Lagrangian transport characteristics of a class of three-dimensional inline-mixing flows with fluid inertia

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Lagrangian transport characteristics of a class of three-dimensional inline-mixing flows with fluid inertia

M. F. M. Speetjens,¹ E. A. Demissie,¹ G. Metcalfe,² and H. J. H. Clercx³

¹Department of Mechanical Engineering, Energy Technology Laboratory, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands
²Commonwealth Scientific and Industrial Research Organisation (CSIRO), VIC 3190 Melbourne, Australia
³Department of Applied Physics, Fluid Dynamics Laboratory, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

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Laminar mixing by the inline-mixing principle is a key to many industrial fluids-engineering systems of size extending from micrometers to meters. However, insight into fundamental transport phenomena particularly under the realistic conditions of three-dimensionality (3D) and fluid inertia remains limited. This study addresses these issues for inline mixers with cylindrical geometries and adopts the Rotated Arc Mixer (RAM) as a representative system. Transport is investigated from a Lagrangian perspective by identifying and examining coherent structures that form in the 3D streamline portrait. 3D effects and fluid inertia introduce three key features that are not found in simplified configurations: transition zones between consecutive mixing cells of the inline-mixing flow; local upstream flow (in certain parameter regimes); transition/inertia-induced breaking of symmetries in the Lagrangian equations of motion (causing topological changes in coherent structures). Topological considerations strongly suggest that there nonetheless always exists a net throughflow region between inlet and outlet of the inline-mixing flow that is strictly separated from possible internal regions. The Lagrangian dynamics in this region admits representation by a 2D time-periodic Hamiltonian system. This establishes one fundamental kinematic structure for the present class of inline-mixing flows and implies universal behavior in that all states follow from the Hamiltonian breakdown of one common integrable state. A so-called period-doubling bifurcation is the only way to eliminate transport barriers originating from this state and thus is a necessary (yet not sufficient) condition for global chaos. Important in a practical context is that a common simplification in literature, i.e., cell-wise fully-developed Stokes flow (“2.5D approach”), retains these fundamental kinematic properties and deviates from the generic 3D inertial case only in a quantitative sense. This substantiates its suitability for (at least first exploratory) studies on (qualitative) mixing properties. © 2014 AIP Publishing LLC.

I. INTRODUCTION

Mixing of fluids under laminar flow conditions is omnipresent in industrial processes, ranging from the traditional mixing of viscous fluids¹–³ via compact processing equipment⁴–⁶ to microfluidic applications for heat and mass transfer,⁷–⁹ labs-on-a-chip,¹⁰, ¹¹ and biotechnology.¹², ¹³ Many (components of) devices operate by the same “inline-mixing” principle: a continuous throughflow that is “stirred” in transverse direction via fixed internal elements or moving boundaries. Macroscopic industrial systems include the well-known Kenics mixer,¹⁴–¹⁷ the multi-flux mixer,¹⁸ the SMX mixer,¹⁹ and the Rotated Arc Mixer (RAM).²⁰–²² The inline-mixing concept admits downscaling⁷, ⁸ and has found application in an array of compact and micro-fluidic counterparts such as the micro-Kenics mixer,²³ the staggered herringbone mixer,²⁴, ²⁵ and the serpentine micro-channel.²⁶, ²⁷ These
configurations have been the subject of investigation in numerous studies. However, such analyses typically concentrate on design and optimization of transport processes instead of on fundamental mechanisms (as, e.g., formation or breakdown of transport barriers). Moreover, they often are restricted to simplified conditions such as piece-wise developed flow or non-inertial (i.e., Stokes) flow. This motivates the present study, which concentrates specifically on fundamental transport properties in inline-mixing flows and then under essentially three-dimensional (3D) flow conditions with significant fluid inertia.

The RAM is adopted as representative system for inline mixers with flow domains that are topologically cylinders, that is, any geometry that can be created by continuous deformation of a cylinder including, e.g., the beforementioned staggered herringbone mixer and the serpentine micro-channel. Throughflow in the RAM is driven by an axial pressure gradient; transverse flow is induced by an encasing rotating cylinder acting on the fluid via an axial sequence of reoriented apertures. Its (qualitative) flow and transport properties are (in principle) transferable to any device with the same domain topology. Transport is investigated from a Lagrangian perspective by identifying and examining coherent structures that form in the 3D streamline portrait of the 3D steady flow. Such coherent structures define the flow topology and geometrically determine the transport properties of the flow. This, arguably, affords the deepest fundamental insight into transport and mixing phenomena.

The present study expands on the Lagrangian transport analysis in Ref. 21, where the 3D RAM flow was approximated by an aperture-wise fully-developed Stokes flow (i.e., depending only on the transverse \((x, y)\) coordinates) composed of reorientation of the flow at the first aperture (“2.5D approach”). This ansatz is common in analyses of inline mixers (refer, e.g., to Refs. 1, 3, 24, and 25) and implicitly assumes relatively elongated mixing cells (i.e., the cylinder segments associated with individual apertures) such that cell-wise transition is negligible for the total flow field, admitting its approximation by discontinuous change-overs at cell interfaces. Moreover, fluid inertia is omitted altogether. However, both transition and inertia may, depending on design and operating conditions, significantly affect flow and transport. The underlying changes in Lagrangian mechanisms remain unclear, though. The 2.5D system possesses a well-defined Hamiltonian structure, causing the Lagrangian transport to be governed by Hamiltonian mechanics. This results in properties and behavior that, basically irrespective of fluid rheology, hold universally and differ only quantitatively between individual systems. Key issue to be addressed in the study is the impact of three-dimensionality and fluid inertia on the Lagrangian behavior and kinematic structure of the RAM flow. This will be investigated by theoretical studies and numerical simulations.

The exposition is organized as follows. Problem definition and flow models (including numerical methods) are given in Sec. II. Properties of the 3D flow field are examined in Sec. III. Section IV investigates the associated flow topology and Lagrangian transport characteristics. Conclusions are drawn in Sec. V.

II. PROBLEM DEFINITION AND FLOW MODELS

A. 3D Flow model

Figure 1 gives a schematic of the Rotated Arc Mixer (RAM). It consists of two snugly fit concentric cylinders in which an axial throughflow with mean velocity \(U\) is created by a constant pressure drop. The inner cylinder is stationary and accommodates an axial sequence of apertures (“windows”) of arc angle \(\Delta\) and axial length \(L\); the subsequent windows are offset in azimuthal direction by an angle \(\Theta\). The outer cylinder rotates at a constant angular velocity \(\Omega\) and drives a transverse flow via the windows by viscous drag. The window-wise offset systematically reorients the transverse flow and thus accomplishes cross-wise mixing of the axial throughflow.

Offsets \(\Theta\) are in this study restricted to values commensurate with \(2\pi\), i.e., \(\Theta = 2m\pi/n\), meaning the window sequence repeats itself after \(N\) windows (or \(N\) “mixing cells”). An infinite repetition of such sequences of \(N\) cells is assumed, which admits reduction of the infinitely-long flow domain to the first \(N\) cells with periodic inlet-outlet. Thus the flow domain in the study hereafter is defined as \(D = \{r, \theta, z\} \in [0, R] \times [0, 2\pi] \times [0, NL]\). The flow is assumed to be incompressible and
B. Non-dimensional form and control parameters

such that the constant forcing conditions yield a steady flow. Under this premise, the flow is isothermal and the fluid is considered Newtonian. Furthermore, the flow is assumed to be laminar such that the constant forcing conditions yield a steady flow. Under this premise, the flow is isothermal and the fluid is considered Newtonian. Furthermore, the flow is assumed to be laminar of the inlet; the \((x, y)\)-coordinates and \(z\)-coordinate correspond with the cross-sectional and axial positions, respectively.

\[
\begin{align*}
\nabla \cdot \mathbf{u} &= 0, \\
-\mu \nabla^2 \mathbf{u} + \rho \mathbf{u} \cdot \nabla \mathbf{u} + \nabla p &= 0, \\
\end{align*}
\]

(1)

describing mass and momentum conservation, respectively. These laws are given in the primitive variables \(p\) and \(\mathbf{u}\), where \(p\) is the pressure and \(\mathbf{u} = (u_x, u_y, u_z)\) is the fluid velocity. Quantities \(\rho\) and \(\mu\) denote the density and dynamic viscosity, respectively. The corresponding boundary conditions are

\[
\begin{align*}
u|_{z=0} &= \mathbf{u}|_{z=N_L}, \\
u_{r,z}|_{r=R} &= 0, \\
u_{\theta,z}|_{r=R} &= V_{\theta}(\theta - \bar{\theta}(z), z), \\
\end{align*}
\]

(2)

with \(V_{\theta}(\theta, z) = \Omega R [H(\theta) - H(\theta - \Delta)]\) the angular boundary velocity at the first window, \(\bar{\theta}(z) = \Theta \sum_{k=1}^{N} H(z - kL)\) the reorientation angle of the window at axial position \(z\) and \(H(\cdot)\) the Heaviside function.

The Lagrangian motion of passive tracers released in the flow field is governed by the kinematic equation

\[
\frac{dx}{dt} = \mathbf{u}(x), \quad x(0) = x_0,
\]

(3)

with formal solution \(x(t) = \Phi_t(x_0)\). This uniquely determines the current position \(x\) of a tracer along its Lagrangian trajectory for a given initial position \(x_0\). Note that the tracer trajectories identify with streamlines in the present context of steady flows.

B. Non-dimensional form and control parameters

The non-dimensional form of the flow model follows from the scaling \(\tilde{x} = x/R, \tilde{u} = u/U\), and \(\tilde{p} = p/P\), with \(P = \mu U/R\), where tildes indicate dimensionless variables. Note that the characteristic pressure assumes momentum conservation to be dominated by a balance between pressure gradient and viscous forces. Substitution into the flow model yields \(\tilde{D} : (\tilde{r}, \tilde{\theta}, \tilde{z}) \in \left[0, 1\right] \times \left[0, 2\pi\right] \times \left[0, N\Lambda\right]\) as non-dimensional flow domain and

\[
\begin{align*}
\nabla \cdot \tilde{\mathbf{u}} &= 0, \\
R e \tilde{r} \tilde{u} \cdot \nabla \tilde{u} &= -\nabla \tilde{p} + \nabla^2 \tilde{u},
\end{align*}
\]

(4)

as corresponding conservation laws. The associated non-dimensional parameters are window opening angle \(\Lambda\), offset \(\Theta\), aspect ratio of one mixing cell \(\Lambda = L/R\), Reynolds number \(Re = U R / \nu\) (with \(\nu\) the kinematic viscosity) and ratio of axial to transverse flow \(\gamma = U / \Omega R\). An alternative to \(\gamma\) exists in \(\beta = \Lambda / \gamma = \Omega L / U\) (ratio of cell-wise residence time \(\tau_{cell} = L/U\) to eddy turn-over time \(\tau_e = \Omega^{-1}\) of the transverse flow), which is adopted hereafter for consistency with literature.

The non-dimensional kinematic equation follows from (3) upon rescaling time as \(\tilde{t} = t / \tau_e\) and retains the form of its dimensional counterpart. This yields no further parameters, meaning
that the full set of non-dimensional parameters comprises \{\Delta, \Theta, \Lambda, Re, \beta\}. The following study adopts fixed aspect ratio \Lambda = 2 and window angle \Delta = \pi/4 and restricts the offset to \Theta = \pm 2\pi/3, implying a RAM consisting of \(N = 3\) mixing cells. The remaining parameters are varied within the ranges \(\beta \in [0, 15]\) and \(Re \in [0, 50]\). Moreover, tildes indicating non-dimensional variables are omitted below. The study in Ref. 21 exposed fundamentally different behavior for \(-\pi < \Theta < 0\) and \(0 < \Theta < \pi\), respectively denoted negative and positive offset. This nomenclature is adopted here as well.

