Transport and mixing of scalar quantities in fluid flows is ubiquitous in industry and Nature. Turbulent flows promote efficient transport and mixing by their inherent randomness. Laminar flows lack such a natural mixing mechanism and efficient transport is far more challenging. However, laminar flow is essential to many problems and insight into its transport characteristics is of great importance. Laminar transport, arguably, is best described by the Lagrangian fluid motion (“advection”) and the geometry, topology and coherence of fluid trajectories. Efficient laminar transport being equivalent to “chaotic advection” is a key finding of this approach.

The Lagrangian framework enables systematic analysis and design of laminar flows. However, the gap between scientific insights into Lagrangian transport and technological applications is formidable primarily for two reasons. First, many studies concern two-dimensional (2D) flows yet the real world is three dimensional (3D). Second, Lagrangian transport is typically investigated for idealised flows yet practical relevance requires studies on realistic 3D flows.

The present review aims to stimulate further development and utilisation of know-how on 3D Lagrangian transport and its dissemination to practice. To this end 3D practical flows are categorised into canonical problems. First, to expose the diversity of Lagrangian transport and create awareness of its broad relevance. Second, to enable knowledge transfer both within and between scientific disciplines. Third, to reconcile practical flows with fundamentals on Lagrangian transport and chaotic advection. This may be a first incentive to structurally integrate the “Lagrangian mindset” into the analysis and design of 3D practical flows.

1 Introduction

The scope of this review is transport and mixing of scalar quantities such as additives, chemical species, heat and nutrients in realistic three-dimensional (3D) fluid flows under laminar flow conditions. This flow regime sets in for a Reynolds number $Re = UL/\nu$ below the threshold of turbulence and is common to many systems and processes in industry and Nature due to high fluid viscosities $\nu$, small length scales $L$ and/or low velocities $U$. Industrial examples are found abundantly in fluids processing and span a wide range of scales from conventional food or polymer processing [1, 2, 3, 4, 5] to emerging technologies as e.g. process intensification and micro-fluidics [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19]. Further technological applications include (at first glance) less obvious systems as Darcy representations of flow and transport in porous media, relevant e.g. for in situ mining, enhanced oil recovery, geothermal energy extraction or groundwater remediation [20, 21, 22, 23, 24, 25], as well as continuum descriptions of granular flows [26, 27, 28].

Scalar transport in laminar flows is also key to many systems beyond industry and technology. Consider, for example, transport of oxygen or pathogens in physiological flows [29, 30, 31, 32, 33, 34] and geophysical flow problems as plate tectonics driven by mantle convection [35] and dispersion of nutrients or spreading of pollutants in oceans [36, 37, 38, 39].

Scalar transport in flows generically involves an interplay of the physical motion of the fluid (“advection”) with

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1Oceanographic flows typically are turbulent yet often admit approximation by Euler flows or spatio-temporal averaged flows obtained via e.g. RANS or LES. Such approximations are deterministic in the sense of being robustly reproducible and thus effectively behave as laminar flows. The underlying turbulent flows, on the other hand, are stochastic in being realisations from an ensemble of states and thus are inherently unpredictable.
molecular mechanisms as diffusion and chemical reactivity. Advection is, given its pivotal role in scalar transport, the focus of this review and for laminar flow conditions, arguably, best viewed from the Lagrangian perspective of the motion of individual fluid parcels. The stochastic nature of turbulent flows gives rise to Brownian motion of fluid parcels that typically admits expression as an effective diffusion (“turbulent diffusion”) [40]. This is absent in laminar flows and, in consequence, the structure of Lagrangian fluid trajectories is essential to the transport characteristics. Moreover, the connection between Eulerian velocity field and Lagrangian motion may be non-trivial and counter-intuitive in that extremely tangled fluid trajectories can coexist with very simple velocity fields. Hence transport studies in laminar flows should on account of these fundamental characteristics concentrate on the Lagrangian transport of fluid parcels.

The most striking disconnect between flow and Lagrangian transport is the intriguing phenomenon of “chaotic advection” that, in a “rough–and–ready” definition, concerns the rapid deformation of material fluid elements into highly ramified and filamented structures. This promotes efficient transport reminiscent of the rapid dispersion by stochastic fluctuations in turbulent flows and thus renders chaotic advection the laminar counterpart to turbulent mixing. Chaotic advection has first been demonstrated in the seminal study of Hassan Aref in the early 1980s [41] and can occur for very general (and often deceptively simple) flows [42, 43].

Consider as an illustration of the versatility of Lagrangian transport phenomena a few examples in completely different settings. Fig. 1(a) gives 3D trajectories (visualised by fluorescent tracer particles) in the steady flow inside a cylindrical container stirred by an impeller [44]. The system serves as laboratory model for industrial batch mixers and the Lagrangian transport – in particular the accomplishment of global 3D chaotic advection – evidently is crucial to the functionality and performance of such devices. The disordered nature of the trajectories suggests chaos yet dedicated Lagrangian analysis is necessary to conclusively establish this. The web of trajectories, by virtue of continuity and mass conservation, namely contains hidden “pathways” and coherent structures that geometrically guide the transport. Relevant here are in particular toroidal (i.e. donut-shaped) material surfaces that may emerge around the impeller axis, which, if indeed occurring, act as barriers to chaotic trajectories and thus compromise the mixing performance. Such structures cannot be inferred from the velocity field nor are directly visible, meaning they elude conventional analyses and engineering. Hence many mixer designs and operating conditions likely are sub-optimal.

A non-technological example is found in physiology. Fig. 1(b) gives the 3D Lagrangian trajectories inside a human aortic aneurysm (balloon-like malformations of blood vessels) visualised by in vivo measurements using phase-contrast cardiovascular magnetic-resonance imaging [45]. This reveals vortical structures (indicated by open arrows) akin to the batch mixer in Fig. 1(a) driven by the main bloodstream (closed arrows) and thus suggests comparable (chaotic) dynamics due to a similar “hidden organisation” of the fluid trajectories into chaotic tangles and toroidal barriers. However, unlike mixers, chaotic advection has a potentially adverse impact by tending to entrap blood-borne platelets and substances implicated in thrombosis and atherosclerosis [29]. Hence Lagrangian transport is a key factor in these vascular diseases and investigation of the pathways and coherent structures embedded in the fluid trajectories may shed more light on their origin and causes.

A biological counterpart exists in the cilia-driven flow at polyps in coral reefs visualised (for polyp explants) by fluorescent particles in Fig. 1(c) [46, 47, 48]. The “stirring” by the cilia aims, similar to the batch mixers, at creating favourable transport conditions near the coral surface for the benefit of photosynthesis and suppression of adverse effects as carbon fixation and pathogen invasion. However, what conditions are in fact “favourable” (this must not necessarily be chaos; transport barriers as mentioned before may very well benefit these processes) and how Nature accomplishes this remains unclear. Lagrangian transport analyses may again contribute to the understanding of these phenomena.

The examples in Fig. 1 give a first flavour of the richness and universality of Lagrangian transport and demonstrate that seemingly different systems have much more in common than meets the eye. The fluid trajectories are namely organised into elementary “building blocks” that form the network of transport pathways and barriers according to general “rules” depending only secondarily on the particulars of the flow (forcing). This implies unifying structures that transcend specific flow configurations (as well as scientific and engineering disciplines) and thus enable a universal strategy for analysis of Lagrangian transport by the geometry, topology and coherence of fluid trajectories.

The relevance and usefulness of a (cross-disciplinary) unified Lagrangian approach towards laminar transport is evident from the above. Moreover, efficient laminar transport being synonymous to chaotic advection is in itself recognised in the fluid-dynamics community [43]. However, the gap between scientific research and insights into (chaotic) Lagrangian transport and technological applications is still formidable primarily for two reasons. First, scientific studies on Lagrangian transport to date mainly concern two-dimensional (2D) flows. The real world is three dimensional (3D), on the other hand, yet 3D Lagrangian transport remains elusive. The additional spatial degree of freedom greatly increases the dynamical richness and geometric complexity and a comprehensive theoretical and conceptual framework, in contrast with the basically complete picture of 2D Lagrangian transport, is therefore still non-existent in 3D [49, 43]. Second, Lagrangian transport is typically investigated via theoretical and computational studies and often concerns idealised flow situations that are difficult (or even impossible) to create in laboratory experiments. Dedicated experimental studies on Lagrangian transport, crucial for (conclusively) establishing physical meaningfulness and practical relevance of results, remain scarce, though. The only systematic technological application to date exists in a subclass of industrial and micro-fluidic mixers that admit reconciliation with 2D flows and the associated theory.
However, dissemination even of the proven principle of chaotic advection for such devices to the practicing engineering community remains a challenge.

The growing importance and urgency to close the gap between fundamentals and applications motivates this review. Its principal aim is to stimulate further utilisation and development of know-how on 3D Lagrangian transport for technological applications and its dissemination to practitioners in industry and beyond. To this end 3D practical flows to which (non-)chaotic Lagrangian transport is essential are categorised into canonical problems so as to (i) identify analogies and similarities, (ii) establish connections with configurations in fundamental 3D Lagrangian studies, (iii) outline a unified Lagrangian framework for 3D transport studies and (iv) isolate challenges specifically regarding applications. The focus will be primarily on flows and applications in industry and technology with excursions into life sciences and on occasion beyond. Furthermore, the canonical flow problems will be exemplified and represented by experimentally-realisable cases to ensure practical relevance and robustness of phenomena.

The Lagrangian framework and the concept of chaotic advection have, expanding on Aref’s pioneering work \[41\], been developed in the 1980s and 1990s mainly for 2D time-periodic flows (as approximations for inline industrial mixers) using dynamical-systems methods as Hamiltonian mechanics and vector-field topology \[51\]–\[57\]. Two important extensions of the Lagrangian framework in the early 2000s include granular-media flows that admit a continuum description \[26\]–\[27\] and deterministic descriptions of large-scale oceanographic flows \[36\]–\[37\]. Recent and ongoing efforts focus on generalisations of the Lagrangian approach to aperiodic and finite-time flows \[58\]–\[61\], and the interaction between chaotic advection and other transport physics (e.g. diffusion, reaction, propulsion, particle inertia) \[60\]–\[63\]–\[61\].

The present review expands on the above body of work by explicitly positioning this in the context of 3D practical flows and is organised as follows. \textsection{}3 introduces the general concepts and methods of Lagrangian transport studies.

\textsection{}2 categorises 3D practical flows into canonical problems in order to, first, identify analogies and similarities and, second, establish connections with fundamental 3D Lagrangian studies. \textsection{}4 outlines a unified Lagrangian framework for 3D transport studies by reconciling the flow categories and canonical problems with theory and fundamentals on Lagrangian transport. \textsection{}5 investigates the degree of dissemination and technological utilisation of the concept of chaotic advection via a survey on relevant patents and commercial applications. Concluding remarks including an overview of challenges exposed by this review are in \textsection{}6.

