c = 50 \), for the three flow protocols considered (symbols) and the corresponding regression curve equation (39).

By using the value of \( p_0 \) so obtained, and the data deriving from purely convective analysis, Eq. (39) predicts the behavior of \( \Lambda_{mp}(Pe) \) for any value of \( N_c \) without the use of any further fitting parameter. The comparison between Eq. (39) and the numerical data for different grid resolutions is depicted in Figure 14. For the three flow protocols considered, the value of the prefactor \( p_0 \) has been obtained from the data on the \( N_c = 50 \) grid. As can be observed, the agreement between Eq. (39) and numerical data for \( \Lambda_{mp}(Pe) \) is excellent, with the exception of the low \( Pe \)-range behavior at \( T_p = 1.18 \).

The discrepancy observed in this case finds a simple explanation if one consider more accurately the spectral properties of this flow protocol. Figure 15 compares the results for \( \Lambda_{mp}(N_c = 50) \) and the results of the dominant decay exponent of the advection-diffusion equation obtained by means of Fourier analysis. Two main conclusions can be derived by the data reported in Figure 15: (i) in the low Péclet number region, i.e., up to values of \( Pe \) at which the saturation effects for \( \Lambda_{mp} \) induced by numerical diffusion becomes significant (\( Pe < 5 \times 10^3 \)), \( \Lambda_{mp} \) provides an accurate numerical estimate for \( \Lambda(Pe) \); (ii) the scaling \( \Lambda(Pe) \sim Pe^{-\beta} \) with \( \beta = 0.55 \) sets in starting from \( Pe \geq 10^3 \), and a deviation from this scaling occurs for lower \( Pe \) values (see also Figure 17 in Cerbelli et al.29).

Therefore, it is not surprising that the continuous line in Figure 14 based on Eq. (39) shows a discrepancy for low values of \( Pe \), just because this equation is based on a unique (global) power-law scaling, which is not fully fulfilled for \( Pe \leq 10^3 \).

To conclude the analysis, let us consider another typical class of flow protocol, namely, that of almost globally chaotic flows. An example of this class is given by the TPSF at \( T_p = 1.6 \). For \( Pe \in (5 \)
The dominant decay exponent $\Lambda \mp(Pe)$ is practically independent of the Péclet number, and attains an almost constant value $\Lambda \mp(Pe) \simeq 0.34$. In this situation, the analysis developed above for predicting the behavior of $\Lambda \mp(Pe)$ and for estimating the prefactor $p_0$ does not apply. Figure 16 depicts the behavior of $\Lambda \mp(Pe)$ at $T_p = 1.6$ for several resolutions $N_c = 30, 100, 200$.

In this case, it is very difficult to determine the value of $p_0$ from the graph of $\Lambda \mp(Pe)$ vs $Pe$, since the saturation is a consequence of both numerical diffusion and the eventual singular (saturating) behavior of the dominant decay exponent for large Péclet numbers. For this reason, the most convenient strategy to determine $p_0$ is to estimate the value of $Pe_{eff}$ at a low resolution grid, say $N^*_{c}$, by comparing the value of $\Lambda \mp_{c}$ with that of the dominant decay exponent of the continuum advection-diffusion operator, i.e.,

$$\Lambda(Pe_{eff}) = \Lambda(p_0 N^*_{c}) = \Lambda \mp_{c} \big|_{N_c = N^*_{c}}. \quad (40)$$

We choose $N^*_{c} = 30$, to which corresponds the value $\Lambda \mp_{c} \simeq 0.38$.

The value of the prefactor $p_0$ so obtained, as well as the values of $p_0$ for the other flow protocols considered above are depicted in Figure 17. The prefactor $p_0$ increases with the flow period $T_p$, although its graph is not monotonic. For example, the value of $p_0$ at $T_p = 1.18$ is larger than at $T_p = 1.6$.