The analysis below will in part be performed relative to local references frames \(x'\) for each mixing cell \(k\). Global and local frames relate via the coordinate transformation
\[
F_k : x = (r, \theta, z) \rightarrow x' = (r, \theta - \theta_k, z - z_k),
\]
where \(\theta_k = \Delta/2 + \Theta(k - 1)\) and \(z_k = (2k - 1)\Lambda/2\) \((k = 1, 2, \ldots, N)\) are symmetry axis of window and midplane for cell \(k\), respectively. Within the cell-wise local reference frame \(x' = F_k(x)\), the symmetry axis bi-secting the window corresponds to the \(x'\)-axis and the cell inlet/midplane/outlet correspond to \(z' = -\Lambda/2, 0, \Lambda/2\).

### C. 2.5D flow model

Studies in the literature on inline-mixing flows typically concern 2.5D simplifications of the 3D system (Sec. I); the present study, on the other hand, considers the full 3D case. Here the corresponding 2.5D case will be included in the analysis as a reference for the actual 3D behaviour. This enables specific highlighting of similarities and differences between 2.5D and 3D representations of the RAM in particular and inline-mixing flows in general.

The 2.5D simplification of the RAM is defined in Refs. 20 and 21 and leans on decoupling between axial and transverse flows and omittance of the (essentially 3D) transition between consecutive mixing cells. Moreover, Stokes flow \((Re = 0)\) is assumed. The transverse flow \(\hat{u} = (\hat{u}_r, \hat{u}_\theta)\) is given by a cell-wise reorientation of the 2D analytical solution to the first cell \(\hat{u}^{(0)} = \hat{u}^{(0)}(r, \theta)\) by Hwu et al.\textsuperscript{31} the axial velocity \(u_z\) is given by the Poiseuille solution for pipe flow. Thus the 2.5D flow becomes
\[
\hat{u}(r, \theta, z) = \hat{u}^{(0)}(r, \theta - \hat{\theta}(z)), \quad u_z(r) = \frac{2\Lambda}{\beta}(1 - r^2),
\]
with \(\hat{\theta}(z)\) the reorientation angle at position \(z\) as before. This results in a discontinuous change-over in the transverse field at the cell interfaces. The axial flow, on the other hand, remains continuous due to its invariance to rotation. Moreover, the decoupling in flow components ensures that the discontinuity in transverse flow is immaterial for the continuity constraint.\textsuperscript{20} Note that here only the specific parameter combination \(\gamma = \Lambda/\beta\) (non-dimensional axial flow) is relevant, reducing the parameter set to \{\Delta, \Theta, \gamma\}. For the present case of fixed \(\Lambda\), this can be readily translated into \{\Delta, \Theta, \beta\} without loss of generality. Note that the 2.5D studies in Refs. 20 and 21 employ an identical parameter set, i.e., \{\Delta, \Theta, \beta_{2.5D}\}, for mixing cells with unit length \((\Lambda = 1)\). This implies that parameter \(\beta\) of the current configuration relates to \(\beta_{2.5D}\) of said 2.5D studies via \(\beta_{2.5D} = \beta/\Lambda\).

### D. Numerical methods

The 3D steady Navier-Stokes equations (4) are resolved in the entire mixer by way of the commercial computational fluid dynamics (CFD) package ANSYS-FLUENT. The employed settings are: double-precision implicit pressure-based solver for 3D steady laminar flows; pressure-velocity coupling via SIMPLE scheme; second-order upwind scheme for convective term; convergence criterion residuals \(\varepsilon = 10^{-12}\). This strategy has proven its worth in numerous computational analyses of inline industrial mixers.\textsuperscript{14-17,32} The non-dimensional flow domain (Sec. II B) is implemented in the dimensional solver by setting \(R = 1\) m and \(L = \Lambda\) \((= 2)\) m and is discretized by a computational mesh consisting of about \(2.5 \times 10^6\) tetrahedral elements per mixing cell. Standard grid-dependence tests determined this as sufficient for adequate resolution of the flow. Periodic boundary conditions as per (2) are imposed; the Dirichlet condition in (2) is prescribed via a user-defined function in
III. FLOW PROPERTIES

A. Non-inertial case \( Re = 0 \)

1. Generic properties of the velocity field

Periodic boundary conditions (2) and a net axial pressure drop suggest axial Fourier decomposition of the primitive variables \( p \) and \( u \) according to

\[
p(r, \theta, z) = -C z + \sum_{k = -\infty}^{\infty} \tilde{p}_k(r, \theta) e^{2\pi i k z / N \Lambda}, \quad u(r, \theta, z) = \sum_{k = -\infty}^{\infty} \tilde{u}_k(r, \theta) e^{2\pi i k z / N \Lambda},
\]

with \( (\tilde{p}_k, \tilde{u}_k) \) the Fourier modes, \( C > 0 \) the constant axial pressure gradient and \( i = \sqrt{-1} \). (Only the pressure gradient needs to be periodic in \( z \) in order to satisfy the conservation laws on the current periodic domain yet not the pressure itself. This admits non-periodic terms that are linear in \( z \).) The Fourier modes of the axial velocity component satisfy

\[
\langle \tilde{u}_{z,0} \rangle = \Phi / \pi > 0, \quad \langle \tilde{u}_{z,k} \rangle = 0 \quad \forall k \neq 0,
\]

with \( \langle g \rangle = \pi^{-1} \int_0^1 \int_0^{2\pi} g(r, \theta) r dr d\theta \) the cross-sectional average and \( \Phi = \int_0^1 \int_0^{2\pi} u_z r dr d\theta = \pi \langle u_z \rangle \) the volumetric flux (Appendix A). Thus flux \( \Phi \) is contained entirely in mode \( \tilde{u}_{z,0} \); Fourier modes \( \tilde{u}_{z,k} (k \neq 0) \) have zero net axial flux. Hence, \( \tilde{u}_{z,k} > 0 \) in some subregion implies \( \tilde{u}_{z,k} < 0 \) elsewhere so as to satisfy condition (8).

The non-inertial limit \( Re = 0 \) admits decomposition of the fields into \( p = p_1 + p_2 \) and \( u = u_1 + u_2 \) on account of the linearity of the Stokes equations. Splitting pressure as \( p_1 = -C z \) and \( p_2 = p - p_1 \) and boundary conditions as \( u_1|_{r=1} = 0 \) and \( u_2|_{r=1} = (u_{2,0}, u_{2,0}, u_{2,0})|_{r=1} = (0, 0) \) with \( V_0 \) according to (2), yields two separate flow problems: pressure-driven field \( u_1 = u_p \) and rotor-driven field \( u_2 = u_\Omega \). Thus the total flow decomposes as

\[
u = u_p + u_\Omega,
\]

where \( u_p = (0, 0, u_z(r)) \) identifies with the standard Poiseuille flow, with \( u_z = u_z(r) \) following (6). The \( z \)-components of contributions \( u_p \) and \( u_\Omega \) correspond with Fourier modes \( k = 0 \) and \( k \neq 0 \) of the net axial velocity \( u_z \), respectively (Appendix A). This, via (8), implies that the net axial flux \( \Phi \) is contained entirely in the pressure-driven component \( u_p \). Decomposition (9) has the important implication that the 3D Stokes flow in essence retains the structure of the 2.5D flow in Sec. II C. The principal difference is an essentially 3D rotor-driven component \( u_\Omega \) (i.e., dependent on all coordinates and including a non-zero axial component) due to cell-wise transition. Note that (9), similar to the 2.5D case, enables an efficient analysis in that numerical resolution only once of \( u_\Omega \) is required for each RAM geometry.
FIG. 2. Typical 3D flow field (top) and streamline portrait (center/bottom) demonstrated for positive offset at $\beta = 11$ and $Re = 0$. Panels (a) and (b), respectively, show transverse velocity $\hat{u}$ (magnitude: $0 \leq |\hat{u}| \leq 5.5$) and axial velocity $0 \leq u_z \leq 2.13$ at inlet/midplane/outlet of the mixing cells (blue/red indicate lower/upper bound). Dashed outlines indicate windows. Panels (c) and (d) give head-on upstream views of panels (c) and (d). Labels “L” and “H” in panel (b) indicate low/high-pressure zones due to transversal fluid acceleration/deceleration at upper/lower window edges; arrows indicate axial flow promoted by these zones. Color in panels (c)–(f) distinguishes streamlines with (cyan/blue) and without (red) backflow; blue indicates backflow segments.

A typical 3D velocity field is given in Fig. 2 (top) in the inlet/midplane/outlet of the mixing cells for positive offset and $\beta = 11$. The transverse flow (Fig. 2(a)) is appreciable only in the direct proximity of the windows. The axial flow (right) exhibits minor departures from a standard axisymmetric Poiseuille profile in the inlet/outlet regions; the axial velocity in the midplanes closely resembles this profile.

The departures near the cell inlets/outlets include localized backflow regions, i.e., where $u_z < 0$, which result from the formation of local low-pressure/high-pressure zones at the longitudinal edges of windows $1 \leq k \leq N$ due to transversal acceleration of the fluid by the rotor at the leading window edge $\theta = (k - 1)\Theta$ and subsequent deceleration upon reaching the trailing edge $\theta = (k - 1)\Theta + \Delta$. Former and latter zones are indicated with “L” and “H”, respectively, at window $k = 2$ in Fig. 2(b). The low-pressure/high-pressure zones promote flow towards/away from the window center, as indicated by the arrows in Fig. 2(b). This effect, if sufficiently strong, leads to backflow near the lower-left and upper-right corners of shown window and in the same way at the other windows. (Here
Backflow peaks at $\min u_z \approx -0.12$ in a highly-localized area at an axial distance of about $\Delta z \approx 0.1$ inwards from said corners. The associated streamline portrait, shown in Figures 2(c)–2(f), reveals that streamlines with backflow (cyan/blue) envelop a “core” of streamlines devoid of backflow (red). Backflow segments (blue) originate near the leading and trailing edges of the windows and, due to Poiseuille flow being positive everywhere, must emanate exclusively from the rotor-driven component $u_{2z}$ in (9). Note that backflow similar to that demonstrated here may occur under various conditions (i.e., is not specifically tied to a certain non-dimensional parameter). The backflow is weak compared to the main flow and diminishes the closer to the midplane. Figures 3(a) and 3(b) characterize the backflow by the velocity ratio $r_u \equiv u_{\text{back}}/(|u|)$ (left), with $u_{\text{back}} = \max |u_z|_{z < 0}$ the strongest backward velocity in the flow domain, and the flux ratio $r_\Phi \equiv \Phi_{\text{back}}/\Phi$ (right), with $\Phi_{\text{back}} = \iint u_z \, dx \, dy |_{z < 0}$ the strongest backward volumetric flux in the axial cross-section, versus $\beta$. Important to note is that non-zero $r_u$ and $r_\Phi$ does not signify net backflow from outlet to inlet; this typically concerns a local effect due to backflow segments on streamlines running from inlet to outlet (i.e., with net downstream flux). This reveals that, consistent with, significant backflow starts to occur upon $\beta$ exceeding a certain threshold, here $\beta \gtrsim 2$ (negative offset) and $\beta \gtrsim 4$ (positive offset). This can be understood by realizing that increasing $\beta$ signifies stronger transverse forcing, which promotes formation of vortical structures at window corners and edges. Moreover, the effect is far stronger for negative offset, where local magnitudes may become comparable to the main flow. However, the impact in all cases remains highly localized and thus of only marginal importance for the overall transport characteristics. This is evidenced by the weak backward flux (Fig. 3(b)), which even in the worst case amounts to just 10% of the net forward flux in a cross-section. Hence, further exploration of backflow is beyond the present scope.