2 Lagrangian approach to transport and mixing

In the absence of chemical reactions, transport of a scalar quantity \(C\) in fluid flows occurs by an interplay of two physical mechanisms: advection by the fluid motion and molecular diffusion along concentration gradients (Fig. 2). The corresponding Eulerian evolution is governed by the advection-diffusion equation

\[
\frac{dC}{dt} = \frac{∂C}{∂t} + \bar{u} \cdot \nabla C = \frac{1}{Pe} \nabla^2 C, \quad C(\bar{x},0) = C_0(\bar{x}), \tag{1}
\]

with \(\bar{u}\) the velocity field and \(Pe = UL/\alpha\) the well-known Pécel number. Here \(U\) and \(L\) are characteristic velocity and length scales, respectively, and \(\alpha\) is the scalar diffusivity. The present review concerns systems with \(Pe \gg 1\), which holds for many practical systems, meaning that scalar transport is dominated by advection and \((1)\) effectively reduces to \(dC/dt = 0\). Thus scalar \(C\) is passively advected by fluid parcels, i.e. \(C(\bar{x}(t)) = C(\bar{x}_0)\), with \(\bar{x}(t)\) the current parcel position governed by the kinematic equation

\[
\frac{d\bar{x}}{dt} = \bar{u}(\bar{x}(t),t) \quad \Rightarrow \quad \bar{x}(t) = \bar{Φ}_t(\bar{x}_0), \tag{2}
\]

\(^{2}\)Advection and diffusion of heat is usually denoted “convection” and “conduction”, respectively.
Fig. 2. Transport of scalar quantity $C$ (red) in fluid flows by interplay of advection of material region (left) and diffusion across its interface (right). Arrows indicate transport by respective mechanisms.

and flow $\mathbf{\Phi}$, its formal solution describing the Lagrangian motion of a parcel released at $\mathbf{x}(0) = \mathbf{x}_0$. Hence, $dC/dt = 0$ and $d\mathbf{x}/dt = \mathbf{u}$ are the equivalent Eulerian and Lagrangian representations of scalar advection. The fluid is assumed incompressible, implying solenoidal flow ($\nabla \cdot \mathbf{u} = 0$), which has fundamental ramifications for Lagrangian transport.

The exposition hereafter adopts the Lagrangian perspective on scalar transport with kinematic equation (2) as its cornerstone. The latter defines a generically nonlinear dynamical system and enables investigation of advective transport by the geometry and topology of the Lagrangian fluid trajectories, i.e. the “Lagrangian flow topology”, which is composed of elementary structures denoted “Lagrangian coherent structures” (LCSs) [52,59,43]. The nature of LCSs is described and illustrated by the time-periodic flow

$$\mathbf{u}(\mathbf{x}, t + pT) = \mathbf{u}(\mathbf{x}, t),$$

with $T$ the period time and $p$ the period, in a 2D cavity driven by the alternate steady motion of the left and right walls. Fig. 3 gives the translation direction of each wall and the corresponding velocity vectors and streamline patterns obtained by numerical solution of the non-dimensional Navier-Stokes equations. Note that the steady flow during the second half of each period is a reorientation of that of the first half. This composition of time-periodic flows from reorientations of a steady base flow is common practice both in scientific studies on mixing and industrial applications.

The scalar advection illustrated in Fig. 2(a) may take place in two fundamentally different ways: non-chaotic advection versus chaotic advection. This is demonstrated in Fig. 4 for the advection of blue and red material elements in the 2D lid-driven cavity, revealing a dramatic difference in behaviour. The blue element undergoes a moderate shear-like deformation that yields (at most) linear stretching of the interface. This signifies non-chaotic advection. The red element, on the other hand, exhibits strong deformation due to repeated stretching and folding, resulting in exponential elongation of the interface. This is the hallmark of chaotic advection [52].

Fig. 3. Time-periodic 2D lid-driven cavity flow $\mathbf{u}(\mathbf{x}, t + pT) = \mathbf{u}(\mathbf{x}, t)$ due to alternate steady translation of left and right wall during first ($0 \leq t' \leq T/2$) and second ($T/2 \leq t' \leq T$) half, respectively, of each period $p$ with time interval $0 \leq t' \leq T$. Heavy black arrows indicate translation direction; blue arrows and closed curves indicate velocity vectors and streamlines, respectively.

Fig. 4. Non-chaotic versus chaotic advection illustrated by evolution of blue and red material elements, respectively, in 2D time-periodic lid-driven cavity.

Chaotic advection promotes efficient mixing in laminar flows by (i) rapid global scalar distribution (Fig. 4) and (ii) creation of large “working areas” and steep gradients for diffusion (Fig. 2(b)) by exponential interface stretching. Hence, it is the laminar counterpart to efficient mixing by turbulent diffusion in turbulent flows. However, unlike turbulence, chaotic advection is not a natural mixing mechanism of the flow and in fact can be challenging to accomplish. This is a direct consequence of the fundamental disconnect between the Eulerian velocity field and the Lagrangian transport in laminar flows, allowing complex fluid trajectories to coexist with simple flow fields. Compare to this end the simple step-wise flows in Fig. 3 with the intricate Lagrangian advection in Fig. 4. Thus the Lagrangian approach is essential to describe and understand scalar transport in laminar flows.

The Lagrangian flow topology underlying scalar advvec-
tion in time-periodic flows (as the lid-driven cavity) admits visualisation by so-called “stroboscopic maps” of tracer particles. Such maps visualise the motion of a tracer released at \( \vec{x}_0 \) via the sequence of positions at the end of each period, i.e.

\[
S(\vec{x}_0) = \{ \vec{x}_0, \vec{\Phi}_T(\vec{x}_0), \vec{\Phi}_T^2(\vec{x}_0), \ldots \}, \tag{4}
\]

with

\[
\vec{x}_p = \vec{\Phi}_T(\vec{x}_{p-1}) = \vec{\Phi}_T^p(\vec{x}_0), \tag{5}
\]

the period-wise mapping, as if illuminated by a stroboscope. This is a common technique also often referred to as “Poincaré sectioning” [43].

Fig. 5(a) gives the Lagrangian flow topology visualised by the combined stroboscopic maps (black dots) of a number of tracer particles released on the vertical line \( x = 0.5 \). This exposes a first kind of LCSs, viz. islands (white circular patches), embedded in “chaotic seas” (densely filled regions). Such islands systematically return to their initial position after a given number of periods and act as transport barriers by entrapping material regions during their excursion through the flow domain. The blue element in Fig. 5 is trapped in the island near the top wall that cyclically returns to its initial position after 3 periods through intermediate positions at the island near the bottom and left walls after the first and second periods, respectively. The centres of the islands are periodic points, i.e. material points cyclically returning to the initial position after \( p \geq 1 \) periods following

\[
\vec{x}_0 = \vec{\Phi}_T^p(\vec{x}_0), \tag{6}
\]

commonly denoted elliptic points (Sec. 4.1) [52]. Periodic points for \( p > 1 \) always emerge as sets of \( p \) material points

\[
X_p(\vec{x}_0) = \{ \vec{x}_0, \vec{\Phi}_T(\vec{x}_0), \ldots, \vec{\Phi}_T^{p-1}(\vec{x}_0) \}, \tag{7}
\]

through which each element of \( X_p \) cyclically progresses in the course of time.

A second kind of LCSs in Fig. 5(a) concerns structures termed “manifolds”, which are associated with another type of periodic points known as hyperbolic points (Sec. 4.1) [52]. At each hyperbolic point (marked with a cross in Fig. 5(a)), a stable (blue) and unstable (red) manifold exists that wind their way through the chaotic sea. (The type of a periodic point is determined by the local deformation characteristics; Sec. 4.1) These manifolds are “special” material curves that delineate the principal transport directions in this sea and thus dictate the chaotic advection [43]. The red element in Fig. 5 coincides with the chaotic sea and its chaotic transport – characterised by the repeated stretching and folding – is determined by the unstable manifold. Figs. 5(b-d) demonstrate this by the rapid convergence of the red element on the unstable manifold (coloured in grey); essentially the same convergence occurs for any material element released in the chaotic sea, exposing the unstable manifold as its fundamental mixing pattern (or “mixing template” [43]).

Solenoidal 2D velocity fields \( (\nabla \cdot \vec{u} = 0) \) exclusively admit elliptic/hyperbolic points and associated islands/manifolds [52]. Hence these LCS are the fundamental “building blocks” of Lagrangian flow topologies of 2D incompressible flows. Moreover, solenoidality in 2D imposes a Hamiltonian structure on kinematic equation (2) that makes unsteadiness a necessary (yet not sufficient) condition for chaotic advection in 2D flows. This Hamiltonian structure and its ramifications for the dynamics are detailed in Sec. 4.2. Time-periodic 2D flows – here represented by the lid-driven cavity – meet this requirement and their Lagrangian flow topologies therefore typically consist of arrangements of islands and chaotic seas as in Fig. 5(a) [43].

The Lagrangian flow topology of 3D flows (both steady and unsteady) admits an essentially similar decomposition into distinct LCSs yet encompasses a larger set of such “building blocks” and corresponding interaction scenarios. Furthermore, the aforementioned Hamiltonian structure of kinematic equation (2) is retained only for certain conditions (refer to Sec. 4.3, Sec. 4.4 and Sec. 4.6). Thus 3D flows in general exhibit much greater topological complexity and, in consequence, far richer dynamics than their 2D counterparts.

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1Refer to [www.youtube.com/watch?v=B3dwryNgPXY](https://www.youtube.com/watch?v=B3dwryNgPXY) for an experimental demonstration of chaotic advection and the emergence of a mixing pattern due to the unstable manifold.
parts. LCSs enable systematic and rigorous exposure of this topology and associated dynamics and therefore are indispensable for in-depth transport studies and characterisations. Furthermore, LCSs, given their robustness, can (in principle) be instrumentalised for specific purposes. Efficient mixing requires systematic “destruction” of transport barriers as the islands in 2D; containment of polluted groundwater, on the other hand, requires establishment of such barriers in the subsurface flow. This can (basically) be realised by appropriate flow conditions. Sec. 3 below reconciles the Lagrangian framework and the utilisation of LCSs as “tools” for analysis, characterisation and engineering 3D practical flows to which Lagrangian transport is essential.

3 Lagrangian transport in 3D practical flows

3.1 Introduction

The examples in Fig. 1 clearly demonstrate the diversity and relevance of 3D Lagrangian transport yet applications of concepts and insights remain limited due to the still formidable gap between theory and practice for reasons mentioned in Sec. 1. The only systematic technological application is found in the subclass of industrial and micro-fluidic mixers that, for “proper” conditions, admit reconciliation with 2D flows and the associated theory (Sec. 3.2). Julio Ottino and co-workers laid the groundwork for mixing technology based on Lagrangian chaos in such 2D analogons in the late 1980s (resulting in his classical textbook [52]) and the framework thus developed has since found frequent application in engineering sciences [62, 63, 50, 64, 65, 7, 66, 67, 9, 13, 68, 16, 69, 70, 71, 72, 73]. The Ottino group extended this framework to granular flows that behave as a continuum [26, 27, 28].

Employment of the Lagrangian framework for transport studies beyond industry and technology occurs primarily in the geophysical sciences. Large-scale oceanographic and atmospheric flows, due to their quasi-2D nature, namely also connect well with existing 2D theory [36, 37, 38, 39]. Development of Lagrangian methods specifically for aperiodic flows is in fact strongly motivated by such systems [74, 59, 75].

Dissemination of the above chaos-based mixing technology to the practicing engineering community poses a major challenge in its own right. Many recent handbooks on fluids processing, despite overwhelming evidence of its crucial role in (at least) inline mixers, mention chaotic advection only perfunctorily and continue to rely on properties of the Eulerian velocity field for standard metrics as the “coefficient of variation” (CoV) [76, 77, 5, 78]. Two leading manufacturers

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5 This may lead to erroneous results. A common misconception is e.g. that vortices imply efficient mixing. Reconsider to this end the lid-driven cavity. The flow consists of two alternating vortices (Fig. 4) yet mixing is inefficient (Fig. 5) due to islands in the Lagrangian flow topology (Fig. 5a).