There is another issue that can be inferred from the data of $\Lambda \mp$ at $T_p = 1.6$ depicted in Figure 16. While in the range $N_c \in (30, 100)$ the limit value of $\Lambda \mp(Pe)$ is almost independent of $Pe$ (and this is a consequence of the fact that in the corresponding range of effective Péclet numbers the dominant decay exponent of the continuum advection-diffusion operator saturates to a constant, Péclet-independent, value), if the resolution increases further, see the data for $N_c = 200$, the value of $\Lambda \mp_{c}$ start to decrease significantly. This is not a numerical artifact, but rather it indicates that for
this flow protocol the dominant decay exponent \( \Lambda(Pe) \) does not saturate towards a constant value but decreases to zero. This observation is further addressed in Sec. VI.

VI. THE CONVECTIVE LIMIT: CONCLUDING OBSERVATIONS

This section addresses further the application of purely convective mapping matrices for predicting the spectral properties of the advection-diffusion operator and the evolution of scalar fields advected by a flow field in the presence of diffusion.

Given the prefactor \( p_0 \) that, as discussed in Sec. V, can be estimated in several different ways, it is possible to make a quantitative comparison of the spectral properties of the continuum advection-diffusion operator respect to the coarse grained mapping counterpart in purely convective conditions.

Figure 18 depicts this comparison of \( \Lambda_{mp}^c \) (symbols •) and \( \Lambda \) (symbols ◦) vs the effective Péclet number \( Pe_{eff} \) for the TPSF protocols at \( T_p = 0.56, 0.8, \) and 1.18. Here, \( Pe_{eff} = Pe \) for the data obtained by using the continuum advection-diffusion operator that are taken from Cerbelli et al.\(^{29}\) As can be observed there is a strict quantitative agreement between the values of the dominant decay exponent obtained from the purely convective mapping matrix and \( \Lambda \) associated with the propagator of the continuum operator.

The analogous graph for \( T_p = 1.6 \) is depicted in Figure 19. In this figure, data taken from Cerbelli et al.\(^{29}\) (symbols ◦) have been complemented with new results for the continuum advection-diffusion

![Figure 18](image1.png)

**FIG. 18.** Comparison of the prediction of dominant decay exponent by using the purely convective mapping matrix \( \Lambda_{mp}^c \) with respect to the results obtained by the spectral analysis of the advection-diffusion operator. Symbols (•) refer to \( \Lambda_{mp}^c \), (◦) to \( \Lambda \). Line (a) corresponds to \( T_p = 0.56 \), (b) to \( T_p = 0.8 \), (c) to \( T_p = 1.18 \).

![Figure 19](image2.png)

**FIG. 19.** Comparison of the dominant decay exponent \( \Lambda_{mp}^c \) of the purely convective mapping matrix (•) and the dominant decay exponent of the corresponding advection-diffusion operator (◦, □) at \( T_p = 1.6 \).

operator at higher Pécelt numbers (symbols □) than those considered in Cerbelli et al.\textsuperscript{29} These data have been obtained by using the same approach described in Cerbelli et al.,\textsuperscript{29} that is projecting the advection-diffusion operator into a truncated Fourier basis $\{e^{i2\pi(mx+ny)}\}_{m,n=-N_f}^{N_f}$ with $N_f$ up to 200 for $Pe = Pe_{\text{eff}} = 5 \times 10^5$.

This protocol is particularly interesting, as it is one of the classical examples of an almost globally chaotic flow.\textsuperscript{25, 29} As can be observed, the scaling $\Lambda \sim \text{constant}$, occurring up to $Pe = Pe_{\text{eff}} = 10^5$, reveals not to be an asymptotic property, but solely an intermediate regime, corresponding to the occurrence of “strange eigenmodes” distributed all over the mixing domain (see Figure 20 panel (a)). As the Pécelt number increases beyond $Pe = 10^5$, a new asymptotic regime occurs characterized by the localization of the eigenmodes near the low stretching regions (see Figure 20 panel (b)), and by the scaling of $\Lambda \sim Pe^{-\beta}$ where $\beta \simeq 0.5$. A further discussion of this flow protocols—that in point of fact proves to be partially chaotic—is addressed in Sec. VII B. The connection between eigenfunction structure and kinematic properties (stretching distribution, invariant measures associated with the unstable foliation, etc.) is beyond the scope of this paper and is a topic for future research.