2. Transition between mixing cells

Continuity dictates a smooth change-over in the flow field between consecutive mixing cells, precluding fully-developed (i.e., $z$-independent) cell-wise flow, thus introducing three-dimensionality to the flow field. This effect in fact underlies the departure of the axial flow $u_z$ from a Poiseuille flow in Fig. 2. Transition is absent in the 2.5D approximation according to Sec. II C and, in consequence, the axial flow identifies with a Poiseuille flow $u_z^P$ at any cross-section. Thus the departure of $u_z$ from the latter, defined following Ref. 22 as $\delta u_z(z) \equiv \langle u \rangle^{-1} \sqrt{\sum_{i=1}^M |u_z(x_i, y_i, z) - u_z^P(x_i, y_i)|^2} / M$ for the equidistant grid of $M$ evaluation points $(x_i, y_i)$ in a cross-section, may serve as measure for the effect of transition. Fig. 4(a) shows deviation $\delta u_z$ at 20 cross-sections in the second cell (expressed in local coordinates $x'$ according to (5)) of the RAM for positive offset and $\beta = \{3, 5\}$. This reveals that the strongest deviation happens at the inlet/outlet $z' = \pm \Lambda/2$ of the cell and the closest resemblance of the Poiseuille flow occurs in the midplane, consistent with Fig. 2. Deviation $\delta u_z$ is symmetric about the midplane $z' = 0$ – and always reaches its minimum $\epsilon_z \equiv \min \delta u_z(z') = \delta u_z(0)$ here – due to symmetries in the flow field for the non-inertial limit $Re = 0$ (Sec. III A 3). Growing $\beta$ amplifies the deviation and at some point even in the midplane gives rise to substantial departures from the

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**Image Description**: The figure shows a graph illustrating the backflow in the 3D non-inertial flow characterized by velocity ratio $r_u = u_{\text{back}}/(|u|)$ (left) and flux ratio $r_\Phi = \Phi_{\text{back}}/\Phi$ (right) as a function of $\beta$ for positive ($\Theta > 0$) and negative ($\Theta < 0$) offset. The graph indicates the backflow peaks at $\min u_z \approx -0.12$ in a highly-localized area at an axial distance of about $\Delta z \approx 0.1$ inwards from said corners. The associated streamline portrait, shown in Figures 2(c)–2(f), reveals that streamlines with backflow envelop a “core” of streamlines devoid of backflow. Backflow segments originate near the leading and trailing edges of the windows. The backflow is weak compared to the main flow and diminishes the closer to the midplane. Figures 3(a) and 3(b) characterize the backflow by the velocity ratio $r_u$ and the flux ratio $r_\Phi$, revealing that non-zero $r_u$ and $r_\Phi$ does not signify net backflow from outlet to inlet. However, the impact in all cases remains highly localized and of only marginal importance for the overall transport characteristics.
Poiseuille profile, as demonstrated in Fig. 4(b), signifying transition zones that cover the entire mixing cell. The impact of the transition is case-specific yet the same qualitative trend occurs in all cases: transition effects intensify with increasing $\beta$ and/or decreasing $\Delta$.

The fact that deviation $\delta u_z$ is symmetric and monotonic between midplane and inlet/outlet for $Re = 0$ facilitates distinction between transition zones ($\delta u_z > \varepsilon_k$) and internal (nearly) developed zones ($\delta u_z \leq \varepsilon_k$). Fig. 4(c) shows the proportion $r_\Lambda \equiv \Delta \Lambda / \Lambda$ of the total cell length $\Lambda$ occupied by the transition zones at inlet/outlet (with transition length $\Delta \Lambda$) versus $\beta$ for positive offset and $\varepsilon_k = \{5\%, 10\%\}$. This further consolidates the finding that increasing $\beta$ augments the transition effect. The $r_\Lambda - \beta$ diagram for negative offset (not shown) is nearly indistinguishable from Fig. 4(c), implying only a minor role of the offset in the emergence and extent of the transition itself.

The above revealed that transition between subsequent mixing cells has a significant effect on the flow field. Similar observations have been made for other inline mixers.14, 17, 36, 37 This means that simplifications such as the 2.5D approximation in Sec. II C in general compromise reliability of mixing analyses. However, a comparative study of the 2.5D/3D RAM revealed that deviations mainly concern location and extent of transport barriers yet not correct prediction of their emergence per se.22 Thus, the 2.5D method in general properly distinguishes poor and good mixing cases and is suitable for at least first exploratory studies. Detailed (quantitative) analysis necessitates fully 3D analysis, though.

3. Symmetries in the flow field

Symmetries in geometry and boundary conditions lead to symmetries in the flow field, which, in turn, have fundamental implications for the Lagrangian transport properties (Sec. IV). Boundary conditions (2) under laminar flow conditions yield an internal 3D field of the form

$$u(r, \theta, z) = \bar{u}(r, \theta - \bar{\theta}(z), \bar{z}(z)), \quad \bar{z}(z) = z \mod \Lambda,$$  

with $\bar{\theta}(z)$ as before and $\bar{u}(r, \theta, z)$ the 3D velocity field in the first cell $0 \leq z \leq \Lambda$. Thus the 3D field assumes the same form as the 2.5D approximation (6) yet with the essential difference that the 3D base flow $\bar{u}$ depends on $z$ (due to continuous change-over between cells). Important to note is that global symmetry (10) is upheld for both $Re = 0$ and $Re > 0$. The simulated velocity fields $u_{num}$ considered below satisfy (10) up to at least $\bar{u}_{num}(r, \theta, z) - \bar{u}_{num}(r, \theta - \bar{\theta}(z), \bar{z}(z)) \sim \mathcal{O}(10^{-3})$ relative to $|u_{num}| \sim \mathcal{O}(1)$; errors are attributed entirely to numerical effects.

The Stokes limit $Re = 0$, besides global symmetry (10), furthermore possesses internal symmetries within each cell. Expressing the flow field in the local reference frame (5) gives

$$u'_x(x') = cu_x \left(F_k^{-1}(x')\right) + su_y \left(F_k^{-1}(x')\right), \quad u'_y(x') = -su_x \left(F_k^{-1}(x')\right) + cu_y \left(F_k^{-1}(x')\right),$$  

and $u'_z(x') = u_z \left(F_k^{-1}(x')\right)$, with $c = \cos \theta_k$ and $s = \sin \theta_k$. The flow exhibits an internal symmetry in this frame, viz.,

$$u'_x(x', y', z') = -u'_x(x', -y', z'), \quad u'_y(x', y', -z') = u'_y(x', y', z'),$$  

$$u'_z(x', y', z') = u'_z(x', -y', -z'),$$

where $F_k^{-1}$ is the inverse Fourier transform. This further consolidates the finding that increasing $\beta$ augments the transition effect. The $r_\Lambda - \beta$ diagram for negative offset (not shown) is nearly indistinguishable from Fig. 4(c), implying only a minor role of the offset in the emergence and extent of the transition itself.
resulting in (anti-)symmetry about the \( x' \)-axis (i) \textit{within} the midplane \( z' = 0 \) (Figs. 5(a)–5(c)) and (ii) \textit{between} cross-sectional pairs \((z', -z')\) about this plane (Figs. 5(d)–5(o)). Hence, per cell two symmetry planes exist: \( \theta' = 0 \) and \( z' = 0 \). Former and latter translate into \( \theta_k = \Delta/2 + \Theta(k - 1) \) and \( z_k = (2k - 1)\Lambda/2 \), respectively, in global coordinates. Note that Figs. 5(j)–5(m) have the same (anti-symmetry as Figs. 5(d)–5(g)) yet in a less pronounced manner due to the closer proximity to the midplane \( z' = 0 \). The symmetry breaking about the \( x' \)-axis occurs solely in cross-sections within the transition zones; flow in the 2.5D simplification and developed regions in the 3D case is symmetric about the \( x' \)-axis.

Symmetries (12) interact with global symmetry (10) in cross-sections \( z' = \pm \Lambda/2 \). This results in a further symmetry about the axes \( \theta' = -\Theta/2 \) and \( \beta' = \Theta/2 \) following:

\[
\begin{align*}
\mathbf{u}_{11}(r', \theta', \pm \Lambda/2) &= -\mathbf{u}_{11}(r', 2\alpha - \theta', \pm \Lambda/2), \\
\mathbf{u}_{12}(r', \theta', \pm \Lambda/2) &= \mathbf{u}_{12}(r', 2\alpha - \theta', \pm \Lambda/2),
\end{align*}
\]

\[(13)\]

with \( \alpha = \pm \Theta/2 \) for \( \pm \Lambda/2 \). Here \( u_{1}' = \cos \alpha u_{x}' + \sin \alpha u_{y}' \) and \( u_{2}' = -\sin \alpha u_{x}' + \cos \alpha u_{y}' \) are the transverse velocity components relative to axes \( \theta' = \pm \Theta/2 \). Symmetry (13) is demonstrated in Fig. 6 and is essentially the same as (12) in the midplane \( z' = 0 \) yet about different axes. Note that the transverse magnitude \( u_{xy} = \sqrt{u_x'^2 + u_y'^2} = \sqrt{u_x'^2 + u_y'^2} = \sqrt{u_x'^2 + u_y'^2} \) is symmetric about \( \alpha = \pm \Theta/2 \). This, combined with the symmetry within midplane \( z' = 0 \), explains the symmetry in the transverse magnitude in Fig. 2.

### B. Impact of fluid inertia \((\text{Re} > 0)\)

Fluid inertia couples the components of the momentum via the nonlinear term \( \mathbf{u} \cdot \nabla \mathbf{u} \) and thus prohibits a strict separation into (and independent resolution of) pressure-driven and rotor-driven flow problems. However, decomposition (9) is nonetheless retained in that the flow field can still be expressed as a superposition of a Poiseuille flow \((\mathbf{u}_P)\) and a rotor-induced contribution \((\mathbf{u}_{12})\) with

\[
\begin{align*}
\mathbf{u} &= \mathbf{u}_P + \mathbf{u}_{12}, \\
\mathbf{u}_P &= \mathbf{u}_P(\alpha, \beta, z), \\
\mathbf{u}_{12} &= \mathbf{u}_{12}(\alpha, \beta, z).
\end{align*}
\]
Fluid inertia has fundamental consequences for the symmetry properties of the flow field. Global symmetry (10) is preserved for Re > 0, but internal symmetries (12) and (13) are broken. This is demonstrated in Fig. 7 by the transverse and axial flow fields in the midplane (z' = 0) and outlet (z' = A/2) of the mixing cells for Re = 0, 5, 10, and β = 5 (the vertical line separates positive (left) and negative (right) offsets). The profiles develop clear asymmetries about the local symmetry axes (dashed) already from Re = 0 contained entirely in the Poiseuille component of the flow.

Zero net axial flux (Appendix A). Note that for both Re = 0 and Re > 0 the net axial velocity uz of the 3D flow, respectively, given by (A6) and (A7), deviates from a Poiseuille flow. Hence, the net axial flux Φ is also for Re > 0 contained entirely in the Poiseuille component of the flow.

FIG. 6. (a)–(f) Internal symmetries within cross-sections z' = ±A/2 of each mixing cell in the Stokes limit Re = 0 demonstrated for β = 5 and positive offset. Results are represented in the local reference frame according to (5) and transverse velocity components (ux, uz) Blue/red indicate minimum/maximum values; dashed lines indicate local symmetry axes; the vertical line separates positive (left) and negative (right) offsets.

FIG. 7. (a)–(x) Symmetry breaking by fluid inertia demonstrated by the transverse (uxs) and axial (uz) flow fields in the midplane (z' = 0) and outlet (z' = A/2) of the mixing cells for β = 5. Results are represented in the local reference frame according to (5); blue/red indicate minimum/maximum values; dashed lines indicate local symmetry axes; the vertical line separates positive (left) and negative (right) offsets.
FIG. 8. Impact of fluid inertia on the cell-wise flow field (positive offset). (a) Symmetry breaking in deviation $\delta u_z$; (b) degree of asymmetry $\sigma$ versus Re; (c) proportion $r_\Lambda \Lambda$ occupied by the transition zones.

The symmetry breaking can be examined via the extent of the transition zones. Fig. 8(a) shows deviation $\delta u_z$ for $\beta = 11$ and positive offset as a function of Re. The Stokes limit $Re = 0$ exhibits symmetry about the midplane similar to Fig. 4(a). Fluid inertia profoundly affects the transition zones, as quantified by the degree of asymmetry $\sigma \equiv (2/L) \int_0^L |\delta u_z(z') - \delta u_z(-z')| \, dz$ in Fig. 8(b). Moreover, the minimum deviation $\varepsilon_z$, first, shifts away from the midplane towards the cell outlet and, second, increases substantially with growing Re. Thus fluid inertia, besides symmetry breaking, also strongly intensifies transition effects, as demonstrated by the growth of $r_\Lambda$ as a function of Re in Fig. 8(c). The corresponding $r_\Lambda - Re$ diagram for negative offset (not shown) practically coincides with Fig. 8(c), signifying a marginal role of the offset in the impact of fluid inertia on transition. Note that similar observations have been made regarding the effect of $\beta$ (Fig. 4). The trend in Fig. 8(c) can be estimated by scaling arguments. Disturbances (e.g., induced by reorientation at cell inlets) decay by viscous dissipation in a typical time span $t_v = R^2/\nu$, yielding $\Delta L \sim U t_v$ as their typical downstream region of influence. This gives $r_\Lambda = \Delta L/L = \Delta L/L \sim UR^2/\nu L = Re/\beta$ and suggests an approximately linear dependence on Re with a slope $\beta^{-1} \approx 0.09$, which is in reasonable agreement with the numerical predictions.