The CoV is in essence the “intensity of segregation” introduced by Danckwerts in the early 1950s [79]. An important shortcoming is arbitrariness of the cell size to determine the scalar concentration, which may cause non-physical effects as artificial diffusion and thus yield misleading results [80]. More sophisticated measures seek to overcome this yet their practical utilisation is scarce to non-existent [43].

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7 Refer to brochures “Mixing and Reaction Technology” (sulzer.com) and “Kenics KMX-V Static Mixer” (chemineer.com).
3.2 Flow in ducts

3.2.1 General configuration

Flows in ducts are assumed periodic in space unless stated otherwise. This assumption makes these problems tractable by allowing representation of the complete system by a periodic subsection and is a valid approximation in important industrial devices as the aforementioned inline mixers. Using this approximation, such periodic duct flows consist of a duct of infinite length and (in principle) arbitrary cross-section composed of repeated duct segments of length $L$. Each duct segment $p$ accommodates a 3D steady flow that is a periodic repetition of that in the first segment $p = 1$, i.e.

$$\bar{u}(x,y,z + pL) = \bar{u}(x,y,z),$$

(8)

rendering the duct flow the spatially-periodic counterpart to the time-periodic flow \([3]\). The duct segments are, in turn, partitioned into $N$ axial “cells” of length $\Delta = L/N$. Each cell $k$ accommodates a 3D steady flow that is a reorientation of flow $\bar{v}$ in the first cell $k = 1$, i.e.

$$\bar{u}(r, \theta, z + k\Delta) = \bar{v}(r, \theta - \Theta_k(z), z),$$

(9)

with axial reorientation following

$$\Theta_k(z) = \Theta \sum_{k=1}^{N-1} \mathcal{H}(z - kL),$$

(10)

$\Theta$ the cell-wise reorientation angle such that $N\Theta$ is commensurate with $2\pi$ and $\mathcal{H}$ the Heaviside function \([83]\).

3.2.2 Industrial and technological relevance

Periodic duct flows constitute the generic configuration for industrial devices involving continuous processing or treatment of laminar fluid streams. Applications are numerous and include [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19].

- inline mixers for food processing;
- extruders for polymer processing;
- compact inline mixers for process intensification;
- inline micro-mixers and throughflow compartments in micro-fluidic (lab-on-a-chip) devices.

The purpose is mixing of a continuous axial throughflow at velocity $u_c$ (typically pressure-driven) by systematic reorientation of the transverse flow ($u_x, u_y$) via e.g. static internal mixing elements [1, 2, 3, 4]. Fig. 6 demonstrates this principle for the RAM introduced above [66]. Here the transverse flow is driven by viscous drag exerted by a rotating outer cylinder via apertures in the stationary inner cylinder (Fig. 6(a)); the cell-wise reorientation angle $\Theta$ following (10) is the angular offset of consecutive apertures accomplishes systematic downstream reorientation of the transverse flow following Fig. 6(b). Each tube section containing an aperture defining one cell and shown black/blue/red transverse streamline patterns correspond with flow (9) in cells $k = 1, 2, 3$, respectively, for $\Theta = 2\pi/3$ of duct segments of $N = 3$ cells.

The RAM is inspired by the Partitioned-Pipe Mixer (PPM) shown in Fig. 6(c) \([85]\). The PPM achieves reorientation by the combined effect of a rotating cylinder and alternating horizontal/vertical static internal elements that repeatedly split and recombine the axial throughflow. The PPM serves as model problem for the pioneering work on industrial mixers by the Ottino group (Sec. 5.1). The RAM and PPM capture the essentials of two kinds of periodic duct flows relevant in the present context:

- **Open duct flows**: consist of a duct devoid of internal walls and rely on reorientation by the transverse boundary forcing as primary mixing mechanism.
- **Partitioned duct flows**: consist of a duct partitioned by internal walls and rely on reorientation by splitting and recombination of the axial flow as primary mechanism.

Moreover, the RAM and PPM have both been the subject of in-depth (experimental) Lagrangian transport analyses and thus are well-suited to bridge the theory–practice gap. Hence, the RAM and PPM are adopted hereafter as the two representative periodic duct flows.

**Generic flow topology** The generic 3D flow topology of periodic duct flows is demonstrated in Fig. 7(a) for the RAM.
It typically consists of families of concentric stream tubes running from inlet to outlet of the duct segment (3 specimens highlighted in colour) embedded in chaotic streamlines (black). Fig. 7(b) gives the corresponding axial cross-section at the inlet and reveals a transverse topology reminiscent of the stroboscopic map of the lid-driven cavity in Fig. 5(a) islands in a chaotic sea. This reflects the fundamental property that 3D steady duct flows (in principle) are dynamically equivalent to 2D time-periodic flows and kinematic equation (2) admits a (formal) transformation $\mathcal{F}$ following

$$\frac{d}{dt} \begin{bmatrix} x \\ y \\ z \end{bmatrix} = \begin{bmatrix} u_x \\ u_y \\ u_z \end{bmatrix} \Rightarrow \frac{d}{d\tau} \begin{bmatrix} \zeta_1 \\ \zeta_2 \end{bmatrix} = \begin{bmatrix} v_1 \\ v_2 \end{bmatrix}, \quad (11)$$

such that the transverse and axial Lagrangian motion corresponds with transport in the 2D domain $(\zeta_1, \zeta_2)$ and periodic evolution in “time” $\tau$, respectively. The LCSs relate as follows: periodic points, islands and 1D manifolds (i.e. curves) in 2D materialise as axially reconnecting streamlines, tubes and 2D manifolds (i.e. surfaces), respectively, in 3D (Sec 4.1.4).

The Lagrangian framework developed by the Ottino group (Sec 3.1) implicitly relies on transformation (11) following Sec 4.3.2 and studies on the RAM explicitly demonstrate its existence (at least) for generic open-duct flows [83,86]. Localised phenomena such as reversed axial flow or internal recirculations may restrict this transformation – and the equivalence with 2D dynamics – to a “net through-flow region” yet this is inconsequential for the functionality of open-duct mixers [86].

The equivalence (of at least said net throughflow region) with 2D unsteady flows means that 3D steady duct flows can basically always be designed to achieve global chaotic advection. Parametric studies enable systematic isolation of the appropriate operating conditions and thus are a common approach for optimisation and design of actual devices [62,63,50,64,65,71,66,67,9,13,68,16,69,71,72,73]. Fig. 7(c) gives the stroboscopic map for the RAM optimised by this approach, yielding a chaotic state devoid of stream tubes and, inherently, efficient mixing.

The generic composition of 3D Lagrangian flow topologies according to Fig. 7(a) and the impact upon the transport is demonstrated experimentally via dye visualisation for the RAM in Fig. 8(a) and the PPM in Fig. 8(b) [85,66]. Two situations are considered for each system, viz. a poor-mixing case (top) with cross-sectional stroboscopic map containing islands similar to Fig. 7(b) and a good-mixing case (bottom) exhibiting global chaos as in Fig. 7(c) (Stroboscopic maps of the PPM are essentially similar as in Fig. 7 [85,87]). The dye visualisations in both RAM and PPM indirectly expose the coexisting tubes and chaotic regions of the poor-mixing case via the entrainment of the red dye and transverse spreading of the green dye, respectively. The global chaotic advection of the good-mixing case is clearly demonstrated by the transverse spreading of both the red and green dye and the resulting formation of a homogeneous (yellowish) mixture.
Chaotic advection implies cross-sectional mixing patterns with downstream progression similar to Fig. 4. This is demonstrated in Fig. 9(a) by the simulated evolution of a binary concentration distribution in the RAM, revealing the characteristic stretching and folding imparted by the unstable manifold (not shown) following Fig. 5 [88]. Experimental visualisation using fluorescent dye illuminated by a transverse light sheet near the outlet of the RAM set-up of Fig. 8(a) exposes (for comparable operating conditions) an essentially similar cross-sectional mixing pattern (Fig. 9(b)) [66]. This experimentally validates the key mechanism underlying chaotic advection in inline mixers and indirectly visualises the governing LCS(s), viz. the unstable manifold(s).

Open-duct flows: micro-mixers using patterned walls

Inline mixers of the open-duct type are, apart from the RAM, found mainly in micro-fluidics and usually accomplish transverse flow and reorientation by patterned duct walls. Consider as an example the square micro-channel with cross-baffles extending from top and bottom channel walls shown in Fig. 10(a) [89]. The cross-baffles merely “deform” the duct boundary without partitioning the domain and thus the system indeed constitutes an open-duct flow. This holds, irrespective of the geometric complexity, for any wall pattern and duct shape. Fig. 10(b) gives a typical cross-sectional mixing pattern in the above micro-mixer according to the simulated evolution of a patch of black tracer particles (left) versus experimental visualisation by fluorescent dye via transverse light sheets (right). This reveals the same progressive stretching and folding as in the RAM (Fig. 9) and thus demonstrates (i) the universality of Lagrangian (chaotic) transport phenomena of open-duct flows and (ii) the intrinsic similarity in mixing characteristics of (practical) flows belonging to this class. Moreover, the close agreement between computational and experimental results further substantiates the physical validity and meaningfulness of the Lagrangian concept.

Other inline micro-mixers adopting patterned walls for reorientation – thus defining open-duct flows with basically the same transport characteristics as above – include the “staggered-herringbone mixer” (using grooved walls) and the “C-shaped serpentine micromixer” (using C-shaped duct segments) and numerous variations on these designs [90, 13]. State-of-the-art micro-manufacturing technologies make commercial fabrication of such devices increasingly feasible and may thus become a key enabler for the practical application of chaos-based micro-mixing technology [91].

Partitioned-duct flows: static mixers using baffles

Inline mixers of the partitioned-duct type have a long history as
static mixers in the traditional processing industry. These devices rely on partitioning of the flow domain by internal static mixing elements ("baffles") for reorientation and the Kenics mixer is perhaps the best known incarnation of this principle. Its original design dates back to the 1960s \cite{92} and to this day finds widespread utilisation in fluids engineering (Sec. 5.2.1) \cite{1,2}. Modified Kenics designs have even been implemented in micro-mixers \cite{93}. Hence, the Kenics mixer, arguably, is the archetypical static mixer and on that grounds served as model for the PPM \cite{52}.

The Quatro mixer (Primix BV, Mijdrecht, The Netherlands) is a static mixer reminiscent of the Kenics mixer \cite{94}. Its mixing elements consist of chevron-shaped central plates with perpendicular elliptical segments extending to the cylinder wall and are alternately reversed and rotated about the cylinder axis (grey elements 1 and 2 in Fig. 11). This yields a periodic repetition of pairs of mixing elements analogous to the consecutive pairs of reordered plates in the PPM.

Fig. 11 demonstrates the transverse mixing within each duct segment by horizontal and vertical splitting (and subsequent recombination) of the axial throughflow by the central plates of elements 1 and 2, respectively, via the 3D streamline pattern measured with 3D Particle Tracking Velocimetry (3DPTV) \cite{94,84}. Mixing element 1 splits the incoming fluid into two separate streams (indicated in red and green) that, in turn, undergo further splitting by element 2, resulting in a "mixture" of red and green streamlines at the segment exit. Repetition of this process by consecutive duct segments yields ever finer striations as visualised in Fig. 12(a) by reverse cuts of the 3D distribution of a fluorescent dye (bright) injected at the inlet and measured by 3D Laser-Induced Fluorescence (3DLIF) \cite{84}. Here mixing elements 1,3,5 are the upstream elements (i.e. element 1 in Fig. 11) in the consecutive duct segments 1,2,3, respectively, and shown cuts are halfway the central horizontal plate (white bar).