Figure 20 shows the modulus of the second eigenfunction of the purely convective mapping matrix at $N_c = 100$ (panel (a)) and $N_c = 200$ (panel (b)), which correspond to values of the effective Pécelt number $Pe_{\text{eff}} = 7.8 \times 10^4$, and $Pe_{\text{eff}} = 3.1 \times 10^5$, respectively, below and above the transition point (around $Pe = 10^5$) separating the occurrence of global strange eigenmodes, and of asymptotic localized ones. The localization effects are clearly evident in panel (b) (higher values of the modulus correspond to the light-shaded striations, lower values to the dark region).

It is worth observing that, even with relatively small grids order of $N_c \leq 200 - 300$, (the data depicted in Figure 18 refer to grid size up to $N_c = 200$, the data depicted in Figure 19 to $N_c \leq 300$), it is possible to address advection-diffusion problems for relatively high values of the Pécelt number. This indicates that the use of the purely convective mapping method can be a very powerful, and computationally not expensive, alternative with respect to other numerical techniques to address transport problems at very high Pécelt values (see further the discussion in Sec. VII B).

A. Critical cases

As a further example, let us consider the TPSF protocol at $T_p = 2.0$. The Poincaré map of this flow exhibits a large chaotic region, extending almost all over the flow domain, and parabolic periodic points at $(1/4 \pm 1/2, 1/4 \pm 1/2)$, in the neighborhood of which the kinematics is regular. Figure 21 depicts a zoom-in of the Poincaré map close to one of these periodic points, in the neighborhood of which the Lagrangian evolution of passive particles exhibits long-range correlation\textsuperscript{51, 52} (i.e., “stickiness,” as sometimes referred to in the specialized literature\textsuperscript{55}).

Figure 22 shows the behavior of the dominant decay exponent $\Lambda_{\text{mp}}$ of the purely convective mapping matrix vs the lattice Pécelt number $Pe_c = N_c^2$ for several lattice resolutions. As can be
FIG. 21. Zoom-in of the Poincaré map of the TPSF at $T_p = 2$ near the periodic point (0.25, 0.25).

FIG. 22. Dominant decay exponent $\Lambda_{mp}^c$ of the purely convective mapping matrix vs the lattice Péclet number $Pe_c = N_c^2$ for $T_p = 2$. The solid line (a) represents the scaling $\Lambda_{mp}^c \sim Pe_c^{-\beta}$ with $\beta = 0.38$, solid lines (b) and (c) the scaling $\Lambda_{mp}^c \sim Pe_c^{-\beta}$ with $\beta = 0.45$.

observed, the behavior of $\Lambda_{mp}^c$ is highly non-monotonic as a function of $Pe_c$ (and ultimately of $N_c$). Since we have considered solely even values of $N_c$, two different situations arise, depending whether $N_c/2$ is even or odd. For $N_c = 4n$, with $n$ integer, (i.e., $N_c/2$ even), one observe higher values of $\Lambda_{mp}^c$ corresponding to lines (a) and (c) in Figure 22. The data for $N_c = 2(2n + 1)$ correspond to line (b) in Figure 22. Depending on the parity of $N_c/2$, the coarse-graining procedure used to construct the convective mapping matrix samples in different ways the dynamic evolution of passive particles in the neighborhood of the parabolic fixed points, and this seems to be the origin of the non-monotonic behavior of $\Lambda_{mp}^c$ shown in Figure 22, which is controlled exclusively on the parity of $N_c/2$. At present, no further theoretical explanation is available of this rather singular and “pathological” behavior as it regards coarse-graining.

Nevertheless, if one consider separately even and odd values of $N_c/2$ (i.e., data lying on lines (a) and (c) and on line (b)) if they were two different numerical experiments, the data for $T_p = 2.0$ depicted in Figure 22 can be treated as any other flow protocol discussed so far, with the methods discussed in Secs. III–V.