Fluid inertia, remarkably, tends to suppress backflow. Evaluation of $r_\Phi$ as a function of Re revealed a decline with stronger inertia (not shown) and backflow reduces roughly by a factor two within the range $0 \leq Re \leq 10$. This is believed to be a consequence of the fact that inertia causes fluid parcels to more strongly “resist” deflection from straight trajectories compared to the non-inertial case, thus promoting uni-directional downstream motion.

IV. FLOW TOPOLOGY OF GENERIC 3D RAM FLOWS

The present study examines mixing properties from a Lagrangian perspective by considering the coherent structures that form in the 3D streamline portraits. Such coherent structures define the flow topology and geometrically determine the transport properties of the flow. This Lagrangian approach has found successful application in the analysis of laminar inline-mixing flows. However, fundamental insight into the Lagrangian transport properties of such flows is mainly restricted to 2.5D cases on grounds of (i) their straightforward reduction to axial maps and (ii) their well-defined Hamiltonian structure. Three-dimensionality and fluid inertia introduce conceptual and technical complications (due to, e.g., reversal or symmetry breaking of the underlying flow field; Sec. III) that render extension and generalization of the Lagrangian framework of the 2.5D simplification to generic 3D inline flows non-trivial. This generalization is worked out below in a rigorous and step-by-step way. This employs generic concepts from dynamical-systems theory.

A. Existence of net throughflow regions

The mixing properties hinge on specific topological entities in the 3D streamline portrait. Key organizing entities are isolated stagnation points (possibly merged into stagnation lines) and closed
streamlines. The latter may emerge in two ways in the current periodic domain. First, periodic streamlines, i.e., streamlines connecting inlet and outlet at the same cross-sectional position after \( P \) spatial periods: 

\[
x_0 = (r_0, \theta_0, 0) \rightarrow x_P = (r_0, \theta_0, PN/\Lambda_w)
\]

Second, internal closed streamlines (i.e., disconnected from any domain boundary). (So far there is no evidence of closed internal streamlines in the current example flow. However, they do occur in similar flow configurations. Refer, e.g., to Ref. 42 for emergence of internal closed stream tubes in a micro-channel with net axial flow, which implies internal closed streamlines. Hence, these entities are included for generality of the analysis.) Isolated stagnation points are invariably accompanied by 1D/2D manifold pairs (delineating the principal transport directions in their zones of influence); periodic/closed streamlines are either elliptic or hyperbolic. The former define the centers of continuous families of (closed) concentric tubes; the latter define the origin of 2D/2D manifold pairs. Note that 1D and 2D manifolds correspond with individual streamlines and stream surfaces, respectively.

Tubes of elliptic/closed streamlines are completely separated from other coherent structures and in that sense demarcate isolated flow regions. Manifolds, on the other hand, may exhibit various point-line and line-line interactions (essentially smooth merger or intersection) and thus may give rise to intricate topological structures. (Interactions between manifolds originating from different points or lines are termed “heteroclinic”.) Relevant in the present context is the heteroclinic interaction between manifolds of hyperbolic periodic lines with those of hyperbolic closed lines and stagnation points. This interaction is investigated below on the basis of the elementary heteroclinic interactions between points and closed streamlines in Ref. 40. (Note closed streamlines are denoted “cycles” in Ref. 40.) Generalizing the line-line interactions examined in Ref. 40 to include both periodic and closed lines results in:

**Conjecture 1. Hyperbolic periodic streamlines cannot interact heteroclinically with hyperbolic closed streamlines**

**Rationale.** Such heteroclinic interaction can occur in two ways: (i) formation of a continuous heteroclinic surface; (ii) intersecting manifolds. 2D manifolds are “extrusions” of their underlying streamlines: extruding a periodic line defines an open surface, which is topologically a flat plane; extruding a closed periodic line defines a tube-like surface, which is topologically a cylinder. The different topologies preclude a smooth merger of former and latter kind of surface, meaning that heteroclinic surfaces cannot form between periodic and closed lines. Intersection yields two heteroclinic trajectories that must intersect the periodic line. This implies stagnation points on the latter, which is in conflict with the uni-directional motion along a periodic line.

Conjecture 1 dictates a strict separation between manifolds of periodic lines and those of closed lines. This may in principle happen in three ways: (i) generic separatrices (e.g., symmetry planes); (ii) encapsulation of periodic-line entities by 2D manifolds of closed lines; (iii) encapsulation of closed-line entities by 2D manifolds of periodic lines. However, situation (ii) prohibits the periodic lines from connecting periodic inlet/outlet and thus is impossible. Intersecting manifolds of periodic lines, similar to those of closed lines, form (highly-convoluted and infinitely-long) stream tubes that accommodate uni-directional transport from one line to the other. Such tubes can therefore not hold closed streamlines, thus ruling out situation (iii) for periodic lines with intersecting manifolds. This leads to:

**Conjecture 2. Hyperbolic periodic lines and hyperbolic closed streamlines (and associated manifolds) can coexist only in subregions divided by generic separatrices or non-intersecting manifolds of hyperbolic periodic lines.**

Similar considerations result in an equivalent for the interaction between periodic lines and stagnation points:

**Conjecture 3. Hyperbolic periodic lines and isolated stagnation points (and associated manifolds) can coexist only in subregions divided by generic separatrices or non-intersecting manifolds of hyperbolic periodic lines.**
**Rationale.** Manifolds of periodic lines and isolated stagnation points can interact in two ways. First, 2D manifolds of periodic lines versus 1D manifolds of points. Intersection yields new stagnation points and therefore cannot occur. Hence, generically 1D manifolds of points can exist only in subregions demarcated by 2D manifolds of periodic lines. (Heteroclinic merger of 1D and 2D manifolds is in principle possible yet non-generic and thus highly improbable. Compare with essentially similar situations for closed streamlines in Ref. 40). Moreover, on the same grounds as for closed streamlines, only subregions bounded by non-intersecting 2D manifolds can hold stagnation points. Second, 2D manifolds of periodic lines versus 2D manifolds of points. The former and latter topologically are flat planes and disks, respectively, and thus cannot merge into a continuous surface. Intersection, on the other hand, is possible yet only between 2D manifolds of opposite stability. This restricts such interaction to periodic lines with non-intersecting manifolds. One intersection between the manifolds of a line, namely, implies an infinite set, precluding intersection of the 2D manifolds of stagnation points with one stability type only.

Conjectures 2 and 3 preclude closed streamlines and stagnation points existing in regions occupied by intersecting manifolds of hyperbolic periodic lines, or equivalently, in chaotic zones induced by such lines. However, intersecting 2D manifolds – and, inherently, chaos – is the generic situation for hyperbolic periodic lines.40 These considerations strongly suggest that, save exceptional conditions, periodic lines and associated tubes and manifolds normally are strictly separated from (entities associated with) stagnation points and closed streamlines. This implies that periodic lines, if existent, create an isolated “net throughflow region” between inlet and outlet by which mixing of the supplied material and its net axial transport occurs.

A net throughflow region must always exist by virtue of a non-zero net axial flux $\Phi > 0$ (Appendix A) and in its most general form consists of a set of $M$ coexisting “throughflow tubes” $\mathcal{N}_m (m = 1, \ldots, M)$ that each connect inlet and outlet. In the 2.5D case, isolated stagnation points are absent and the entire flow domain always constitutes one throughflow tube: $M = 1$ and $\mathcal{N}_1 = D$. Similarly, efficient global mixing, the ultimate objective of inline mixers, also results in one global net throughflow region (Appendix B). However, in general tubes $\mathcal{N}_m$ may coexist with flow regions associated with closed streamlines and/or isolated stagnation points (e.g., internal recirculation zones comparable to Ref. 42). Brouwer’s fixed-point theorem implies that each $\mathcal{N}_m$ accommodates at least one periodic streamline on account of periodicity and convexity of their cross-sections (which topologically are disks). Hence, tubes $\mathcal{N}_m$ and periodic lines are inextricably linked. Arbitrary streamlines within tubes $\mathcal{N}_m$, in consequence, belong either to chaotic zones or elliptic tubes and, notwithstanding possible local backflow $u_z < 0$, always accommodate net axial flow from inlet to outlet (Appendix B). For the situation shown in Fig. 2, the net throughflow region, akin to the 2.5D and global-chaos cases, takes up the entire flow domain. Here all streamlines, including those with backflow, run from inlet to outlet. The net throughflow region is key to the functioning of the RAM (and inline mixers in general) and the principal subject of investigation for the remainder of this study.

**B. Hamiltonian structure**

The net throughflow region accommodates a 3D steady volume-preserving flow devoid of stagnation points (Conjecture 3). This admits transformation of kinematic equation (3) into a 2D time-periodic Hamiltonian system with (canonical) time associated with the throughflow from inlet to outlet. The transformation generically involves two steps: (i) transformation $\mathcal{G}$ from Cartesian coordinates $x = (x, y, z)$ to right-handed orthonormal curvilinear coordinates $\xi = (\xi_1, \xi_2, \xi_3)$; (ii) transformation $\mathcal{H}$ from physical reference frame $\xi = (\xi_1, \xi_2, \xi_3)$ to canonical space-time reference frame $\xi = (\zeta_1, \zeta_2, \tau)$. Thus the full transformation formally reads

$$
\frac{dx}{dt} = u(x) \quad \Rightarrow \quad \frac{d\xi}{dt} = v(\xi) \quad \Rightarrow \quad \nu \left( \frac{d\xi_1}{d\tau}, \frac{d\xi_2}{d\tau} \right) = \left( \frac{\partial H}{\partial \zeta_1}, \frac{\partial H}{\partial \zeta_2} \right),
$$

with $\mathcal{G}$ and $\mathcal{H}$ elaborated in the supplementary material.43 Here transformation $\mathcal{H}$ employs the methodology by Ref. 44. Important to note is that non-trivial transformation $\mathcal{G}$ is necessary only for cases with backflow; frame $\xi$ can identify with the original Cartesian reference frame otherwise.
The canonical time \( \tau \) concerns evolution along curvilinear coordinate \( \xi_3 \); the canonical spatial coordinates \( (\xi_1, \xi_2) \) concern the corresponding Hamiltonian dynamics transverse to this evolution. In both the 2.5D case and 3D case without backflow, canonical time and space are tied to the z-axis and \((x, y)\)-positions in an arbitrary axial cross-section, respectively. In the 3D case with backflow, this is generalized to the curvilinear coordinates \( \xi_1 \) and \( (\xi_1, \xi_2) \) so as to ensure a monotonic relation between throughflow and canonical time. Retention of the Hamiltonian structure by the generic 3D system has the fundamental ramification that the relevant Lagrangian transport properties, despite three-dimensionality and fluid inertia, remain governed by the Hamiltonian mechanisms of a 2D time-periodic flow. The period time \( \mathcal{T} \) identifies with the arc length of the coordinate axis of the streamwise coordinate \( \xi_3 \) (refer to Ref. 43) and relates to the total length of the RAM via \( \mathcal{T} = cN\Lambda \), with \( c > 0 \) in general and \( c = 1 \) in cases without backflow. This gives \( \mathcal{T} = cNY\beta \) through the scaling following Sec. II B and, via \( \gamma > 0 \), yields \( \mathcal{T} \propto \beta \). Thus period time \( \mathcal{T} \) and kinematic parameter \( \beta \) relate monotonically: \( d\mathcal{T}/d\beta > 0 \) and \( \mathcal{T}(0) = 0 \).