The cross-sectional mixing pattern in Fig. 12(a) exhibits stretching and folding (signifying chaotic advection) akin to the RAM (Fig. 9) and the micro-mixer (Fig. 10) and thus the Quatro mixer, notwithstanding the entirely different designs of the devices, accomplishes efficient mixing by essentially the same Lagrangian mechanism. However, the underlying 3D fluid motion is markedly different. Said open duct mixers effectively coil up streamlines reminiscent of twirling spaghetti around a fork, illustrated in particular by the black chaotic streamlines in Fig. 7(a) giving rise to cross-sectional swirling (Fig. 9). The (partitioned-duct) Quatro mixer, on the other hand, progressively "slices" the downstream fluid stream not unlike a meat grinder, as visualised in Fig. 12(b) by longitudinal cuts of the 3D dye distribution in the indicated mixing elements (top view of central horizontal plate). This manifests itself in the cross-sectional filamentation shown in Fig. 12(a).

Inline static mixers, regardless of baffle shape and fluid rheology, exhibit basically the same Lagrangian transport characteristics as the Quatro mixer. Consider to this end e.g. the qualitative resemblance of the cross-sectional mixing patterns in Fig. 12(a) with the dye visualisations of those for shear-thinning fluids in Kenics static mixers \cite{80}. The wide variety of baffles employed in industry is nonetheless designed primarily by empirical studies and case-specific numerical simulations using standard mixing measures such as the CoV (Sec. 3.4). Design and optimisation of static mixers using Lagrangian concepts, on the other hand, is rare and mainly restricted to studies in engineering sciences on the beforementioned Kenics and Sulzer mixers \cite{62,63,95,70}. This strongly suggests that many mixer designs are in fact sub-optimal and industry may greatly benefit from dedicated engineering tools as e.g. post-processing modules in a (commercial) CFD package for determination of cross-sectional stroboscopic maps. The latter namely enables systematic and conclusive isolation of the operating conditions that yield global chaotic advection as in Fig. 7(c).

An open fundamental issue concerns the impact of the internal walls on the Lagrangian framework. The existing approach implicitly relies on transformation \cite{11} and studies to date basically confirm its validity. However, internal walls may, besides the beforementioned reversed axial flow and recirculation, introduce singularities that (locally) break the link with 2D unsteady systems and thus pave the way to essentially 3D phenomena (Sec. 4.3.2). In particular LCSs emerging from critical points on the internal boundaries (instead of periodic points \cite{8} and corresponding reconnecting streamlines in the flow interior) are likely to play a (decisive) role in the transport characteristics on grounds of the analogy with transport in porous media. The latter namely involves similar splitting and recombination of throughflows by internal walls and is in fact dominated by such LCSs (Sec. 3.5.1). Hence investigation of transformation \cite{11} for partitioned flow domains and wall-induced LCSs according to Sec. 3.5.1 may strengthen the theoretical foundation and yield important insights with possible new applications.

Partitioned-duct flows: branching micro-channels The partitioned-duct principle finds application also in micro-fluidic inline mixers yet, owing to the difficult fabrication of microscopic baffles, typically in the form of branching and
reconnecting channels. Consider for illustration the chaotic serpentine micro-mixer (CSM) [96, 97]. Fig. 13(a) shows one duct segment comprising of two branching-reconnecting cells that each are composed of a pair of reversed L-shaped channels. Thus each cell in the CSM, despite distributing the fluid streams over two physically separate channels, gives rise to basically the same splitting and recombination of the axial throughflow as imparted by each mixing element in the Quatro mixer. Fig. 13(b) demonstrates this by the simulated foliation of the internal 3D material interface separating the incoming fluid streams, revealing a “slicing” as in Fig. 11 and Fig. 12(b). The corresponding cross-sectional mixing pattern is shown in Fig. 13(c) and exhibits similar filamentation as in Fig. 12(a). Hence, transport in the CSM, despite completely different designs and length scales, indeed emanates from essentially the same Lagrangian mechanisms as the Quatro mixer and, inherently, any partitioned-duct mixer.

**Aperiodic duct flows** A class of inline mixers intimately related to the above devices exists in aperiodic duct flows, that is, axial throughflows without the periodic structure following (8) and (9). Consider for illustration the T-shaped micro-mixer in Fig. 14(a): two fluid streams are brought together via separate inlets and undergo transverse mixing by a vortical flow in one common outlet duct (reminiscent of open-duct flows) induced at the junction in case of sufficiently strong fluid inertia (Fig. 14(b)) [98, 9]. Said vortical flow (termed “engulfment flow” in [98, 9] and occurring if the Reynolds number $Re$ exceeds a certain threshold) is characterised by two adjacent counter-rotating longitudinal swirling flows that intensify in downstream direction and each cross the vertical axial midplane of the duct (Fig. 14(b)). This yields a 3D mixing pattern consisting of two longitudinal “mixing rolls” visualised in Fig. 15(a) by iso-contours of a 3D dye distribution – and corresponding cross-sectional evolution (Fig. 15(b)) – using 3DLIF [98]. Numerical simulations for similar flow conditions (Fig. 15(c)) reveals a roll-wise “stretching and folding” pattern reminiscent of the RAM (Fig. 9). This implies similar transport characteristics as the periodic counterparts and thus strongly suggests that the nature of the axial evolution, i.e. periodic versus aperiodic, is of secondary importance to this process.

The (spatial) aperiodicity has in fact primarily theoretical/conceptual ramifications by considerably limiting the
usefulness of the existing Lagrangian machinery for transport analysis due to its strong reliance on stroboscopic maps and LCSs associated with periodic points (6) (Sec. 2). This limitation is of even greater significance for geophysical flows and in particular the latter spurred scientific efforts, with George Haller and co-workers at the vanguard, to generalise the notion of coherence imparted by “special” Lagrangian entities (i.e. LCSs) to aperiodic flows [59]. Relevant here are particularly so-called attracting LCSs, i.e. the aperiodic counterparts to the unstable manifolds in Fig. 5(a), which in a similar manner as the latter govern stretching and folding – and thus chaotic advection – in aperiodic flows [59] (Sec. 4.1). Isolation of attracting LCSs in the T-shaped micro-mixer is (to the best of our knowledge) outstanding yet their identification in an analogous flow in life sciences, i.e. an artery bifurcation (Fig. 16 in Sec. 3.2.3), provides compelling evidence of their existence. This, in turn, underpins the above conjecture that the transport characteristics of (a)periodic duct flows are in essence the same. However, generalisation of the Lagrangian framework to a universal and unambiguous approach for aperiodic and, intimately related, finite-time (transient) flows is still in progress (Sec. 6).

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10Term “Lagrangian coherent structures” (LCSs) in fact stems from these efforts and here denotes such entities in both periodic and aperiodic flows.
using the experimental flow, are given in Fig. 16(b) in the transverse cross-sections 1 and 3 (indicated by \( T < 0 \)) and exhibit similar stretching and folding as their periodic counterparts, i.e. the unstable manifolds, in Fig. 5(a). Shown also are the corresponding repelling LCSs (indicated by \( T < 0 \)) in cross-section 1 and stream-wise cross-section 2, which are the aperiodic counterparts to the stable manifolds in Fig. 5(a). Existence of these entities – and their similarity to manifolds in chaotic seas – strongly suggests chaotic advection in the artery bifurcation and, by analogy, also in the T-shaped micro-mixer. Moreover, the implication of chaotic advection with atherosclerosis in carotid arteries is consistent with its purported role in blood-clot and plaque formation in aneurysms (Sec. 3.1 and Sec. 3.3.3). Hence, chaotic advection could be, by tending to augment the residence time of blood-borne substances \[29\], a central player in the development of vascular diseases. Further exploitation of the resemblance with industrial inline mixers and application of the corresponding (scientific) knowledge on (chaotic) Lagrangian transport may contribute to the understanding of these processes.

**Tokamak fusion reactors** A remarkable analogy exists between periodic duct flows \( \vec{u} \) and the magnetic fields \( \vec{B} \) of magneto-hydrodynamic (MHD) descriptions of plasmas in tokamak fusion reactors (Fig. 17(a)). First, a periodic duct is topologically a torus and thus equivalent to the interior of said reactors. Second, flow and magnetic fields are both solenoidal, i.e. \( \vec{\nabla} \cdot \vec{u} = 0 \) and \( \vec{\nabla} \cdot \vec{B} = 0 \). Third, the toroidal-poloidal magnetic forcing imposes similar net flux and reorientation as the axial-transverse forcing in periodic ducts. Thus the tokamak admits a transformation \( \mathcal{F} \) following \((11)\) of magnetic field \( \vec{B} \) and toroidal reference frame \((\sigma, \eta, \phi)\) such that poloidal \((\sigma, \eta)\) and toroidal \((\phi)\) dynamics correspond with canonical space \((\zeta_1, \zeta_2)\) and time \(\tau\), respectively. These commonalities render periodic duct flows and magnetic fields in MHD tokamak models topologically and kinematically equivalent \[54,101,100\].

The objective in tokamak fusion plasmas is accomplishment of so-called “magnetic confinement”, that is, a magnetic topology consisting entirely of one global family of concentric tubes (denoted “magnetic flux surfaces” and indicated schematically by the purple torus in Fig. 17(a)) similar to the stream tubes in the RAM (Fig. 7(a)). However, this remains a major challenge and is in fact the principal hindrance to the envisaged application of fusion plasmas as heat source for power plants (and safe alternative to nuclear fission) \[100\].

Magnetic confinement may be broken by the disintegration of certain flux surfaces through the nonlinear process of reconnection, yielding metastable states denoted “neoclassical tearing modes” (NTMs) that precede global instability of the magnetic field \[101,109\]. Such NTMs are often investigated in an oversimplified manner, though. First, their (poloidal) magnetic topology is typically assumed to entirely consist of one central island with multiple satellite islands arranged by “o-points” and “x-points” following Fig. 17(b) \[100\]. (Former and latter are the magnetic equivalents to elliptic and hyperbolic points, respectively, in flow topologies.) The analogy with periodic duct flows strongly suggests far more complex NTM topologies similar to Fig. 7. Both numerical studies and experimental evidence support this \[102,103,104,105\]. Fig. 17(c) gives a typical simulated poloidal NTM topology that indeed exhibits the expected characteristics; the red curve is a shearless transport barrier and is (considered) key to the survival of the remaining flux surfaces \[105\]. Second, transport by chaotic magnetic field lines is commonly regarded a stochastic turbulent-diffusion-like mechanism (“magnetic turbulence”) devoid of spatial organisation \[100,106\]. However, chaotic advection in mixing flows implies a (though highly complex) well-defined spatial structure imparted by manifolds (Fig. 5) and thus fundamentally contradicts this key assumption. Dedicated studies on chaotic magnetic field lines are indicative of such a spatial structure in NTM topologies \[107,106\].

The above strongly suggests that incorporation of the true (3D) topological structure of the magnetic field and in—

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\[11\] This holds equally for the so-called “flux coordinates” often employed in tokamaks; these are namely intimately related to the toroidal frame \[100\].
3.3 Flow in containers

3.3.1 General configuration

Containers are defined as 3D flow domains confined by a closed surface (i.e. flux across the boundaries is absent) that are topologically equivalent to spheres. This includes, besides the latter, common geometries as cylinders and cubes. The key difference with periodic ducts exists in the nature of the domain. Periodic ducts and containers namely are topologically a torus and a sphere, respectively, which can have fundamental consequences for the Lagrangian flow topology and associated transport properties.