Figure 23 panels (a) and (b) depict the modulus of the dominant eigenfunction for $N_c = 50$ (i.e., for $N_c/2$ odd, corresponding to the lower system of data depicted in Figure 22, line (b)) and for $N_c = 52$ ($N_c/2$ even, associated with the upper system of data depicted in Figure 22, lines (a) and (b)). For even values of $N_c/2$, the coarse-graining procedure induces a lower value of the effective Péclet number, and this phenomenon can be observed from the occurrence of larger localization regions of the dominant eigenfunction close to the parabolic fixed points ($1/4 \pm 1/2$, $1/4 \pm 1/2$). This is evident from the contour plot depicted in panel (b) of Figure 23, where a larger localization “wake” extends...
FIG. 23. Modulus (multiplied by $10^{-3}$) of the second eigenfunction of the purely convective mapping matrix at $T_p = 2$.
Panel (a): $N_c = 50$. Panel (b): $N_c = 52$.

close to the four periodic point, while it is absent (or at least greatly reduced) for $N_c = 50$ (panel (a)).

By consider odd and even values of $N_c/2$ separately, it is possible to define two effective Péclet numbers for even and odd values of $N_c/2$, and two different prefactors, $P_{\text{eff, even}} = p_{0, \text{even}} N_c^2$, $P_{\text{eff, odd}} = p_{0, \text{odd}} N_c^2$, from diffusive mapping matrix data using a low resolution grid, as discussed in Sec. V (these data are not reported since these are qualitatively identical to those depicted in Figures 13 and 14). The values of prefactors so obtained are $p_{0, \text{even}} = 38$, $p_{0, \text{odd}} = 700$, i.e., the effect of numerical diffusion in odd grid is more than one order of magnitude smaller than for even grid. In Figure 17 solely $p_{0, \text{even}}$ has been reported. From the knowledge of the prefactors, it is possible to represent the spectral data in Figure 22 as a function of the effective Péclet number, and compare them with the spectral results of the continuum advection-diffusion operator (the data for $\Lambda$ are taken from Cerbelli et al.\textsuperscript{29}). Figure 24 provides this comparison. As can be observed, all of the data fall into the graph of a single curve (no matter the parity of $N_c/2$), and are quantitatively consistent with the continuum data taken from Cerbelli et al.\textsuperscript{29}

It can be observed that the dominant decay exponent for this flow protocol exhibits a crossover behavior at high values of $Pe$. For $Pe = P_{\text{eff}} < 3 \times 10^5$, $\Lambda(Pe) \sim Pe^{-\beta}$ with an exponent $\beta = 0.37$ (and this is consistent with the data reported in Cerbelli et al.\textsuperscript{29}), while for $P_{\text{eff}} > 5 \times 10^5$, the scaling exponent attains the value $\beta = 0.45$ (this range of high $Pe$ values was not explored in Cerbelli et al.\textsuperscript{29}). In point of fact, it is rather remarkable that the mapping-matrix approach can be used to

FIG. 24. Comparison of the dominant decay exponent $\Lambda_{\text{mp}}$ of the purely convective mapping matrix (symbols • and □) and the dominant decay exponent of the corresponding advection-diffusion operator (○) for $T_p = 2$. Symbols (●) refer to the lower branch of $\Lambda_{\text{mp}}$ for $N_c = 2(2n + 1)$, (□) to the upper branch for $N_c = 4n$, with $n$ integer. Solid lines (a) and (b) represent the scalings $\Lambda \sim Pe_{\text{eff}}^{-\beta}$ with $\beta = 0.37$ and $\beta = 0.45$, respectively.
simulate advection-diffusion processes for very high values of the Péclet number, using reasonable small lattice sizes (the data depicted in Figure 24 refer to $N_c \leq 200$).

To sum up, TPSF protocol at $T_p = 2.0$ is a rather peculiar case, revealing an “anomalous” behavior related to symmetry and coarse-graining sampling influencing the construction and the spectral properties of the associated mapping matrices. Nevertheless, a careful analysis of these symmetries and pathologies (the example depicted in Figure 22 is just a case, other situations may in principle occur for other flow protocols) makes it possible also in this “pathological” case to use the methods developed in Secs. III–V, and ultimately to apply purely convective mapping matrices for predicting the properties of advection-diffusion processes.

**B. Norm evolution**

The analysis developed in Secs. III–V and VI A has been focused mainly on the dominant (second) eigenvalue. We have found a quantitative correspondence between spectral properties of the purely convective mapping matrices and of the corresponding advection-diffusion operators through the introduction of an effective Péclet number.