C. Reduction to axial maps

Kinematic equation (3) admits for 2.5D flow straightforward reduction to an axial map between subsequent cross-sections following:

\[
\Phi_{3D} : (x_k, y_k, z_k) \rightarrow (x_{k+1}, y_{k+1}, z_{k+1}), \quad x_{k+1} = \Phi(x_k), \quad z_{k+1} = z_k + N\Lambda, \tag{15}
\]

on account of the uni-directional axial flow, i.e., \( u_z > 0 \) everywhere, which acts as a progression in time.\(^{21}\) Here \( x = (x, y) \) is the cross-sectional reference frame in an axial cross-section \( \mathcal{A}_z : z = z_0 \), with \( 0 \leq z_0 \leq \Lambda \) an (in principle arbitrary) axial position in the first cell. Consecutive cross-sections \( z_k \) in an infinite RAM correspond with periodic crossings of \( \mathcal{A}_z \) in its current periodic representation. Here map \( x_{k+1} = \Phi(x_k) \) describes the period-wise evolution of tracer positions \( x_k \) within \( \mathcal{A}_z \) and is analogous to a time-periodic progression of the 2D cross-sectional system.

For 3D flow fields such a reduction is a priori non-trivial due to the possibility of backflow (Sec. III A 1). The axial position, namely, evolves non-monotonically on backflow segments (e.g., the blue segments in Fig. 2) and thus cannot act as time. Transformation \( \mathcal{G} \) in (14) incorporates this essential monotonicity in the curvilinear coordinates \( \xi \) and thus enables reduction to a map map adopts the same form as (15) yet now in the \( \xi \)-frame.

The (practical) link with axial cross-sections can be re-established for those \( \mathcal{A}_z \) with \( u_z > 0 \) everywhere. Here intersections of the 3D streamline portrait with \( \mathcal{A}_z \) and \( \mathcal{A}_x \), namely, are topological identical (and thus interchangeable), since increasing \( z \) locally corresponds with progression in canonical time \( \tau \). The analysis hereafter assumes that at least one such axial cross-section \( \mathcal{A}_z \) can be designated, which can be done without loss of generality. This enables expression of the map associated with \( \mathcal{A}_z \) as an axial map following (15). The sequence of positions of a given tracer,

\[
\mathcal{X}(x_0) = \{x_0, x_1, \ldots, x_k, \ldots\}, \tag{16}
\]

are the subsequent intersections of its corresponding (periodically re-entering) 3D streamline with cross-section \( \mathcal{A}_z \) and constitute its so-called Poincaré section. Coherent structures in the Poincaré section are inextricably linked to those in the 3D streamline portrait. Periodic lines in the net throughflow region emerge as periodic points \( \Phi^p(x_0) = x_0 \) of the same type (i.e., elliptic or hyperbolic), with \( p \) the number of cycles prior to returning to the initial position, in the corresponding Poincaré section. Moreover, tubes of elliptic periodic lines appear as islands in these sections. Any map \( \Phi \) for any \( R \geq 0 \) and associated with any cross-section \( \mathcal{A}_z \) has at least one period-1 point on account of Brouwer’s fixed-point theorem: \( \Phi(x_0) = x_0 \). Thus map \( \Phi \) enables investigation of the flow topology both in terms of the dynamics within a cross-section \( \mathcal{A}_z \) and 3D streamlines. (This principle underlies any Lagrangian analysis of inline-mixing flows.\(^{[1,3,20-22]} \)) Both representations will be employed here.
D. Symmetries

Symmetries in the flow field manifest themselves as symmetries in the tracer trajectories and, inherently, in the flow topology. Thus symmetries may offer important insight into the transport properties and must therefore be an integral part of any Lagrangian transport analysis.\textsuperscript{21,29,45,46} Two kinds of symmetries are relevant here: global symmetry between mixing cells and internal symmetries within mixing cells (Sec. III A 3).

1. Global symmetry between mixing cells

The global symmetry \((10)\) in the flow field carries over to the map \(\Phi\) in axial cross-sections \(A_z\) following \((15)\) (holding for 2.5D and 3D model; Sec. IV C). This implies the composite form

\[
\Phi = \Phi_N \Phi_{N-1} \cdots \Phi_1, \quad \Phi_k = R^{k^{-1}} \Phi R^{1-k} \Rightarrow \Phi = (R^{-1} \Phi)^N,
\]

(17)

with \(R: \theta \rightarrow \theta + \Theta\) the reorientation operator and \(\Phi\) the cross-sectional component associated with the axial map \(\Phi_{1D}: (x, z_0) \rightarrow (\Phi(x), z_0 + \Lambda)\) from initial cross-section \(z_0\) over one cell length (i.e., to cross-section \(z_0 + \Lambda\)). Composition \((17)\) is essentially the same as in the 2.5D case\textsuperscript{21} and also holds for maps \(\Phi'\) and \(\Phi''\) relative to reference frame \(x'\) following \((5)\). Important to note is that symmetry \((17)\) holds for \(Re \geq 0\).

2. Internal symmetry within mixing cells

The cell-wise internal flow symmetries \((12)\) and \((13)\) of the non-inertial limit \(Re = 0\) result in symmetries of the base map \(\Phi'\) in reference frame \(x'\). Relevant in the present context are so-called time-reversal symmetries, which have the generic form

\[
\Phi' = S_a \Phi'^{-1} S_a, \quad S_a : \theta' \rightarrow \alpha - \theta',
\]

(18)

with symmetry operator \(S_a = S_{a}^{-1}\) reflecting about the associated symmetry plane \(I_S: \theta' = \alpha/2\).

Symmetries \((12)\) and \((13)\) translate into time-reversal symmetries\textsuperscript{21}

\[
\Phi_{-\Lambda/2}^I = S_0 \Phi_{-\Lambda/2}^I S_0, \quad \Phi_{0}^I = S_\Theta \Phi_{0}^I S_\Theta, \quad \Phi_{\Lambda/2}^I = S_{2\Theta} \Phi_{\Lambda/2}^I S_{2\Theta},
\]

(19)

for base maps \(\Phi'\) associated with the distinct cross-sections \(z'_0 \in \{-\Lambda/2, 0, \Lambda/2\}\). Other cross-sections have a symmetry

\[
\Phi_{\alpha}^I = S_\Theta \Phi_{\alpha}^I S_{-\alpha},
\]

(20)

between pairs \(z'_0 \in \{-\alpha, a\}\). Note that the above single reflections in the cross-sectional frame \(x'\) correspond with double reflections in the 3D frame \(x\), i.e., \(\Phi_{1D} = S_{1D} \Phi_{3D}^{-1} S_{1D}\), with \(S_{1D} = S_a S_{c}\) and \(S_{c}: z' \rightarrow -z'\). Substitution of symmetries \((19)\) and \((20)\) into the global symmetry \((17)\) yields

\[
\Phi' = R^{-1}(S_a \Phi'^{-1} S_a) \cdot R^{-1}(S_a \Phi'^{-1} S_a) = S_{r} \Phi'^{-1} S_{r}, \quad S_{r} = R^{-1} S_a, \quad \gamma = \alpha - \Theta,
\]

(21)

with associated symmetry plane \(\theta' = (\alpha - \Theta)/2\), which is essentially the same as in Ref. 21. This yields the specific planes

\[
\theta'_{-\Lambda/2} = -\Theta/2, \quad \theta'_{\Lambda/2} = \Theta/2, \quad \theta'_{\pm \alpha} = 0,
\]

(22)

in the \(x'\)-frame. For the present configuration this gives (in the above order) sets of angles \(\{-\pi/3, 0, \pi/3\}_{0=2\pi/3}\) and \(\{\pi/3, 0, -\pi/3\}_{0=2\pi/3}\) for positive and negative offset, respectively. Former and latter (via \(\theta = \theta' + \Delta/2\)) become \(\{-5\pi/24, \pi/8, 11\pi/24, \pi/8\}_{0=2\pi/3}\) and \(\{11\pi/24, \pi/8, -5\pi/24, \pi/8\}_{0=2\pi/3}\) in the original reference frame \(x\).

Time-reversal symmetries \((20)\) constrain the Poincaré sections for the non-inertial limit \(Re = 0\) and, inherently, the 3D flow topology. For cross-sections \(z'_0 \in \{0, -\Lambda/2, \Lambda/2\}\) they imply symmetry within Poincaré sections; for other cross-sections they imply symmetry between Poincaré sections \(z' = -a\) and \(z' = a\). These symmetries are demonstrated in Fig. 9 for map \(\Phi\) following \((15)\) in the indicated cross-sections \(z'_0 = z_0 - \Lambda/2\) at \(\beta = 5\) and negative offset. Note that Poincaré
sections for $z_0' = \pm \Lambda/2$ possess self-symmetry about $\theta' = \pm \Theta/2$ and, by virtue of symmetry (20), simultaneously form symmetric pairs about $\theta' = 0$.

The symmetry analysis reveals that the 3D RAM flow for $Re = 0$ in essence exhibits the same time-reversal symmetry in its flow topology as the 2.5D case studied in Ref. 21. This has, in conjunction with the Hamiltonian structure established in Sec. IV B, the fundamental implication that the non-inertial 3D RAM flow is dynamically equivalent to its 2.5D counterpart. Differences between 3D and 2.5D cases are entirely quantitative in a manner akin to the effect of variable rheology considered in Ref. 21. Thus the generic structure and transport properties of the 2.5D case determined in Ref. 21 generalize to the (net throughflow region of the) 3D non-inertial RAM flow.

E. Actual system state as departure from an integrable state

The behavior of Hamiltonian systems is generally investigated in terms of perturbations of so-called integrable states (i.e., states with a priori known non-chaotic dynamics). Here a given system state is effectively considered as the perturbation of – or, equivalently, the departure from – such an integrable limit. This same approach is employed for the RAM in order to further strengthen its connection with generic Hamiltonian mechanics. To this end kinematic parameter $\beta$ is adopted as “perturbation parameter” that controls the departure from integrability. Diminution of transverse forcing by decreasing $\beta$ causes the 3D flow for any $Re \geq 0$ to approach a decomposition of the form (9), with $u_{\Omega} \sim \mathcal{O}(\beta)$ here a weak rotor-induced 3D flow superimposed upon a Poiseuille profile $u_P$. The pipe-flow limit $\beta = 0$ (provided laminar conditions) for any $Re \geq 0$ is Poiseuille flow. The 2.5D flow asymptotes towards this same pipe-flow limit for diminishing $\beta$. This means that all RAM configurations share a weakly-perturbed Poiseuille flow as universal limit for vanishing $\beta$. The 2.5D analysis in Ref. 21 revealed that only the averaged transverse flow $u_{\Omega}(r, \theta, \zeta) = \int_0^{N\Lambda} u_{\Omega}(r, \theta, z) dz/N\Lambda$ is relevant to the transport in this limit. The fact that the 2.5D and 3D cases converge on the same limit state for $\beta \to 0$ then implies that this situation extends to the 3D case for any $Re \geq 0$. The flow then assumes the form

$$u = u(r, \theta) = u_P(r) + u_{\Omega}(r, \theta), \quad u_{\Omega}(r, \theta) = \sum_{k=0}^{N-1} \bar{u}^*(r, \theta - k\Theta), \quad \bar{u}_{\Omega}(r, \theta) = \int_0^{\Lambda} \bar{u}_{\Omega}(r, \theta, z) dz/\Lambda,$$

(23)

where expression in terms of the 3D velocity field in the first cell, i.e., $\bar{u}(r, \theta, z)$, follows from property (10). The $z$-independence of the averaged transverse flow $\bar{u}_{\Omega}$ (via volume-preservation) implies the Hamiltonian form

$$\nabla \cdot \bar{u}_{\Omega} = \partial \bar{u}_{\Omega,x}/\partial x + \partial \bar{u}_{\Omega,y}/\partial y = 0 \quad \Rightarrow \quad (\bar{u}_{\Omega,x}^*, \bar{u}_{\Omega,y}^*) = (\partial \bar{H}^*/\partial y, -\partial \bar{H}^*/\partial x),$$

(24)

using Cartesian coordinates for simplicity of notation. Thus the transverse motion of fluid parcels throughout the entire RAM is governed by the global autonomous Hamiltonian system

$$\left(\frac{dx}{dt}, \frac{dy}{dt}\right) = \left(\frac{\partial H^*}{\partial y}, -\frac{\partial H^*}{\partial x}\right), \quad H^*(r, \theta, \Phi) = \frac{1}{N} \sum_{k=0}^{N-1} \bar{H}^*(r, \theta - k\Theta),$$

(25)
with $\hat{H}^*$ the Hamiltonian function according to (24). Hamiltonian system (25) defines the integrable limit of (14) and exists for any RAM configuration, irrespective of the other control parameters $\{\Delta, \Theta, \Lambda, Re\}$. This limit is of the same form as found for the corresponding 2.5D case. Fundamental consequence is that fluid parcels are restricted to invariant surfaces defined by axial extrusion of isopleths of $H^*$ in the cross-section. These isopleths coincide with the 2D streamline portrait of flow field ($u^*_z, u^*_{\beta z}$) in the $xy$-plane and, given an impenetrable cylindrical boundary, for $N \geq 3$ constitute a continuous family of concentric tubes centered on the $z$-axis that connect inlet and outlet. The associated Poincaré section consists entirely of one global elliptic island centered on the origin. System (25) admits transformation into so-called action-angle form for $N$ (midplane 113601-17 Speetjens et al. Phys. Fluids for axis.)