Flow and transport in periodic ducts and containers constitute the two principal canonical problems in the present categorisation. The surface classification theorem namely states that orientable closed surfaces are topologically equivalent to the surface of either a connected sum of tori or a single sphere \([108]\). Hence, tori and spheres are the fundamental building blocks of 3D (realistic) flow domains and Lagrangian transport in any such configuration thus intimately relates to that in periodic ducts and/or containers.

3.3.2 Industrial and technological relevance

Flows in containers are the generic configuration for industrial devices involving batch processing or treatment of fluid bodies under laminar conditions. Practical applications include \([1, 2, 109, 11, 110, 111, 112]\):

- bio-reactors;
- food processing;
- electro-magnetic materials processing;
- batch micro-mixers and processing chambers in micro-fluidic (lab-on-a-chip) devices.

The confinement by the closed boundary imparts a flow (topology) that generically involves a global circulation and consists of (multiple) vortical structures. Consider as a first illustration the impeller-driven flow in Fig. 1(a) here the toroidal tracer paths reveal a primary circulation about the impeller and a secondary circulation transverse to this. However, unlike periodic duct flows, the flow (topology) in containers may vary significantly with driving mechanism (i.e. surface versus body forcing) and time signature (i.e. steady versus unsteady). Hence, this flow class is discussed below in terms of the following basic practical configurations:

- steady flow driven by body forcing;
- steady flow driven by surface forcing;
- unsteady flow.

This classification is somewhat pragmatic by leaning on relevant practical characteristics of container flows and is adopted here for reasons of accessibility. A more rigorous (yet at this point inscrutable) differentiation can be made on the basis of dynamic characteristics and emerges naturally in the course of the following discussion.

**Steady flow driven by body forcing** The abovementioned impeller-driven flow is archetypal of a steady container flow driven by body forcing; the immersed stirring device namely effectively acts as a (local) body force. This is the most common configuration in industrial batch mixers and thus of great practical relevance.

The study by \([113, 114]\) adopts the flow inside a cylindrical container driven by a tilted rotating disk following Fig. 1(a) as physical model for industrial batch mixers. (The specific domain and stirrer geometry are of secondary importance.) Fig. 1(b) visualises the Lagrangian transport using fluorescent dye and reveals, consistent with the toroidal paths in Fig. 1(a), a 3D flow topology comprising of nested closed stream tubes (one specimen highlighted by red dye) encircling the rotation axis of the impeller. The corresponding vertical cross-section, visualised in Fig. 1(c) by red and green dye, and intersections of 3D computational streamlines (Fig. 1(d)) imply an internal structure resembling the transverse topology of duct flows (Fig. 2(b)): islands in a chaotic sea. This has the fundamental implication that, despite topologically different flow domains, the 3D Lagrangian motion and flow topology of the impeller-driven flow are dynamically similar to periodic duct flows. The similarity is particularly apparent upon comparison with the tokamak fusion reactor in Fig. 17. The cross-sectional and azimuthal
dynamics in the impeller-driven flow are equivalent to the poloidal/toroidal dynamics of the tokamak and, in turn, to the transverse/axial dynamics of periodic duct flows. This implies for the impeller-driven flow a transformation (11) of the cylindrical reference frame \((r,\theta,z)\) such that canonical space \((\zeta_1,\zeta_2)\) and time \(\tau\) correspond with \(rz\)-plane and azimuthal component \(\theta\), respectively. (Refer to Sec. 4.3.3 and Sec. 4.6.2 for the conditions permitting this transformation.) The (closed) stream tubes as shown in Fig. 7(b) and Fig. 18(b) and the magnetic flux surfaces in Fig. 17(a) thus constitute equivalent LCSs and are denoted “Kolmogorov-Arnold-Moser (KAM) tori” in dynamical-systems terminology [52,100]. This nomenclature is adopted hereafter to indicate such entities.

Lagrangian flow topologies consisting of KAM tori embedded in chaos is generic for steady flows driven by body forcing. The particular forcing and/or domain geometry may (significantly) affect the emergence and arrangement of KAM tori and other LCSs, though. Consider to this end the impact of the impeller shape and positioning on the flow topology of the stirred container in Fig. 18(a). A single impeller consisting of blades and rotating about the cylinder axis (i.e. a so-called “Rushton impeller” widely used in industry) results for sufficiently strong body forcing: electro-magnetic stirring of glass melts [110]. This relies on buoyancy (induced by Joule heating via an electric current) in conjunction with a Lorentz force (induced via interaction of said current with an external magnetic field).

The above experimental analyses reveals a dramatic impact of the impeller geometry on the mixing properties. Stirring by disks rotating about the cylinder axis yields only KAM tori; tilting of the rotation axis is essential to induce chaos yet this remains highly localised (Fig. 18) [114]. The Rushton impellers, on the other hand, yield significant chaotic regions (Fig. 19) and, potentially, global chaos. The generic composition of the flow topology (i.e. KAM tori and chaos) nonetheless is essentially the same in all cases.

The generality of KAM tori as key LCSs in 3D steady container flows – and the dynamic analogy with periodic duct flows – admits further substantiation by an entirely different (industrial) example involving (global) body forcing: electro-magnetic stirring of liquid metals. However, this occurs (unlike glass melts) under turbulent flow conditions and thus is beyond the present scope [117].
following Fig. 20(a) (shown is one torus of one family described by a single streamline). Activation of the magnetic field (\( \vec{B} \neq \vec{0} \)) changes this completely into a steady swirling flow and corresponding topology of one global family of tori centered on the central electrode (Fig. 20(b)) [110]. KAM tori nonetheless persist as the topological building blocks and their arrangement is similar to that of the individual families in the above impeller-driven flows (Fig. 18 and Fig. 19).

However, an essential difference with the latter is (at least in the considered parameter ranges) the absence of (local) inertial forces, their capacity as barriers to fluid transport, are detrimental to chaotic advection; this requires unsteady electro-magnetic stirring and is addressed below.

The above concerns practical utilisations of Lagrangian transport seeking to accomplish chaotic advection for the purpose of efficient mixing and dispersion. KAM tori, in their capacity as barriers to fluid transport, are detrimental to this process. However, industrial batch processing often aims at solid-liquid separation in suspensions of finite-size particles for e.g. waste-water treatment or biorefinery. KAM tori offer a way to realise such “unmixing” for (at least) small particle parcels. The weak deviation in Lagrangian motion namely has a remarkable effect by causing such particles to migrate towards KAM tori and thus cluster there. This “unmixing” is demonstrated experimentally in Fig. 21 for the above steady flow driven by a single Rushton impeller via the clustering of polystyrene particles suspended in glycerine in the KAM tori at the impeller (Fig. 19(a)) [44]. Fig. 1(a) shows individual particle trajectories in this process.

Following Fig. 21 stems from the fact that the Lagrangian dynamics, though also described by a kinematic equation as \( \vec{\dot{v}} \), is determined by a particle velocity \( \vec{v} \) instead of the fluid velocity \( \vec{u} \). The fundamental difference between both fields is that the former is non-solenoidal, i.e. \( \nabla \cdot \vec{v} \neq 0 \), and its (asymptotic) dynamics, in consequence, is governed by so-called “attractors”, that is, LCSs associated with \( \vec{v} \) onto which the particles converge in the course of time. Such entities are absent in solenoidal flows and thus non-existent for the Lagrangian flow topology underlying the fluid motion. However, an intriguing connection occurs in that the attractors for the particular kind of particles considered here are in fact the KAM tori [119]. This gives rise to the “unmixing” of particles shown in Fig. 21. General criteria for particle clustering in LCSs are developed in [120].

Steady flow driven by surface forcing Flows in a cylindrical cavity due to rotating or translating lids are representative examples of surface-driven flows and are 3D counterparts of the 2D lid-driven cavity flow introduced in Sec. 2. The flow and transport characteristics depend essentially on the specific lid motion (i.e. rotating versus translating) and time signature (i.e. steady versus unsteady).

Steady rotation of the bottom lid gives rise to a swirling flow and associated flow topology comparable to the steady impeller-driven and electro-magnetically-driven flows in Fig. 18 and Fig. 20(b) respectively: a family of KAM tori centered on the cylinder axis and surrounded by chaos. This is demonstrated experimentally in Fig. 22 by the time-averaged light intensity emitted by a fluorescent dye in a vertical cross-section; its level sets (differentiated by colour) expose the characteristic island structure including localised LCSs as the indicated period-2/3/4 island chains [121]. Moreover, this reveals a progressive breakdown of tori into chaos with increasing Reynolds number \( Re \).

Steady translation of the bottom lid yields a circulatory flow consisting of two symmetric vortices similar to the buoyancy-driven flow in Fig. 20(a) [122][123]). Fig. 23 demonstrates the emergence of the corresponding flow topology by a typical streamline in case of translation in \( +x \)-direction. The non-inertial limit \( Re = 0 \) yields a closed streamline (Fig. 23(a)) that, upon introduction of fluid inertia, becomes non-closed and describes a torus as shown for \( Re = 10 \) in Fig. 23(b). Thus two global families of tori separated by the symmetry plane \( y = 0 \) are created for

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\[^{14}\text{This is largely inherent in the particular configuration, though. Buoyancy alone may (in 3D steady container flows) namely already yield intricate flow topologies with multiple tori families and chaotic regions [118].}\]
“sufficiently small” \( \text{Re} \). The streamline becomes chaotic upon further increasing \( \text{Re} \), as demonstrated for \( \text{Re} = 100 \) in Fig. 23(c), signifying breakdown of certain tori and resulting in KAM tori embedded in chaos (outlined by the corresponding cross-section with the plane \( x = 0 \) in Fig. 23(d)) similar as the above rotating-lid case (Fig. 22). The flow field and streamline pattern are validated experimentally by 3D PTV in [124][125]. Single-wall forcing of cubical cavities yields essentially the same flow topology as in Fig. 23, implying, similar as the previous impeller-driven flows, an only secondary importance of the specific geometry [123]. Moreover, forcing by multiple walls or wall segments typically induces bifurcations into more complex topologies yet nonetheless with a generic composition of (multiple) families of tori embedded in chaos [126][127].

The transition of the flow topology from a global family of closed streamlines in the non-inertial limit \( \text{Re} = 0 \) to two global families of tori, as demonstrated for a generic streamline in Fig. 23 happens for any \( \text{Re} = 0 \). Moreover, other minute perturbations of the non-inertial limit as e.g. asymmetric forcing have the same effect [126][128]. Thus the response of the steady cylinder flow to fluid inertia in fact reflects universal behaviour. Flow topologies in any system consisting entirely of closed streamlines namely constitute a singular limit in that the slightest perturbation induces transition into (multiple families of) tori that robustly survive a finite perturbation strength (here \( \text{Re} \lesssim 10 \)) [129][130]. The impeller-driven flow in Fig. 18 exhibits this behaviour e.g. as a function of the tilt angle \( \alpha \); aligned impellers \( \alpha = 0 \) yield closed circular streamlines about the cylinder axis; tilting \( \alpha \neq 0 \) results in tori formation as in Fig. 22. Hence, given realistic systems are inherently imperfect, tori are the LCSs that make-up the “true” non-chaotic baseline topology of both the cylinder flow and the other cases considered so far as well as most of the configurations discussed hereafter. Flows in micro-droplets are, for instance, a further manifestation of this scenario [Sec. 3.4.2].