It is worth investigating further this correspondence, by analyzing the temporal evolution of scalar fields, especially as it regards the early stages of the mixing process, since the initial evolution involves the entire spectral structure of the advection-diffusion operator and not exclusively the dominant eigenvalue.

Consider as an initial condition the segregated zero-mean field

$$\phi_0(x) = \begin{cases} 
1 & 0 \leq x < 1/2, \\
-1 & 1/2 \leq x < 1,
\end{cases}$$

(41)

where $x = (x, y)$.

Let $\phi_0$ be the discretized representation of $\phi_0(x)$ on a grid. The coarse-grained evolution of $\phi(x, t)$ at time instants multiple of the flow period can be obtained by iterating the mapping matrix $M$, i.e.,

$$\phi_n = \phi|_{t=nT_p} = M^n \phi_0.$$  \hspace{1cm} (42)

Figure 25 panel (a) depicts the time-behavior of the $L^2$-norm of the scalar field $\phi$ obtained by iterating the purely convective mapping matrix, compared with the corresponding norm of the solution of the continuum advection-diffusion equation starting from the same initial condition (41) for the TPSF at $T_p = 0.8$. The latter data have been obtained by expanding the scalar field $\phi(x, t)$ in Fourier modes $\phi(x, t) = \sum_{m, n} \phi_{m, n}(t) e^{2\pi i(mx + ny)}$, projecting the advection-diffusion equation in the

![Figure 25](https://example.com/figure25.png)

**FIG. 25.** $L^2$-norm decay of a scalar field $\phi$ vs time $t$. Symbols refer to the coarse-grained evolution via the purely convective mapping matrix, solid lines (a)-(c) the results obtained by solving the advection-diffusion equation for a value of Pe number equal to the effective Péclet number pertaining to each discretization. Symbols (□) refer to $N_c = 30$, (○) to $N_c = 50$, (●) to $N_c = 80$. Panel (a): $T_p = 0.8$. Panel (b): $T_p = 1.18$. 

Fourier basis, thus obtaining a system of ordinary differential equations for the Fourier coefficients \( \{ \phi_{m,n}(t) \} \). For each grid resolution \( N_c \), the value of the Péclet number in the continuum advection-diffusion equation has been set equal to the effective Péclet number pertaining to that resolution (obtained from the data depicted in Figures 10 and 11).

As can be observed there is a satisfactory quantitative agreement between mapping matrix simulations and the solution of the advection-diffusion equation, even in the early stages of the mixing process. This indicates that the purely convective mapping matrix provides a reliable coarse-grained approximation for the advection-diffusion operator for a value of the Péclet number equal to effective Péclet number associated with each discretization.

Panel (b) in Figure 25 depicts the analogous comparison for \( T_p = 1.18 \) which, as can be observed from the results depicted in Figure 18 and reported in Table I, provides the worst scenario for the application of the mapping matrix formalism amongst those considered in this paper. Even in this case, the purely convective simulations using the mapping matrix formalism provide an acceptable approximation for the initial norm decay of the concentration field.

VII. SPECTRAL ANALYSIS OF REALISTIC FLOWS AND FURTHER DEVELOPMENTS

A. Realistic flow systems

This section discusses the potentialities of the analysis developed in this paper to address mixing and transport problems of relevant physical and practical fluid dynamic interest. The approach outlined in this paper based on the application of the mapping matrix formalism, and described with the aid of the prototypical TPSF, can be straightforward applied and extended practically without any difference to realistic and physically realizable flow fields both in the case of closed and open mixing devices (such as the PPM discussed above, which represents a prototypical case of static mixer). Specifically, the application of the purely convective mapping matrix is particularly intriguing for addressing the simulation of mixing processes (in which diffusion is accounted for in virtue of numerical diffusion) in realistic micro-devices of arbitrary shape and geometrical complexity as it requires exclusively the integration of the kinematic (Lagrangian) equations of motion.