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FIG. 10. (a)–(h) Hamiltonian breakdown of the integrable limit demonstrated by the Poincaré section of map $\Phi$ following (15) (midplane $z_0 = 0$) for 3D non-inertial flow (top) versus 2.5D flow (bottom) at positive offset. (Dashed line: symmetry axis.)

\begin{align}
I_{k+1} &= I_k = I_0, \quad \eta_{k+1} = [\eta_k + \partial(I_0) \kappa(I_0, \eta_k)] \mod \eta^*(I_0), \\
\end{align}

with $\kappa = \kappa(I_0, \eta_k)$ the time for a fluid parcel at position $(I_0, \eta_k)$ to travel from inlet $z = 0$ to outlet $z = NA$, given implicitly by $\int_0^\eta u_z [r(I_0, \eta(\epsilon)), \partial(I_0, \eta(\epsilon))] d\epsilon = NA$. Form (26) is a so-called circle map and provides an important link between the RAM and generic integrable Hamiltonian systems and their response to perturbations. Any state can, namely, be regarded as the departure from integrable state (25) in that it can be attained by continuously increasing $\beta$ from zero to its actual value. This progression from integrable to actual state involves (partial) breakdown of the former state by Hamiltonian mechanisms and has both universal and case-specific elements. The breakdown of the original island is governed by universal scenarios described by the well-known KAM theorem in case of non-resonant orbits ($\partial \kappa$ in (26) non-commensurate with $\eta^*$) and by the Poincaré-Birkhoff theorem in case of resonant orbits ($\partial \kappa$ in (26) commensurate with $\eta^*$). Parameters $\{\Delta, \Theta, \Lambda, Re\}$ have a case-specific impact by quantitatively affecting these scenarios by, e.g., inducing bifurcations in the state (e.g., emergence of new islands) or mode locking.

Fig. 10 demonstrates the Hamiltonian breakdown of the integrable state at positive offset for the 3D non-inertial case (top) and the 2.5D case (bottom) by the Poincaré sections in $z_0 = 0$. This reveals the “classical” picture of Hamiltonian systems: progressive disintegration of the global island of the
FIG. 11. (a)–(d) Hamiltonian breakdown of the integrable limit demonstrated by the Poincaré sections of map $\Phi$ following (15) (midplane $z'_0 = 0$) for 3D non-inertial flow at negative offset. (Dashed line: symmetry axis.)

integrale state in favor of chaotic regions. The disintegration is accompanied by the emergence of the characteristic island chains (most prominent in panel (f)). The remnant of the original island corresponds with an elliptic period-1 point on the symmetry axis (dashed) that gradually shifts towards the left with increasing $\beta$. This results from an interplay between time-reversal symmetry (19) and the positive offset, established for the 2.5D case in Ref. 21 and here generalizing to the 3D non-inertial case. Essentially, the same mechanism causes the emergence of a new elliptic period-1 point on the right segment of the symmetry axis upon exceeding a certain $\beta$-threshold (here $\beta \approx 3$). This underlies the banana-shaped island squeezed against the cylinder wall for $\beta = 5$, 11. Fig. 11 gives the Hamiltonian breakdown for the 3D non-inertial case at negative offset. Here the original island vanishes completely, giving way to a state of almost global chaos, save the small islands near the wall. (The minute remnant of the original island in the center of the chaotic sea disappears at $\beta \approx 11.5$.) The behavior shown in Figures 10 and 11 is (at least qualitatively) representative for any RAM configuration in that non-zero $\beta$ always triggers disintegration of the island(s) of the integrable limit. This reflects the beforementioned universality of the system characteristics. Different conditions affect the progression primarily by the extent to which full disintegration – and thus attainment of global chaos – occurs. Islands may, e.g., persist indefinitely or arise in certain $\beta$-windows. Such processes typically are highly case-specific.

Fluid inertia ($Re > 0$) breaks the internal flow symmetries (12) and (13) and, in consequence, time-reversal symmetry (21) of the map. This is immaterial for the existence of integrable limit (25) and the generic Hamiltonian response scenario in that non-zero $\beta$ still induces breakdown of the original island in a manner akin to that shown in Figures 10 and 11. Non-zero $Re$ manifests itself in a case-specific sense by affecting the particular disintegration process of a given configuration yet not the process perse. This is demonstrated in Fig. 12 by the Poincaré sections for $\beta = 11$ versus $Re$ for

FIG. 12. (a)–(e) Impact of fluid inertia demonstrated by Poincaré sections at $\beta = 11$ for positive (top) and negative (bottom) offset.
positive (top) and negative (bottom) offset. This reveals that fluid inertia may indeed significantly alter the behavior. The banana-shaped island for positive offset disappears around $Re \approx 7$ and eventually gives way to a small island emerging near the top right corner at $Re \approx 50$. The remnant of the original island of the integrable limit, in contrast, survives almost unscathed with increasing $Re$, save the notable excursion from the original symmetry axis (not shown). Fluid inertia has an only minor impact on the behavior for negative offset in the range $Re \lesssim 10$. Small islands emerge at the fringes of the chaotic core for $Re \approx 1$ yet the latter basically remains intact. The state from $Re \approx 20$ onwards again features a central island. However, rather than constituting a new entity emerging at some non-zero $\beta$, this island in fact is the remnant of the original island from the integrable limit (verified by monitoring the progression from $\beta = 0$). This implies that fluid inertia in fact promotes survival of the latter, or equivalently, retards its disintegration (compare with 3D effects).

F. Period doubling as universal route to chaos

Analysis of the 2.5D case revealed a fundamental difference between positive and negative offset regarding response to increasing $\beta$. Positive offset results in indefinite persistence of remnants of the original island in a manner as demonstrated in Fig. 10; negative offset ensures complete disintegration of this island by a so-called period-doubling bifurcation (i.e., split-up into two period-2 islands and their subsequent disintegration into chaos). The underlying mechanism in the 2.5D case is the interplay between the cell-wise reorientation induced specifically by negative offset and the time-reversal symmetry. The existence of essentially the same symmetry in the 3D non-inertial case, i.e., relation (19), implies also here period doubling for negative offset upon exceeding a threshold for $\beta$. This causes the disintegration of the original island (Fig. 11) at $\beta \approx 6.8$, as demonstrated in Fig. 13 by the states directly before and after the bifurcation. This clearly reveals the split-up of the inner tube/island into two adjacent tubes/islands and the transformation of the accompanying elliptic periodic line/period-1 point into a hyperbolic periodic line/period-1 point flanked by two elliptic periodic lines/period-2 points. The twist in the emerging pair of tubes reflects the period-2 nature in the Poincaré sections. The outer tube/island disintegrates and becomes part of the chaotic band delineated by the manifolds of the emerging hyperbolic periodic line/period-1 point (not shown).

The effect of three-dimensionality on the period doubling is entirely quantitative by changing the $\beta$-threshold at which this phenomenon occurs. The 2.5D flow undergoes bifurcation at $\beta \approx 5.5$ for the present RAM conditions; in the 3D case this slightly increases to $\beta \approx 6.8$, suggesting (at least in this parameter regime) a weak delay of the onset of chaos.

![ FIG. 13. Periodic-doubling bifurcation demonstrated for 3D non-inertial flow at negative offset: perspective view (top) and Poincaré section for $z'_0 = 0$ (bottom). (a) $\beta = 6.5$. (b) $\beta = 7$. (c) $\beta = 7.5$.](image_url)
Fluid inertia breaks symmetries and thus (in principle) paves the way to multiple routes to chaos. Consider to this end the generic conditions for transition of the period-1 elliptic point at the center of the global island of the integrable limit to a hyperbolic point. This is for simplicity carried out in canonical space; the monotonous relation between period time $T$ and kinematic parameter $\beta$ (Sec. IV B) enables direct translation of the findings below to physical space. Local linearization around a period-1 point $\xi_0 = \Phi_\xi(\xi_0)$ of the map $\xi_{k+1} = \Phi_\xi(\xi_k)$ associated with velocity $\omega = (\omega_1, \omega_2) = (\partial H/\partial \xi_2, -\partial H/\partial \xi_1)$ in the canonical reference frame $\xi = (\xi_1, \xi_2)$ yields

$$d\xi_1 = F \cdot d\xi_0, \quad F = \nabla_\xi \Phi|_{\xi_0}, \quad \lambda_{1,2} = \frac{tr(F)}{2} \left[ 1 \pm \sqrt{1 - \left( \frac{2}{tr(F)} \right)^2} \right], \quad (27)$$

in terms of local coordinates $d\xi = \xi - \xi_0$, with $F$ the deformation tensor and $\lambda = (\lambda_1, \lambda_2)$ its eigenvalue spectrum as a function of the trace $tr(F)$.\footnote{This incorporates property $\lambda_1 \lambda_2 = det(F) = 1$ due to incompressibility.} The type of the period-1 point is determined by the spectrum of $F$: hyperbolic with $\lambda = (\lambda, \lambda^{-1})$ for $|tr(F)| > 2$; elliptic with $\lambda = (\lambda, \lambda^*)$ for $-2 < tr(F) < 2$ (superscript $*$ indicates complex conjugate). Transition elliptic $\rightarrow$ hyperbolic occurs at $tr(F) = -2$ and $tr(F) = 2$, which involves two essentially different routes to chaos:

- **Route 1**: $tr(F) = 2 \Rightarrow \lambda_1 = \lambda_2 = 1$. This gives period-1 behavior near the period-1 point $d\xi_0 = 0$, i.e., $d\xi_1 = F \cdot d\xi_0 = d\xi_0$ for all $d\xi_0$, implying direct transition from a period-1 elliptic to a period-1 hyperbolic point. Here the island gradually disintegrates via before-mentioned Hamiltonian mechanisms into a chaotic sea.
- **Route 2**: $tr(F) = -2 \Rightarrow \lambda_1 = \lambda_2 = -1$. This gives period-2 behavior near the period-1 point $d\xi_0 = 0$, i.e., $d\xi_1 = F \cdot d\xi_0 = -d\xi_0$ and $d\xi_2 = F \cdot d\xi_1 = F^2 \cdot d\xi_0 = d\xi_0$ for all $d\xi_0$, implying transition to a period-1 hyperbolic point flanked by period-2 structures. This case constitutes a period-doubling bifurcation.