A practical instance of 3D steady container flows driven by translational surface forcing, though not immediately apparent, exists in micro/nano-particle separation in microfluidic channel flows driven by AC electro-osmosis (ACEO) [132]. Fig. 24(a) illustrates the working principle: micro-electrodes at the channel bottom induce a slip velocity in the electric double layer (EDL), setting up vortices that trap and concentrate particles by the resulting centrifugal forces (\( \vec{F}_{\text{HD}} \)). The local dielectrophoretic force (\( \vec{F}_{\text{pDEP}} \)) at the electrode centre supports this process by pushing particles into the bulk flow. The ACEO-driven flow is dynamically equivalent to said container flows: (i) separatrices between adjacent vortices effectively compartmentalise the channel into confined domains; (ii) the EDL has a negligible thickness and the slip velocity thus effectively acts as an elastic translating wall. Fig. 24(b) gives the 3D flow topology associated with each electrode (dark patch) [128]. This exposes a composition of KAM tori (3 specimens highlighted in colour) and chaotic streamlines (black) within each of the compartments bounded by the separatrices (transparent planes) that is indeed similar to the lid-driven container flow in Fig. 23. Measurements of the 3D streamline pattern by astigmatism \( \mu \)-PTV (Fig. 24(c)) provides the first experimental validation of this structure [133]. Here dark/light blue streamlines remain within the subdomains divided by the separatrix at \( x = 35 \mu m \) (transparant plane). Red streamlines cross this separatix due to experimental imperfections...
that, akin to tori formation from closed streamlines, act as natural perturbations of an idealised state. Essentially the same perturbation-induced separatrix crossing may occur in droplet flows (Sec. 3.4.2) and convection cells (Sec. 3.5.2); its underlying mechanisms are elaborated in Sec. 3.4.2.

The ACEO-driven flow is situated in a duct and thus admits transport characteristics of both duct and container flows. Imposition of an axial (x-wise) pressure gradient of “appropriate” strength sets up a throughflow that passes over the vortical structures shown in Fig. 24(b) and thus yields a flow topology comprising of (diminished) tori at the electrode and stream-wise tubes as in Fig. 24(a) surrounded by a chaotic region [128]. Tracers in this chaotic region exhibit both container-flow and duct-flow dynamics by prolonged entrapment in the vortical structure during their excursion from inlet to outlet. Similar hybrid behaviour occurs also in the flow inside an aneurysm shown in Fig. 1(b) and is examined further in Sec. 3.3.3.

**Unsteady flows** Dependence of the flow $\bar{u}$ on time $t$ expands the phase space of kinematic equation (2) from the 3D physical domain to the 4D space-time domain. This affords greater dynamic freedom and generically tends to promote chaotic advection. Reconsider for illustration the flow inside a cylindrical container driven by a rotating disk (Fig. 18). Stirring of a Newtonian fluid yields a steady flow and corresponding flow topology consisting of KAM tori embedded in chaos (Fig. 15). Tilting of the disk is imperative to attain this state; alignment with the cylinder axis yields a topology devoid of chaos and comprising entirely of tori [114]. (Recall that this constitutes the “true” baseline topology emanating from a singular state consisting of closed streamlines.) Non-Newtonian fluid rheology may fundamentally change the latter by (for steady stirring) inducing *time-periodic* oscillations and, in consequence, richer dynamics. Fig. 25(a) demonstrates this for a fluid exhibiting both visco-elastic and shear-thinning behaviour stirred by a rotating disk aligned with the cylinder axis [134]. Shown is a typical instantaneous dye distribution in the vertical cross-section that visualises the mixing pattern and thus outlines the LCSs in the corresponding stroboscopic map as in Fig. 5. The emergence of “rippled” structures (green) enveloping islands (blue/yellow) implies a 3D flow topology of KAM tori and chaos basically similar to that emanating from the tilted disk (Fig. 18(c)) and the single Rushton impeller (Fig. 19(a)) for Newtonian fluids. Thus unsteadiness promotes chaos in a comparable way as a non-trivial geometry in said steady flows. Here in particular the high-shear region around the impeller (indicated by red in Fig. 25(a)) is the physical cause of the rheology-induced time-periodicity.

Introduction of time-periodicity in the electro-magnetic stirring of glass melts by an oscillating magnetic field in Fig. 20 has essentially the same effect: breakdown of the global family of tori in Fig. 20(b) into chaos. Fig. 25(b) demonstrates this by the resulting global dispersion of 25,600 tracers released on a small line segment of length $l/R = 0.02$ in the axial midplane (top view) [110]. First experimental evidence of 3D chaos in unsteady flows such as those in Fig. 20(b) has been obtained by direct measurement of 3D stroboscopic maps of a single tracer via combined 2DPTV in two projections in a time-periodic swirling flow driven by alternate eccentric rotation of upper and lower endwalls [135].

The above container flows, notwithstanding substantial differences in configuration and forcing, in essence all concern cases in which the Lagrangian dynamics are intrinsically linked to the emergence and breakdown of KAM tori. This renders these flows dynamically analogous both to one another and to periodic duct flows with behaviour that, in general, is similar to that of 2D unsteady systems. The link with 2D systems is broken only near possible singularities in transformation (11) and/or upon development of 3D defects.
Global chaos as in Fig. 26(a) (right) sets in after total breakdown of transport-obstructing LCSs as e.g. KAM tori or spheroids in response to some perturbation. The topology of such states generically consists of (multiple) isolated periodic points and associated manifolds that, due to the 3D nature of the flow, materialise as surface–curve pairs. This is exemplified in Fig. 26(b) by the 2D stable (convoluted surface) and 1D unstable (convoluted curve) manifolds of an isolated periodic point (not shown) in the globally-chaotic state that ensues from disintegration of said spheroids due to significant fluid inertia ($Re = 100$) [139].

**Intermezzo: toroidal versus spheroidal “route to chaos”**

The particular “route to chaos” depends on the kind of LCSs constituting the baseline topology. The above findings imply that – from a dynamical perspective – two canonical cases can be distinguished: baseline topologies consisting of (i) tori and (ii) spheroids [7]. Disintegration of KAM tori in both steady and unsteady 3D flows often (including the previous examples) follows the quasi-2D route dictated by their equivalence with 2D unsteady systems: progressive breakdown of tori into chaos with increasing perturbation according to well-defined scenarios (Sec. 4.2). However, 3D systems may furthermore follow two essentially 3D “routes to chaos” associated with tori and spheroids. This is elaborated below.

**Tori** may under certain conditions develop local defects that enable “jumping” of tracers between tori and, in consequence global dispersion. Such defects (locally) break the link with 2D unsteady systems and thus cause essentially 3D behaviour (Sec. 4.6.2). The study [129] investigates this for the time-periodic flow inside the 3D annular region between two concentric spheres driven by their co-axial rotation about axis $\hat{r} = \cos(\alpha) \hat{e}_z + \sin(\alpha) \hat{e}_r$, with $\alpha$ alternating between $\alpha = 0$ and $\alpha = 1^\circ$. The base flow ($\alpha = 0$) consists (for sufficiently small $Re > 0$) of a primary ($\theta$-wise) circulation about the $z$-axis ($\vec{r} = \hat{e}_z$) and a weak secondary circulation in the $rz$-plane [143]. The corresponding streamlines describe two families of tori (with a dense poloidal winding) centred on the $z$-axis and symmetric about plane $z = 0$ following Fig. 27(a) (left), implying a flow topology qualitatively the same as the steady cylinder flow in Fig. 25(b). The time-periodic flow, due to the only minute reorientation of the base flow during rotation about the tilted axis ($\alpha = 1^\circ$), yields a stroboscopic map consisting of basically the same tori as the base flow (Fig. 27(a) left). The principal difference is the emergence of defects in the tori that enable said “jumping” as demonstrated in Fig. 27(a) (left) by the cross-sectional stroboscopic map of a single tracer. This results in a gradual global dispersion due to intermittent dynamics comprising of circulation along and jumping between tori at associated LCSs. Moreover, periodic points preclude double circulation.

**Spheroids** may under certain conditions develop local defects that enable “jumping” of tracers between spheroids and, in consequence global dispersion. Such defects (locally) break the link with 2D unsteady systems and thus cause essentially 3D behaviour (Sec. 4.6.2). The study [129] investigates this for the time-periodic flow inside the 3D annular region between two concentric spheres driven by their co-axial rotation about axis $\hat{r} = \cos(\alpha) \hat{e}_z + \sin(\alpha) \hat{e}_r$, with $\alpha$ alternating between $\alpha = 0$ and $\alpha = 1^\circ$. The base flow ($\alpha = 0$) consists (for sufficiently small $Re > 0$) of a primary ($\theta$-wise) circulation about the $z$-axis ($\vec{r} = \hat{e}_z$) and a weak secondary circulation in the $rz$-plane [143]. The corresponding streamlines describe two families of tori (with a dense poloidal winding) centred on the $z$-axis and symmetric about plane $z = 0$ following Fig. 27(a) (left), implying a flow topology qualitatively the same as the steady cylinder flow in Fig. 25(b). The time-periodic flow, due to the only minute reorientation of the base flow during rotation about the tilted axis ($\alpha = 1^\circ$), yields a stroboscopic map consisting of basically the same tori as the base flow (Fig. 27(a) left). The principal difference is the emergence of defects in the tori that enable said “jumping” as demonstrated in Fig. 27(a) (left) by the cross-sectional stroboscopic map of a single tracer. This results in a gradual global dispersion due to intermittent dynamics comprising of circulation along and jumping between tori at associated LCSs. Moreover, periodic points preclude double circulation.

16 This is a consequence of the domain topology. Brouwer’s well-known fixed-point theorem states that time-periodic flows in convex domains have (at least one) periodic point(s) following [6] [138]. Spheroids and 2D confined domains (as the lid-driven cavity) are convex domains and, by virtue of said theorem, both have flow topologies consisting of periodic points and
local defects (indicated by dashed semi-circles).

The mechanism causing the defects is the local synchronisation of the dynamics with the time-periodic forcing: the poloidal ($\phi$) and toroidal ($\varphi$) angles of certain tracers (in the local toroidal reference frame) evolve with rotations $\omega = \omega(x_0)$ and $\gamma = \gamma(x_0)$ relating via rational fractions, i.e.

$$\phi_p = \phi_0 + p\omega, \quad \varphi_p = \varphi_0 + p\gamma, \quad \omega/\gamma = m/n, \quad (13)$$

where $m$ and $n$ are integers. They describe trajectories that reconnect after $n$ toroidal windings. Tracers exhibiting such “resonance” prevent neighbouring trajectories from densely filling the entire torus and thus cause local defects (refer to Sec. 4.6.2 for the basic mechanisms). Here this occurs in surfaces termed “resonance sheets”; these sheets are unstable and under arbitrarily small perturbation disintegrate into localised chaotic zones that interrupt the poloidal circulation within tori and thus enable random jumps between them. This results in global transport following Fig. 27(a) (right) and is aptly termed “Resonance-Induced Dispersion” (RID) [129]. The actual jumps in RID are facilitated by localised versions of global manifolds similar to Fig. 26(b) corresponding with isolated periodic points (6) that remain from the disintegrated resonance sheets [130].