As an example of the application to a physically realizable flow, consider the PPM that has been briefly described in Sec. II C. This flow system has been addressed by Singh et al. using the purely convective mapping matrix formalism. We use the flow field approximation proposed by Khakhar et al., that depends on a single dimensionless parameter \( \beta_{ppm} \), which is essentially the ratio of the intensity of the cross-sectional velocity field to the mean axial velocity.

Figure 26 depict the Poincaré maps of the PPM for four different values of \( \beta_{ppm} = 1, 8, 10, 20 \) (panels (a)–(d)). As \( \beta_{ppm} \) increases, the islands of quasi-periodic behavior shrink in size, and therefore, intuitively it can be expected that the mixer exhibit better mixing performance (higher values of the dominant decay exponent) as \( \beta_{ppm} \) increases.

We consider steady-state mixing conditions. Since in the dimensionless formulation the length of each of the two units, the alternating repetition of which constitute the PPM, equals 1, the dominant decay exponent is defined as \( \Lambda = -\log |\nu_2|/2 \). Here, we consider solely spectral properties deriving from the purely kinematic mapping matrix. A structured triangular grid has been used.

Figure 27 reviews the behavior of the dominant decay exponent \( \Lambda_{mp} \) vs the lattice Péclet number \( Pe_c = N^2_p \). These data refer to a structured grid of rectangles in the \( r-\theta \) plane possessing the same area, where \( N_p = 10 \), i.e., 100 particles per box (as the estimate of the mapping matrix does not change as \( N_p \) increases). As \( \beta_{ppm} \) increases, the scaling exponent \( \beta \) decreases, from \( \beta = 1 \) (diffusive scaling) occurring for \( \beta_{ppm} = 1 \), the Poincaré map of which is characterized by a system of large and small quasi-periodic islands (Figure 26 panel (a)) to \( \beta = 0.25 \) for \( \beta_{ppm} = 20 \), characterized by a thin layer of quasi-periodic motion near the outer wall of the tube (Figure 26 panel (b)). As discussed above, the estimate of the scaling exponent \( \beta \) is independent of the prefactor \( \nu_0 \) relating the lattice Péclet number to the effective Péclet number, and therefore can be taken as an intrinsic property of the advection diffusion operator that can be extracted directly from purely convective data without the use of any further post-processing (such as the estimate of \( \nu_0 \)).
FIG. 26. Poincaré sections of the PPM. Panel (a): $\beta_{ppm} = 1$, (b) $\beta_{ppm} = 8$, (c) $\beta_{ppm} = 10$, (d) $\beta_{ppm} = 20$.

FIG. 27. Dominant scaling exponent $\Lambda_{mp}$ of the purely convective mapping matrix vs the lattice Péclet number $Pe_c$ for the PPM at different values of $\beta_{ppm}$. Line (a) and (□) $\beta_{ppm} = 1$, line (b) and (■) $\beta_{ppm} = 8$, line (c) and (○) $\beta_{ppm} = 10$, lines (d) and (●) $\beta_{ppm} = 20$. Dashed lines represent the scalings $\Lambda_{mp} \sim Pe_c^{-\beta}$, for the values of the exponent $\beta$ reported in Table II.

Table II reviews the values of the scaling exponent $\beta$ extracted from the data depicted in Figure 27. As expected, $\beta$ decreases with $\beta_{ppm}$, indicating that higher transverse velocity components improve mixing.

In the construction of the diffusive mapping matrix in the presence of more complex velocity fields than parallel flows (as for the two velocity fields characterizing TPSF), more accurate numerical results can be achieved by considering higher-order schemes than the Euler algorithm (20) for solving the stochastic Langevin equation (18). Several higher-order schemes have been proposed, such as the Runge-Kutta-Langevin algorithm (56) or the higher-order quadrature methods for stochastic differential equations discussed by Kloeden and Platen (57) and Mannella (58).
TABLE II. Scaling exponents $\beta$ for the PPM for the different values of $\beta_{ppm}$ considered in the paper.