This exposes period doubling as one of the two ways by which disintegration of period-1 islands may occur. However, hereafter it is shown that Route 2 remains the sole breakdown mechanism in the generic 3D inertial case. To this end the transition criteria of Routes 1 and 2 are expressed in terms of the Lagrangian average

$$\omega^\dagger(\xi_0) = \frac{1}{T} \int_0^T \omega(\xi(\tau)) d\tau, \quad (28)$$

of the canonical velocity $\omega$ following the approach in Ref. 51. Reformulation of map $\Phi_\xi$ in terms of $\omega^\dagger$ gives

$$\Phi_\xi(\xi_0) = \xi_0 + T \omega^\dagger(\xi_0), \quad (29)$$

upon substitution into (27) yielding

$$F = I + TW, \quad W = \nabla_\xi \omega^\dagger|_{\xi_0} \Rightarrow tr(F) = 2 + T tr(W), \quad tr(W) = \nabla_\xi \cdot \omega^\dagger|_{\xi_0}, \quad (30)$$

with $W = \nabla_\xi \omega^\dagger|_{\xi_0}$ the strain-rate tensor of velocity field $\omega^\dagger$, putting forth

$$tr(F) = 2, \quad tr(F) = -2 \quad \Leftrightarrow \quad tr(W) = 0, \quad tr(W) = -4/T, \quad (31)$$

as equivalent transition criteria for Routes 1 and 2, respectively. Criteria (31) identify with the conditions for meso-ellipticity/hyperbolicity of generic material points in Ref. 51 upon substitution of $tr(W) = -T det(W)$. Note that, despite $\nabla_\xi \cdot \omega = 0$, generically $tr(W) \neq 0$. This property is key to the remainder of the argumentation.

By construction, period-1 points $\Phi_\xi(\xi_0) = \xi_0$ coincide with stagnation points $\omega^\dagger = 0$ of the Lagrangian-averaged velocity. Their type is, in a similar way as periodic points, determined by the eigenvalues $\mu_{1,2} = 1/2[tr(W) \pm \sqrt{tr(W)[tr(W) + 4/T]}]$ of $W$. Relevant for the present discussion is that $\mu_1 + \mu_2 = tr(W)$ distinguishes attracting ($tr(W) < 0$) and repelling ($tr(W) > 0$) stagnation points, where former and latter act as fluid sinks and sources, respectively. The fact that the 2.5D case and the 3D non-inertial case admit disintegration of the original island only by period doubling renders, by virtue of criteria (31), the associated stagnation point $\omega^\dagger = 0$ invariably of the attracting
kind \( tr(W) = -4/T < 0 \). Below it is demonstrated that said stagnation point stays attracting – and period doubling remains the sole breakdown mechanism – in the inertial case.

The integrable limit (Sec. IV E) corresponds with \( T = 0 \) and via \( \Phi_\epsilon|_{T=0} = I \) yields \( tr(F) = 2 \) and thus \( tr(W) = 0 \). Occurrence of \( tr(W) \neq 0 \) for non-zero \( T > 0 \) relies on the net divergence of the system, given by

\[
D_Q = \int_Q \nabla \cdot \omega^s \, da_Q = \oint_{\partial Q} \omega^s \cdot n_Q \, ds_Q, \tag{32}
\]

where \( Q \) is the canonical space (area element \( da_Q \)), with boundary \( \partial Q \) (line element \( ds_Q \)) and outward normal \( n_Q \). (Boundary integral follows from Stokes’ theorem.) Cases \( D_Q < 0 \) and \( D_Q > 0 \) signify net influx and outflux across \( \partial Q \), respectively, which can be recast as \( s_Q < 0 \) and \( s_Q > 0 \), with \( s_Q = \text{sign}[D_Q] \). Property (32) translates to

\[
D_A = \int_A \nabla \cdot u^s \, da_A = \oint_{\partial A} u^s \cdot n_A \, ds_A, \tag{33}
\]

in the physical counterpart to \( Q \), viz., cross-section \( A_\epsilon \) (Sec. IV C), with here \( s_A < 0 \) and \( s_A > 0 \) distinguishing net influx and outflux, respectively. Topological equivalence between physical and canonical space implies

\[
s_Q = s_A = s, \tag{34}
\]

which, upon assuming in a first approach that the net throughflow region occupies the entire domain, results in

\[
s = \text{sign} \left( \int u^s_\epsilon (r_0, \theta_0) |_{r_\epsilon = 1} \, d\theta \right). \tag{35}
\]

Property (35) directly relates to the repelling/attracting properties of stagnation points in the Lagrangian-averaged field \( u^s \) and, inherently, to those of the stagnation points in its canonical counterpart \( \omega^s \). Topological equivalence namely dictates that each stagnation point of \( u^s \) has an equivalent in \( \omega^s \) with identical stability properties. The relation between \( s \) and \( u^s \) enables determination of the properties of stagnation points of \( u^s \) (and thus of \( \omega^s \)) by exploiting the fact that property \( s \) is subject to certain bounds. This is elaborated below.

Fluid parcels released at a minute radial distance \( \epsilon \ll 1 \) from solid boundary \( \partial A_\epsilon \), i.e., \( r_0 = 1 - \epsilon \), can exhibit a net radial displacement only in negative-\( r \) direction for initial conditions approaching \( \partial A_\epsilon \) infinitesimally close: \( \lim_{k \to 0} r^s_\epsilon \leq r_0 \). This directly sets the upper bound \( \lim_{k \to 0} u^s_\epsilon (1 - \epsilon, \theta_0) \leq 0 \) for the corresponding Lagrangian-averaged radial velocity and, in consequence, imposes restriction \( s \leq 0 \) on property (35). This range can be further demarcated by realizing that fluid parcels released infinitesimally close to a window propagate parallel to the latter until undergoing an inward deflection upon reaching its edge due to the circulatory transverse fluid motion. Hence, such fluid parcels spend at least part of their excursion in the domain interior, leading to \( \lim_{k \to 0} r^s_\epsilon \leq r_0 \) and thus \( \lim_{k \to 0} u^s_\epsilon (1 - \epsilon, \theta_0) < 0 \) in those cases. Thus fluid parcels starting \( \epsilon \)-close to stationary and moving boundary segments exhibit \( u^s_\epsilon = 0 \) and \( u^s_\epsilon < 0 \), respectively, which invariably gives rise to a net influx \( (s < 0) \). Fig. 14(a) demonstrates this by \( u^s_\epsilon \) on circle \( r_0 = 0.98 \) for the 2.5D case with uniform axial velocity \( u_\epsilon = 1 \) at positive offset and \( \beta = 1 \). Here the shaded regions correspond with the windows. (Note the minuscule overshoot \( u^s_\epsilon > 0 \) near the window edges stems from the large local velocity gradients and vanishes for \( r_0 \) approaching unity.) This results in streamlines emanating from influx boundary segments \( (u^s_\epsilon > 0) \) that, given absence of outflux segments \( (u^s_\epsilon < 0) \), must end in interior stagnation points of attracting type \( (tr(W) < 0) \). The stagnation point associated with the original island is the only such point – and thereby automatically of attracting type – in at least some finite window \( 0 \leq T \leq T_{\text{end}} \). Fig. 14(b) demonstrates the typical streamline portrait by the aforementioned 2.5D example. Restriction to \( tr(W) < 0 \) has the fundamental consequence that (in regime \( 0 \leq T \leq T_{\text{end}} \)) island breakdown can for any RAM occur only through period doubling.

Emergence of new stagnation points in \( u^s \) (period-1 points in map \( \Phi \)) at some \( T_{\text{end}} \) prior to said breakdown in principle admits transition of the stagnation point associated with the original island to repelling type \( tr(W) > 0 \), implying disintegration via Route 1 instead of Route 2. However,
FIG. 14. Lagrangian-averaged field $u^\dagger$ for 2.5D case with uniform axial velocity (positive offset and $\beta = 1$): (a) radial component $u_r^\dagger$ on circle $r_0 = 0.98$; (b) streamline pattern. Ring of dots: initial positions; central dot: stagnation point of original island.

Continuous transition from the attracting state ($tr(W) < 0$) for $T < T_{solo}$ in Fig. 14(b) to a repelling state ($tr(W) > 0$) for $T > T_{solo}$ and simultaneous satisfaction of condition $s \leq 0$ can occur only through the specific sequence of events sketched in Fig. 15:

1. Formation of a global island ($tr(W) = 0$) at $T = T_{solo}$.
2. Emergence of an attracting cycle (dashed) at $T > T_{solo}$ that encloses the (now) repelling point ($tr(W) > 0$) and replaces the former attracting point as attractor for streamlines emanating from the wall.

Note that this scenario concerns a supercritical Hopf bifurcation of the streamline pattern of the Lagrangian-averaged velocity.41

Scenarios other than the Hopf bifurcation inevitably involve discontinuous switching between streamline patterns of $u^\dagger$ and/or outflux boundary segments ($u_r^\dagger > 0$) and thus can immediately be dismissed. However, though less evident, the Hopf bifurcation also conflicts with a realistic scenario, namely, in the following two ways. First, it rules out appearance of new stagnation points for $u^\dagger$ at $T = T_{solo}$, while the current range of interest ($T \geq T_{solo}$) specifically pertains to the case of multiple stagnation points. Second, the global island at $tr(W) = 0$ in Fig. 15(b) exists only in the integrable state $T = 0$ and identifies with the isopleths of Hamiltonian function $H^*$ following (25). However, bifurcation directly at $T = 0$ is prohibited by the fact that elliptic islands of Hamiltonian systems generically survive for finite ranges of the perturbation parameter.41 Thus transition of the stagnation point associated with the original island to repelling type via the scenario in Fig. 15 at some non-zero $T$ cannot occur, retaining period doubling via Route 2 as sole breakdown mechanism for said island in the inertial case ($Re > 0$).

The above findings generically extend to cases with internal isolated regions. First, the latter usually are very localized, causing the bulk of the net throughflow region to remain bounded by

FIG. 15. Bifurcation of stagnation point associated with original island (dot) from attracting type ($tr(W < 0)$) to repelling type ($tr(W > 0)$). The dashed circle is an attracting limit cycle; curves are typical streamlines; arrows indicate flow direction. (a) $tr(W) < 0$. (b) $tr(W) = 0$. (c) $tr(W) > 0$. 
the cylindrical wall. Second, internal regions are strictly separated by transport barriers, which act similarly on these regions as the non-moving boundary segments. Thus the essential net-influx condition \( s < 0 \) that restricts the breakdown scenario to period doubling normally is upheld.

Fig. 16 demonstrates the persistence of period doubling as breakdown mechanism of the original island under inertial conditions \((Re = 10)\) for negative offset. Here it occurs around \( \beta \approx 6 \). The phenomenon remains upon increasing \( Re \) yet typically occurs at higher \( \beta \); the threshold, e.g., increases to \( \beta \approx 11 \) for \( Re = 20 \) (Fig. 12). Thus fluid inertia, comparable to 3D effects in the period-doubling bifurcation of the non-inertial limit, delays the disintegration. These observations are representative for any island disintegration investigated in the regime \( 0 \leq Re \leq 50 \). An essential difference with the non-inertial limit \( Re = 0 \) is that this must no longer be restricted to negative offset. Further demarcation of the specific bifurcation regimes for \( Re > 0 \) is beyond the present scope, though.

V. CONCLUSIONS

The present study concerns the Lagrangian transport properties of inline mixers with flow domains with cylindrical geometries. Key subjects of investigation are the impact of three-dimensionality (3D) and fluid inertia on the dynamics. To this end the Rotated Arc Mixer (RAM) is adopted as representative system. The Lagrangian transport properties are investigated in terms of coherent structures that form in the 3D streamline portrait. The corresponding 2.5D simplification of the RAM (i.e., with cell-wise fully-developed flow) serves as reference for the behavior of the 3D case.

Three-dimensionality emanates from the formation of transition zones between consecutive mixing cells that enable continuous change-over between the cell-wise developed flow. These zones expand with stronger transverse forcing and at some point may even occupy the entire mixing cells. Fluid inertia tends to amplify transition effects. 3D effects may furthermore result in backflow, i.e., local upstream flow, which sets in beyond a certain minimum transverse forcing and intensifies upon the latter becoming stronger. However, backflow generically does not signify net transport from outlet to inlet; it occurs on backflow segments of streamlines running from inlet to outlet (i.e., with net downstream flux). Fluid inertia typically suppresses backflow and thus promotes uni-directional downstream flow.

The 3D flow field always accommodates a global symmetry in that the internal flow fields of consecutive cells relate via a simple reorientation. This admits decomposition of the total flow field into a sequence of reorientations of the flow field in the first cell (“base flow”) in the same way as in the 2.5D case. Essential difference between 2.5D and 3D base flow is that the latter depends on all spatial coordinates due to the continuous change-over between cells. The 3D non-inertial limit furthermore possesses internal symmetries within each mixing cell, which in essence are 3D counterparts to internal symmetries in the 2.5D simplification.