Circumstantial experimental evidence of RID is obtained by the dispersion properties of fluorescent tracer particles in convection cells (Sec. 3.5.2) [144]. Moreover, RID has a direct counterpart in 3D steady flows in global dispersion following Fig. 27(a) (right) enabled by local defects in tori induced by poloidal/toroidal evolutions

$$\phi(t) = \phi_0 + \omega t, \quad \varphi(t) = \varphi_0 + \gamma t, \quad (14)$$

with $\omega$ and $\gamma$ forming rational fractions following (13) [145]. The notion of resonance furthermore generalises to defects in tori caused by significant slow-down of Lagrangian motion [15]. This e.g. underlies the separatrices crossing in the ACEO-driven flows, droplets flows (Sec. 3.4.2) and convection cells (Sec. 3.5.2).

Spheroids may also develop local defects due to resonances yet fundamental disparities exist with tori on grounds of the different topology and dynamics. Key is the formation of periodic lines through coalescence of isolated periodic points (6) in the case of spheroids. These lines intersect the spheroids as illustrated in Fig. 27(b) (left) for the two-step forcing by the bottom wall in Fig. 26(a) (left). (Such points – and thus the resulting periodic lines – always exist on account of Brouwer’s fixed-point theorem. Tori, on the other hand, are generically devoid of periodic points, which sets them fundamentally apart from spheroids [139].) The impact of periodic lines on the response to weak perturbation depends fundamentally on the nature of the associated LCSs: chaotic seas (demarcated by manifolds of the individual points on hyperbolic line segments) survive as thin shells; islands (centered on individual points of elliptic line segments) coalesce into tubes. Resonance here ensues from the isolated degenerate periodic points (termed “parabolic points”) that partition the periodic line into elliptic and hyperbolic segments and (instead of causing local defects in otherwise complete tori as in RID) results in *truncation* of the entire tube. Shells and truncated tubes thus formed merge into intricate LCSs and this process is therefore termed “Resonance-Induced Merger” (RIM). RIM is also an essentially 3D phenomenon by, similar to RID, breaking the link with 2D unsteady systems (again refer to Sec. 4.6.2 for the basic mechanisms).

Fig. 27(b) (right) demonstrates RIM due to perturbation of the flow topology in Fig. 27(b) (left) by weak fluid inertia ($Re = 0.1$), with heavy and normal curves indicating ellipses called “averaging principle”, which assumes motion along trajectories that (i) causes tracer positions to densely fill them over time and (ii) is significantly larger than the drift induced by the perturbation [146]. These assumptions break down near resonances as well as in slow-motion regions and yield local motion in all coordinate directions in the order of the perturbation strength. This results in essentially 3D dynamics that is no longer restricted to tori. Thus slow motion is a generalisation of “true” resonances by (though not strictly precluding) strongly impeding the filling of entire trajectories and, inherently, tori formation. This is further explained in Sec. 4.6.2.
tic and hyperbolic line segments, respectively, separated by the isolated parabolic points (stars) that underlie resonance here [139,147]. Shown is the 3D stroboscopic map of a single tracer outlining an inner and outer shell connected by two tubes centred on the elliptic line segments. The LCS thus visualised (and formed by merger of shells and truncated tubes) is one member of a family of such LCSs that are arranged concentrically according to the truncated tubes. Hence RIM “creates” new LCSs from existing ones; RID, on the other hand, (partially) destroys LCSs. Moreover, RIM, unlike the random jumps in RID, is a predictable and systematic phenomenon in that families of truncated tubes form on elliptic line segments and merge with shells at tube ends at the well-defined positions of parabolic points. (Segmentation of periodic lines therefore is key to RIM; it cannot occur for single-type lines.) The observation of RIM in other flow configurations as well as direct isolation of periodic lines and tube segments by experimental stroboscopic maps using 3DPTV strongly suggests that RIM is a universal and robust phenomenon in flows with spheroidal LCSs and segmented periodic lines [148,149,125,150].

Important to note is that RIM, though creating rather than destroying LCSs, nonetheless is on a “route to chaos”. Said entities namely, instead of being strictly invariant, effectively constitute regions of prolonged (yet not indefinite) entrapment for (tens of) thousands of periods that in the long run admit global dispersion by accumulated transverse drifting. Moreover, incomplete shells may form that exhibit “leakage” of tracers into 3D chaotic zones [148, 139, 147].

The route to 3D chaos from spheroids, in stark contrast with tori, is (to the best of our knowledge) hardly explored in the literature. However, the above findings as well as the considerations below suggest that this scenario may be relevant specifically for unsteady container flows consisting of reoriented base flows.

**Unsteady flows revisited** Mixing of granular media in spherical tumblers is an industrial application that (under the premise of continuum-like flow) exhibits spheroidal LCSs and (signatures of) RIM [151]. The configuration is given schematically in Fig. 28(a) and consists of a time-periodic flow in a half-full sphere driven by alternate rotation about x and z-axes. Equal rotation rates yield a flow topology comprising of spheroids and periodic lines and, in consequence, 2D dynamics similar to that in the lid-driven cylinder flow in Fig. 26(a) (left). Fig. 28(b) demonstrates this by the computational stroboscopic map of multiple tracers (distinguished by colour) released within a given spheroid; green/blue curves are periodic lines consisting entirely of hyperbolic/elliptic points and intersect the spheroid at the chaotic sea and centre of the two opposite islands (outlined by circular orbits). Slight deviation in rotation rates triggers a similar response as weak fluid inertia in the cylinder flow: the chaotic sea forms a thin shell; islands of adjacent spheroids coalesce into tubes. Fig. 28(c) demonstrates former and latter by weak drifting transverse to the spheroid and incipient spiralling about the elliptic line (blue), respectively, resulting in a “fuzzier” stroboscopic map. These dynamics are clear signs of RIM and, though actual tube-shell merger as in Fig. 27(b) is not investigated in [151], thus strongly suggest that the spherical tumbler follows this route to 3D chaos upon perturbation by varying rotation rates. An important outstanding question is whether the periodic lines (in appropriate parameter regimes) admit the segmentation into elliptic and hyperbolic segments that is essential to RIM.

Unsteady transport phenomena as exemplified above are yet to find their way into analysis, design and technological development of other industrial applications. However, accomplishment of 3D global chaos as in Fig. 27(a) (right) by time-periodic agitation of industrial batch mixers may significantly boost performance compared to conventional steady stirring that (implicitly) relies on effectively 2D chaos due to breakdown of tori (Fig. 18 and Fig. 19). Novel concepts such as glass processing by time-periodic electro-magnetic stirring (Fig. 25(b)) take important first steps in this direction.

Micro-fluidic devices and emerging technologies that involve batch processing may particularly benefit from insights into unsteady Lagrangian transport and chaos due to the high degree of flexibility and control offered by micro-fluidic forcing techniques and domain shapes. Proper elec-
trod layouts and (time-periodic) activation in ACEO-driven flows e.g. enable systematic realignments of the steady “base flow” in Fig. 24(b) so as to create various flow and transport conditions in micro-channels similar to the cylinder flow (Fig. 26) including (in principle) deliberate processes such as RIM (Fig. 27(b)). Suspended magnetic nano-particles facilitate well-controlled body forcing reminiscent of the above electro-magnetic glass processing for basically any liquid and find first applications for batch mixing in microfluidic platforms for biomedical and diagnostic purposes [109, 112]. The latter employ time-periodic magnetic forcing to accomplish chaotic advection and incorporation of essentially 3D insights may enable further (technological) development of such devices. Moreover, this may enable new (diagnostic) functionalities by systematically creating and utilising LCSs for e.g. particle trapping and/or separation similar to Fig. 21.

3.3.3 Relevance beyond industry
Lagrangian (chaotic) transport in 3D container flows also play a central role in many (seemingly disconnected) non-industrial flow situations and thus have fundamental similarities with the systems considered above. This is, as for duct flows (Sec. 3.2.3), again illustrated by examples.

Aneurysms Aneurysms are balloon-like malformations of blood vessels that, upon rupturing, lead to internal bleeding with potentially lethal consequences. Transport of blood-borne substances is a key player in this process and in particular chaotic advection (induced by the abnormal shape of aneurysms) has been implicated in the promotion of cardiac diseases (Sec. 3.1 and 3.2.3). The typical flow in aortic aneurysms is visualised experimentally in Fig. 1(b) by 3D Lagrangian trajectories and consists of a vortical structure within the aneurysm (open arrows) driven by the passing aortic flow (closed arrows) [45]. Fig. 29(a) substantiates this generic structure by the 3D streamline portrait of the cyclic mean velocity field in a laboratory replica of a cerebral aneurysm constructed from 2D stereo-PIV measurements [152]. This exposes a similar vortical interior flow (dotted arrow) as well as the local in/outflow (solid/dashed arrow) at the attachment to the cerebral artery that drives the former. Thus the flow in the aneurysm resembles the above time-periodic lid-driven cylinder flow by consisting of a reorientation of a circulatory base flow akin to Fig. 23 forced at its boundary; a non-essential difference is that the flood flow varies continuously (instead of step-wise) during each cardiac cycle. This resemblance implies unsteady 3D (chaotic) transport and its exploitation offers a way to investigate and unravel this process in aneurysms so as to deepen insights into their origins and implicated vascular diseases as thrombosis and atherosclerosis (Sec. 3.1).

A further analogy exists in the larger configuration of aneurysm and blood vessel combined. This situation is comparable to the previous ACEO-driven flow in conjunction with a main throughflow, implying similar interactions between subregions of the vortical flow in the aneurysm and the main blood stream and, in consequence, similar hybrid behaviour including both duct-flow and container-flow char-
characteristics. The computational study by [153] investigates such 3D Lagrangian transport in the 3D time-periodic flow in a cerebral aneurysm induced by the cardiac cycle using a realistic geometry based on CT angiography. Fig. 29(b) shows typical Lagrangian trajectories (distinguished by colour) of three initially close fluid parcels released at the inlet of the single ingoing blood vessel. (The cross-sectional distribution at the inlet gives the residence time of a dense set of parcels released here.) The parcel dynamics nicely demonstrates (i) said hybrid behaviour of prolonged entrainment in the vortical structure inside the aneurysm during excursion from inlet to outlet and (ii) its essentially chaotic nature by the rapid separation of the trajectories. Chaotic trajectories in the above ACEO-driven flow exhibit exactly the same behaviour [128].

Fig. 29(b) exposes a further ramifications of chaotic advection specific for the aneurysm flow: deflection of trajectories into different outgoing blood vessels (labelled 1–3). Such random distribution over multiple branches is in fact an important mechanism for both local and global dispersion in (random) porous media (Sec. 3.5.2) including cardiovascular networks (Sec. 3.5.3).

**Lung alveoli** Flow and transport within the lungs, though physiologically entirely different, intimately relates to the above aneurysms. Air enters the lungs via the respiratory bronchioles, passes through the acinar ducts and ends in the terminal sacs (Fig. 30(a)). These airways become increasingly populated with hemispheroidal protrusions denoted “alveoli” that release air into the bloodstream. The internal flow is primarily driven by the passing ducular flow and the configuration thus resembles that in aneurysms and, in turn, the time-periodic lid-driven cylinder flow. Compare to this end the simulated instantaneous 3D streamline pattern inside an alveolus in Fig. 30(b) with Fig. 23 and Fig. 29(a). The similarity in flow configuration demonstrates interaction between alveolic and throughflow akin to that in aneurysms and, inherently, hybrid dynamics in the chaotic regions as demonstrated in Fig. 29(b).

Aerosol mixing and deposition in the alveoli is essential to the overall functioning of the lungs and motivated a range of studies on Lagrangian transport in the pulmonary acinus [154,155,156,30,157]. This gave the key insight that chaotic advection, in stark contrast with its adverse effect in aneurysms, is vital for the kinematic irreversibility necessary to achieve net aerosol transport during each breathing cycle. Fig. 30(c) experimentally validates stretching and folding – signifying chaotic advection – in the airways by visualisation the cross-sectional mixing pattern using two polymerised liquids of different colour (bar = 500 µm) (reproduced from [156]).