<table>
<thead>
<tr>
<th>$\beta_{ppm}$</th>
<th>$\beta$</th>
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<tbody>
<tr>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>8</td>
<td>0.87</td>
</tr>
<tr>
<td>10</td>
<td>0.83</td>
</tr>
<tr>
<td>20</td>
<td>0.3</td>
</tr>
<tr>
<td>20</td>
<td>0.8</td>
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</table>

*For $\beta_{ppm} = 20$, the scaling $\Lambda_{mp}^c \sim Pe^{-0.3}$ holds up to $Pe_c = 5 \times 10^3$, while for higher $Pe \Lambda_{mp}^c \sim Pe^{-0.8}$, see Figure 27 lines (d) and (e).

B. Large Pe simulations

Numerical analysis of mixing obtained by solving the advection-diffusion equations is limited in the current literature to an upper-bounded interval of Pe values (usually $Pe \leq 10^6$), just because as $Pe$ increases, mixing patterns characterized by higher spatial gradients should be resolved and this gives rise to challenging numerical issues. In the case of Fourier (Galerkin) projections, this would require an increasingly higher number of modes (basis functions) in the expansion of the concentration field beyond the feasibility limits of customarily available computers.

In point of fact, the use of the purely convective mapping matrix, as an emulator of advection-diffusion processes (owing to the effects of coarse-graining and numerical diffusion), permits to extend the range of numerical investigation of mixing in the presence of diffusion to significantly higher $Pe$ values.

To give an example, consider the TPSF protocol at $T_p = 1.6$, that has been discussed in Sec. VI (see Figure 19). Figure 28 depicts the behavior of $\Lambda_{mp}^c$ vs $Pe$ for much larger lattices (up to $N_c = 1500$). In the case of very large mapping matrices, the dominant decay exponent has been obtained by using a power method. As can be observed, for $Pe \in (2 \times 10^5, 2 \times 10^7)$, $\Lambda_{mp}^c$ decays as $Pe^{-\beta}$ where $\beta = 0.5$. This behavior is indeed not surprising, as a more careful investigation of the kinematics of this flow protocol (triggered by the spectral data depicted in Figure 28, and by the localization properties of the corresponding eigenfunctions, see Figure 26 panel (b)), reveals that this system is not globally chaotic, as its Poincaré section possesses very small islands of quasi-periodic behavior (one of which is depicted in Figure 29).

The data depicted in Figure 28 show that it is possible, by means of mapping simulations, to reach values of $Pe_{eff}$ well above $10^7$. Owing to the sparse nature of the mapping matrix, it is further possible at feasible computational costs, to reach linear grid resolutions of about $N_s = 5000$ that

![Figure 28](image_url)

**FIG. 28.** Behavior of the dominant decay exponent $\Lambda_{mp}^c$ vs $Pe$ for very high values of $Pe_{eff}$ obtained using a power-method. Line (a) refers to $\Lambda_{mp}^c \sim$ constant, line (b) to the scaling $\Lambda_{mp}^c \sim Pe_{eff}^{-\beta}$, where $\beta = 0.5$. 
would correspond to values of \( Pe_{\text{eff}} \) close to \( 10^9 \). The availability of such simulations of advection-diffusion dynamics is of great practical (e.g., in connection with highly viscous polymer processing) and theoretical significance.

To give an example of theoretical interest, high \( Pe \) data would permit to investigate some fundamental properties of advection-diffusion dynamics occurring in globally chaotic flows related to the eventual occurrence of a singular limit \(^{25}\) in the behavior of \( \Lambda(Pe) \) for \( Pe \to \infty \), in order to ascertain whether and under what kind of kinematic conditions \( \lim_{Pe \to \infty} \Lambda(Pe) = \Lambda_{\infty} \neq 0 \).

C. Advection-diffusion analysis in the presence of long-range correlated fluctuations

There is another, straightforward, significant extension of the diffusive mapping method discussed in this paper. Throughout this paper, we have considered exclusively the Langevin equation (18) where the stochastic perturbation is a vector-valued Wiener process, owing to the fact that there is a one-to-one correspondence between this stochastic differential equation and the advection-diffusion equation for its probability density function (the associated forward Fokker-Planck equation).