Topological considerations strongly suggest that there always exists a net throughflow region between inlet and outlet of the periodic flow domain that is strictly separated from possible internal regions. The Lagrangian dynamics in this region admits representation by a 2D time-periodic Hamiltonian system – and associated map – via proper coordinate transformations. The latter may
be technically challenging (particularly in case of backflow) yet in principle is always possible. This establishes one fundamental kinematic structure for the present class of inline mixers under realistic operating conditions. Thus Lagrangian transport (in the net throughflow region) remains in all cases, i.e., including 3D effects and fluid inertia, governed by Hamiltonian mechanisms.

The common kinematic structure results in universal behavior. All possible RAM configurations converge on one integrable state, consisting of one family of stream tubes centered on the cylinder axis, for vanishing transverse forcing. All actual states follow from Hamiltonian breakdown of this integrable state induced by continuously increasing transverse forcing from zero to its current level. This reveals that a so-called period-doubling bifurcation is the only way to disintegrate the original tube family. Period doubling thus is a necessary (yet not sufficient) condition for the attainment of global chaos. This kinematic scenario includes cases with fluid inertia. Principal difference between systems with and without fluid inertia is that the latter admit period doubling only for negative offset, which is a direct consequence of the internal symmetries in the non-inertial limit. System parameters other than that of the transverse forcing have an only quantitative impact on the response behavior yet not on the scenario perse.

Important in a practical context is that the widely-used 2.5D approach retains these fundamental kinematic properties and deviates from the generic 3D inertial case essentially only in a quantitative sense. This substantiates its suitability for (at least first exploratory) studies on (qualitative) mixing properties.

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APPENDIX A: GENERIC PROPERTIES RAM FLOW

1. Net flow forcing

The axial flow is governed by the force balance for a fluid parcel, given by

$$Re \frac{du_z}{dt} = Re u \cdot \nabla u_z = f_z, \quad f_z = f_p + f_v, \quad f_p = -\frac{\partial p}{\partial z}, \quad f_v = \nabla^2 u_z, \quad (A1)$$

with $f_z$ the axial body force due to pressure ($f_p$) and viscosity ($f_v$). The corresponding global force balance reads

$$Re \int_D u \cdot \nabla u_z r dr d\theta dz = F_p + F_v = 0, \quad (A2)$$

with $F_p = \int_D f_p r dr d\theta dz = \pi C N A$ and $F_v = \int_D f_v r dr d\theta dz = \int_{\partial D} \partial u_z / \partial r |_{r=1} d\theta dz$ using Gauss' divergence theorem to attain the boundary integral. The net pressure force stems entirely from the pressure drop between inlet and outlet: $F_p = \int_0^1 \int_0^{2\pi} (p(r, \theta, 0) - p(r, \theta, N A)) r dr d\theta = \pi C N A$. The inertial term vanishes identically for any $Re$ due to $\int_D u \cdot \nabla u_z r dr d\theta dz = \int_D \nabla \cdot (u \cdot u)_r d r d \theta d z = \int_{\partial D} u \cdot e_r |_{r=1} u |_{r=1} d \theta d z = 0$, which follows from Gauss’ theorem in conjunction with $\nabla \cdot u = 0$ and $u \cdot e_r |_{r=1} = u_r |_{r=1} = 0$. Relation (A2) implies that the net pressure force $F_p$ is for any $Re$ balanced by viscous wall stress $F_v$. The latter is merely a reaction force, due to absence of $z$-wise wall motion ($u_z |_{r=1} = 0$), thus advancing $F_p$ as the sole driving force of the fluid. The global balance between pressure and viscous forces holds individually for each fluid parcel in the Stokes limit $Re = 0$. The key role of the pressure emerges in an alternative way by recasting the force balance as a Poisson equation for $u_z$, i.e., $\nabla^2 u_z = f$, with nonlinear source term $f(u) = \partial p / \partial z + Re u \cdot \nabla u_z$, and homogeneous Dirichlet conditions $u_z |_{r=1} = 0$. The latter implies that non-zero $u_z$ happens exclusively for non-zero source $f$. Moreover, net axial motion happens only for non-conservative contributions to the source, which here ensue entirely from the pressure gradient: $F = \int_D f r dr d\theta dz = -F_p = -\pi C N A$. 

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2. Net axial flux

Net pressure force \( F_p > 0 \) implies a net axial flux \( \Phi = \int_0^1 \int_0^{2\pi} u_z(r, \theta, z)rdrd\theta > 0 \) that, by virtue of steady conditions, is equal for all cross-sections \( 0 \leq z \leq N\Lambda \). This has fundamental implications for the Fourier decomposition of \( u \). Substitution of (7) yields

\[
\Phi = \int_0^1 \int_0^{2\pi} \tilde{u}_{z,0}(r, \theta)rdrd\theta + \sum_{\forall k \neq 0} \left\{ \int_0^1 \int_0^{2\pi} \tilde{u}_{z,k}(r, \theta)rdrd\theta \right\} e^{2\pi ikz/N\Lambda},
\]

revealing that \( z \)-independent \( \Phi \) can be met only if the trailing term in (A3) vanishes identically. This results in conditions (8) for the Fourier modes of the axial velocity component.

3. Axial flow in the non-inertial limit

The flow decomposes in a pressure-driven and rotor-driven component according to (9) for \( Re = 0 \). Fourier decomposition further exposes the underlying flow structure on substitution of (7) in the \( z \)-component of the Stokes equations,

\[
-C + \sum_k \tilde{p}_k(r, \theta) \frac{2\pi k}{N\Lambda} e^{2\pi ikz/N\Lambda} = \sum_k \left\{ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \tilde{u}_{z,k}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \tilde{u}_{z,k}}{\partial \theta^2} - \left( \frac{2\pi k}{N\Lambda} \right)^2 \tilde{u}_{z,k} \right\} e^{2\pi ikz/N\Lambda},
\]

with mode-wise boundary conditions \( \tilde{u}_{z,k}(1, \theta) = 0 \). The balance must hold individually for each mode \( k \), resulting in

\[
-C = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \tilde{u}_{z,0}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \tilde{u}_{z,0}}{\partial \theta^2} \quad \Rightarrow \quad -C = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \tilde{u}_{z,0}}{\partial r} \right),
\]

for \( k = 0 \). The \( \theta \)-component vanishes due to the homogeneous Dirichlet condition \( \tilde{u}_{z,0}(1, \theta) = 0 \). This implies \( \theta \)-independence and thus reduces this mode of the momentum balance to that for a standard Poiseuille flow in a pipe: \( \tilde{u}_{z,0}(r) = u_p(r) = 2U(1 - r^2) \), with mean flow \( U = C/8 \). Hence, the axial flow is composed as

\[
u_{z}(r, \theta, z) = u_p(r) + \sum_{\forall k \neq 0} \tilde{u}_{z,k}(r, \theta) e^{2\pi ikz/N\Lambda},
\]

with \( \tilde{u}_{z,k} \) the superimposed modes having net zero flux: \( \langle u_z \rangle = \langle u_p \rangle = \Phi/\pi \). Reconciliation with (9) reveals that the \( z \)-components of \( u_p \) and \( u_{z,0} \) correspond with Fourier modes \( k = 0 \) and \( k \neq 0 \) of \( u_z \), respectively.

4. Axial flow in the presence of fluid inertia

Fourier mode \( \tilde{u}_{z,0} \) is no longer governed by (A5) for \( Re > 0 \) yet admits decomposition as \( \tilde{u}_{z,0}(r, \theta) = u_p(r) + 
\tilde{u}'_{z,0}(r, \theta) \), with \( \tilde{u}'_{z,0} \) the departure from the Poiseuille flow, without loss of generality. (Component \( \tilde{u}_{z,0} \) simply follows from subtraction of \( u_p \) from Fourier mode \( k = 0 \) of the net axial velocity \( u_z \)). Property (8) implies \( \tilde{u}_{z,0}(1, \theta) = 0 \) by virtue of \( \langle \tilde{u}_{z,0} \rangle = \langle u_p \rangle + \langle \tilde{u}'_{z,0} \rangle = \Phi/\pi \) and \( \langle u_p \rangle = \Phi/\pi \). Thus the axial flow maintains the generic structure of a Poiseuille flow superimposed by Fourier modes with net zero flux, i.e.,

\[
u_{z}(r, \theta, z) = \tilde{u}_{z,0}(r, \theta) + \sum_{\forall k \neq 0} \tilde{u}_{z,k}(r, \theta) e^{2\pi ikz/N\Lambda} = u_p(r) + \sum_{\forall k} \tilde{u}_{z,k}(r, \theta) e^{2\pi ikz/N\Lambda},
\]

with \( \langle u_z \rangle = \langle u_p \rangle = \Phi/\pi \). Principal difference between (A6) and (A7) is the additional mode \( k = 0 \) in the trailing term (the prime for \( k = 0 \) to distinguish departure from the Poiseuille flow has been dropped). This is due to the coupling of Fourier modes by the nonlinear term. Hence, Fourier modes \( \tilde{u}_{z,k} \) in (A6) and (A7) are different.
APPENDIX B: NET AXIAL MOTION OF INDIVIDUAL FLUID PARCELS IN THE NET THROUGHFLOW REGION

Streamlines are formal solutions $x(t) = \int_0^t x(x(\xi))d\xi$ to kinematic equation (3). The axial component reads
\[ z(t) = \int_0^t u_z(x(\xi))d\xi = \int_0^t u_p(r(\xi))d\xi + \sum_{\forall \xi} \int_0^t \tilde{u}_{z,k}(r(\xi), \theta(\xi))e^{2\pi i z(\xi)/N\Lambda}d\xi, \]
using the generic form (A7) (which holds for $Re \geq 0$). Net backflow from outlet to inlet can in general not be rigorously ruled out due to the (at least hypothetical) possibility of a fluid parcel being confined to a flow region or individual streamline with $\tilde{u}_{z,k}$ such that, despite $u_p > 0$ everywhere outside the cylinder wall, a net axial velocity $\tilde{u}_z(x_0) = \lim_{t \to \infty} t^{-1} \int_0^t u_z(x(\xi))d\xi < 0$ occurs. However, for the net throughput region this can be precluded:

- Fluid parcels on periodic streamlines: $x(0) = (r_0, \theta_0, 0) \to x(t) = (r_0, \theta_0, P\Lambda N\Lambda)$. Here pressure field (7) exerts a net downstream force on fluid parcels during each cycle via net pressure drop $\Delta p = p(r_0, \theta_0, P\Lambda N\Lambda) - p(r_0, \theta_0, 0) = -C P\Lambda N\Lambda < 0$. Hence, fluid parcels that can travel only from inlet to outlet yet not vice versa.
- Fluid parcels on tubes of elliptic periodic lines. The corresponding streamlines densely wind themselves about the tubes and after a given number of cycles, say $K$, closely approximate their initial cross-sectional position. This yields, similar to the periodic lines, a net pressure drop $\Delta p \approx p(r_0, \theta_0, K\Lambda) - p(r_0, \theta_0, 0) = -CK\Lambda N\Lambda < 0$.
- Fluid parcels in chaotic zones of hyperbolic periodic lines. Here the streamlines, similar to the elliptic tubes, also closely approximate their initial cross-sectional position at some point in time by virtue of the Poincaré recurrence theorem. This again gives a net axial pressure drop $\Delta p < 0$.

Efficient global mixing, the targeted state in inline mixers, in fact promotes a global net throughput zone with, on average, a uniform net axial velocity. Global mixing naturally causes fluid parcels to eventually sample the entire cross-section, meaning in the long run they experience averaged Fourier modes. Through properties (8) this yields
\[ \lim_{t \to \infty} z(t) \approx \lim_{t \to \infty} \left[ \int_0^t (\tilde{u}_{z,0})d\xi + \sum_{\forall \xi} \int_0^t (\tilde{u}_{z,k})e^{2\pi i z(\xi)/N\Lambda}d\xi \right] = \lim_{t \to \infty} \frac{\Phi_1}{\pi} > 0, \] implying net downstream axial motion of all fluid parcels.

Alternative integrable limits can be based on vanishing of one of the parameters $\beta$.