However, the same simulation approach can be extended straightforwardly to more general classes of Langevin equations of the form

\[
d\mathbf{x}(t) = \mathbf{v}(\mathbf{x}(t), t) \, dt + \mathbf{G} \, d\mathbf{\xi}_c(t),
\]

where \( \mathbf{G} \) is a \( d \times d \) constant-coefficient matrix and \( d\mathbf{\xi}_c(t) \) are the increments of a generic vector-valued stochastic process. For instance, if \( \mathbf{\xi}_c(t) \) is a \( d \)-dimensional fractional Brownian motion,\(^{59}\) Eq. (43) would correspond to a fractional Langevin equation.\(^{60, 61}\) In this way, the coupling between an advecting field and stochastic fluctuations possessing long-range correlations could be studied directly in terms of a coarse-grained description of the evolution equation for densities, opening the possibilities of performing simple and reliable simulations on the effects of long-range fluctuations not only in fluid dynamics (correlation effects arise in the fluctuation properties of complex polymeric fluids and suspensions) but also in a wealth of different physical phenomenologies.\(^{60}\)

VIII. CONCLUDING REMARKS

This paper has provided a quantitative analysis of the spectral information that can be recovered from the the mapping matrix formalism applied to study transport processes involving the interplay between advection and diffusion. Two major results have been obtained. First of all, the inclusion of molecular diffusion in the mapping approach, that insofar has been grounded exclusively on purely convective simulations. The method chosen is based on the stochastic formulation of advection-diffusion kinematics (Langevin equations) and on the analogy between stochastic differential equations driven by vector-valued Wiener processes and advection-diffusion dynamics.
The stochastic approach is of straightforward implementation and can be regarded as the natural extension of the classical mapping matrix method.

Moreover, the analysis of diffusional effects has enabled to frame in a quantitative way the application of the purely convective mapping method. In point of fact, coarse-graining acts essentially as a superimposed and additional diffusional contribution. This observation can be exploited in several different ways. On one hand, for a fixed value of the Péclet number, it defines quantitatively the minimum grid size above which numerical diffusion becomes practically immaterial. On the other hand, it indicates that the purely convective analysis, performed using the kinematic mapping matrix in the absence of stochastic fluctuations, provides a reliable and robust approximation for the advection-diffusion operator at some well-defined value of the Péclet number, namely $\text{Pe}_{\text{eff}}$. Indeed, the definition of the effective Péclet number and its scaling as a function of the linear lattice size $N_c$ is the key concept in the transposition of purely convective simulations to model advection-diffusion processes.

The quantitative applicability of the purely convective mapping analysis has been verified both using spectral information (essentially, the dominant decay exponent, i.e., the second eigenvalue of the advection-diffusion propagator) and the norm decay in the evolution of scalar fields.

This result lay on a sound and quantitatively firm basis of the application of the purely convective analysis in the study of mixing on finite grids. What is indeed remarkable, is that thanks to coarse-graining, and ultimately to the resulting numerical diffusion effects (that in the majority of the applications are considered as unavoidable, spurious, and undesired fall-out of discretization), a correspondence between purely convective simulations and advection diffusion dynamics can be outlined.

We have considered mainly a prototypical chaotic flow, namely, TPSF for developing the reasoning and the analysis. But we have also shown that, without any additional difficulty, this approach can be applied to physically realizable flows (the case of the PPM provides an illustration for this) in both closed and open mixing systems, operating either in time-dependent way or at steady state.

The possibility of simulating flows at very high Péclet value can be exploited to address several unresolved issues in the theory of fluid mixing, such as under what conditions a singular spectral limit can occur in globally chaotic flows. Similarly, the possibility of simulating advection-diffusion problems in the presence of long-range fluctuations (such as those deriving from fractional Brownian motion processes) will permit a better understanding on the interplay between a deterministic convective field and noise, in the case of complex fluids and suspensions.

The analysis developed in this paper has been focused on mixing properties of scalar fields. On equal footing the mapping analysis including diffusion can be generalized to vector-valued or tensor-valued quantities. This will permit the application of this computational technique to the investigation of fundamental issues in magneto-hydrodynamic theory, especially as it regards the occurrence of positive (diverging) eigenvalues for the vector advection-diffusion operator in three-dimensional flows (the so-called fast-dynamo problem).