**Gastric digestion** A further physiological example of transport in 3D time-periodic container flows exists in the digestive process inside the stomach. The objective is efficient extraction of nutrients by the mixing and “kneading” of the gastric contents via peristaltic movement of the stomach wall through so-called “antral contraction waves”. These waves periodically compress and expand the stomach and its contents and thus set up a 3D time-periodic flow driven by movement at/of the boundary reminiscent of aneurysms and lung alveoli and, by the aforementioned analogy, 3D lid-driven cavity flows. The main differences with said examples are (i) that the stomach, though topologically again spheroidal, has a more complex shape (Fig. 31(a) and (ii) involves elaborate forcing by the entire boundary. This results in the formation of multiple families of vortical structures 

![Image 308x701 to 548x754]

![Image 429x568 to 547x677]
of varying extent and intensity as exemplified in Fig. 31(b) by typical instantaneous 3D streamlines [33]. The individual structures – particularly in the central part of the stomach – clearly resemble those in Fig. 23(c).

The highly non-trivial interplay of the vortical structures in the course of a period are likely to produce (at least local) chaotic advection and first exploratory studies on simplified systems indeed suggest this [161]. However, the contribution of advective transport to actual gastric mixing remains unclear yet is considered important for better understanding of this process [162]. Hence, this is a further problem that may benefit considerably from insights in 3D Lagrangian transport and analogies with the above systems.

Champagne tasting A perhaps surprising (though appealing) instance of 3D transport in container flows emerges in champagne tasting. Transport and mixing due to the internal flow driven by bubble formation upon pouring into the glass is namely believed key to the exhalation of flavours and aromas and thus the taste of the champagne [163].

Champagne glasses typically have engravings at the bottom as nucleation site for a central column of rising CO₂ bubbles as driver of a cross-sectional circulation in the rz-plane following Fig. 32 (top, left). This is visualised in Fig. 32 by vertical cross-sections of fluorescent dye in a coupe glass (bottom, left) and polymer particles in a flute (bottom, right). The vortical structures thus created are qualitatively similar yet their extent depends on the type of glass [163].

The 3D flow constitutes a vortex ring with a corresponding 3D flow topology reminiscent of that below the disk impeller in Fig. 18 yet with one fundamental difference. The impeller actively imposes uni-directional azimuthal flow and, by virtue of the resulting analogy with periodic duct flows, gives rise to KAM tori and associated chaotic regions (Sec. 3.3.2). The champagne flow, on the other hand, stems from passive internal forcing by the (approximately) axi-symmetric bubble column. Its small asymmetric fluctuations induce weak azimuthal flow without one preferential direction and, in consequence, continuous reversal in the course of time. Such dominant cross-sectional circulation in combination with weak azimuthal fluctuations are in fact key ingredients for resonance-induced defects in tori and, in consequence, RID as in Fig. 27(a) (right). RID namely ensues from weak perturbation of an idealised flow topology consisting of closed streamlines (Sec. 4.6.2). These considerations suggest that transport and mixing in the champagne glass may be highly non-trivial and of unexpected richness. In-depth Lagrangian analysis – with a particular focus on resonance phenomena – enables rigorous exploration of this.

Cilia-driven flows at coral reefs Transport in the cilia-driven flow at polyps in coral reefs shown Fig. 1(c), while, as for the ACEO-driven flow (Sec. 3.3.2), not immediately apparent, intimately relates to flows in containers. The cilia, though strictly in an open domain, namely create a boundary-driven flow in a confined region adjacent to the coral surface consisting of vortical structures comparable to that of the boundary-driven container flows shown in Fig. 23 and Fig. 24(b). This is demonstrated in Fig. 33 for typical flow patterns above coral explants obtained by 2DPTV using powdered coral food as tracer particles [47]. The 2DPTV measurements reveal arrays of vortices with alternating circulation (indicated by white arrows) involving flow speeds ranging from stagnant fluid (blue) to a maximum velocity of O(1 mm/s) (red). Note in particular the striking resemblance of the pair of counter-rotating vortices in Fig. 33 (right) with the ACEO-driven flow in Fig. 24(b). These analogies imply similar Lagrangian transport as in said container flows.

3.4 Flow in drops
3.4.1 General configuration

Neglecting two-phase effects as drop coalescence and breakup, the flow inside a drop is in a dynamical sense a subclass of flow in containers considered above. They namely both concern confined flows in (topologically) spheroidal do-
mains and the corresponding Lagrangian transport thus is subject to the same kinematic and topological constraints. The primary difference with container flows is that drops are bounded by a fluid-fluid interface instead of an (elastic) solid wall and thus offer functionalities (particularly in micro-fluidic systems) such as immersion in a bulk flow and external forcing by techniques unavailable to the former. Hence (potential) technological applications involving drops are entirely different from the above container flow and this motivates its distinction as a separate flow class.

### 3.4.2 Industrial and technological relevance

Lagrangian transport and mixing in drops is relevant to a range of micro-fluidic applications and emerging technologies. Two basic configurations can be distinguished:

- **Microfluidic networks** Here droplets propagate in an ambient flow through (interconnected) micro-channels and chambers and serve both as processing and transportation vessels. This principle finds application in micro-fluidic devices for batch processing of samples in e.g. pharmaceutical manufacturing or bio-engineering using droplets as chemical reactors or cell incubators so as to boost reaction rates, shield against contamination and/or provide protected environments [164, 165, 166, 167, 168, 169].

- **Digital microfluidic platforms** Here discrete droplets are trapped (and displaced) on solid substrates. Applications of this approach include processing and analysis of microscopic fluid quantities as e.g. chemical synthesis of drugs or immunoassays for clinical diagnostics of minute blood samples [170, 171, 172].

A great diversity of techniques is available for actuation and manipulation of the internal flow within microscopic drops. The intimate connection with container flows nonetheless means that droplet-based micro-fluidic systems, irrespective of the specific configuration, share fundamental properties with the former as well as among each other. This is elaborated below for several flow-forcing techniques.

#### Internal flow induced by ambient flow

Droplets immersed in a micro-channel flow driven by e.g. a pressure gradient are subject to shear stresses at the fluid-fluid interface (and generically travel slightly faster than the bulk) [6]. This yields a non-zero surfacial velocity in said interface and thus induces an internal flow within the drop akin to lid-driven cavities (Fig. 34). Straight channels, by virtue of constant droplet shape and propagation speed, compare to steady wall forcing and thus yield a steady global circulation with typically non-chaotic dynamics (Fig. 34(a)). Curved channels with a spatially periodic geometry, on the other hand, give rise to oscillating droplet shape and speed and, analogous to time-periodic wall forcing, result in a time-periodic internal flow that admits chaotic dynamics (Fig. 34(b)) [164].

Practical implementation of the above working principle is found in a droplet travelling through a serpentine micro-channel according to Fig. 35(a) [164, 173]. Fig. 35(b) visualises the internal mixing pattern by the evolution of a binary concentration distribution in the droplet midplane using fluorescent lifetime imaging microscopy and reveals the characteristic stretching and folding of chaotic advection. The corresponding simulated evolution is shown in Fig. 35(c) and closely agrees with the experiments.

In-depth experimental analysis of 3D flow and transport within immersed (propagating) microscopic drops is highly challenging and largely beyond current experimental methods. Hence its exploration to date primarily involves theoretical and computational studies. Two standard cases underlying such studies are: (i) buoyant drops driven by gravity; (ii) non-buoyant drops driven by viscous stresses at the fluid-fluid interface. The non-dimensional 3D Stokes flow inside droplets of unit radius is in the co-moving reference frame, with \( z \) the direction of travel, described analytically by

\[
\begin{align*}
  u_x &= zx, \\
  u_y &= zy, \\
  u_z &= 1 - 2r^2 + z^2,
\end{align*}
\]

(15)
which, by consisting of tori, is (dynamically) indeed equivalent to the above container flows. This overall implies similar Lagrangian dynamics and “routes to chaos”; the tori in Fig. 36(b) generically give way to a 3D steady topology of KAM tori and chaos as in e.g. Fig. 22 and Fig. 23 upon intensifying the perturbation of (15) and (16) [176, 174]. The specific behaviour may depend significantly on the relative strength of poloidal versus toroidal flow, though.

The study in [165] adopts base flows (15) and (16) to model 3D flow and Lagrangian transport within the traveling drop in the serpentine channel (Fig. 35). Flow (15) represents the straight channel (Fig. 34(a)) and, taking inevitable imperfections into account, yields a non-chaotic steady topology comprising of a single family of tori following Fig. 36(b) (left). The time-periodic flow in the curved channel (Fig. 34(b)) admits (in the presumed Stokes limit) construction from linear combinations of (15) and (16) and, by effectively switching continuously between the instantaneous base-flow topologies in Fig. 36(b) induces breakdown of tori and thus yields chaotic advection. Fig. 36(c) demonstrates this by the characteristic stretching-and-folding pattern emerging (for 3 typical situations) in the cross-section $x = 0$ of the 3D distribution of a tracer (white) initially occupying the lower droplet hemisphere [165].

Internal flow induced by electro-hydrodynamic forcing

A configuration intimately related to the above serpentine channel exists in a translating droplet in a straight channel subject to an axial electric field $\vec{E} = E \hat{e}_z$. The latter, given the dielectric properties of droplet and bulk fluids are sufficiently apart, induces forces in the fluid-fluid interface that set up a so-called “Taylor circulation” [177]. Its topology corresponds (for a stationary droplet) with Fig. 36(b) (right), where the black axis coincides with the direction of travel, meaning that electro-hydrodynamic forcing has a comparable effect as shear due to curvature in the serpentine channel. Droplets travelling in the z-direction retain this topology yet develop a $z$-wise asymmetry: the leading/trailing family of tori expands/diminishes and, inherently, the separatrix – spanned by the grey axes in Fig. 36(b) (right) – shifts towards the trailing side of the droplet [177].

Chaotic advection by destruction of the tori arrangement following Fig. 36(b) (right) can be achieved basically in two ways: (i) unsteady field strength $E(t)$ so as to change the position of said separatrix in time; (ii) misalignment between electric field $\vec{E}$ and translation direction so as to break the symmetries of the topology. The former is investigated for a falling droplet in a tank subject to a time-periodic electric field $\vec{E}(t) = E \sin(\omega t) \hat{e}_z$ in [178]. Fig. 37(a) visualises typical Lagrangian transport conditions by 2D temporal stroboscopic maps in the $xz$-plane of tracer particles according to simulations (top) and experiments (bottom). This reveals erratic wandering of tracers around surviving KAM tori (white patches in top panel) and thus signifies chaotic advection. The underlying mechanism intimately relates to RID (Sec. 3.3.2) and is elaborated below in the context of thermodiffusional forcing.

Chaos by misalignment is investigated in the
Steady misaligned forcing and exposed to an axial temperature gradient, thermo-capillary effects in droplets immersed in a bulk fluid lead to internal flow induced by thermo-capillary forcing. Field via alternating angles \( \alpha \) demonstrated in [180] by a time-periodic reorientation of the above stationarity, i.e. the imaginary plane in Fig. 37(b) between the vortical structures at the trailing (top) and leading (bottom) droplet side. The slight tilt emanates from the misaligned electric field (\( \alpha \neq 0 \)).

Reorientation of the electric field enables similar accomplishment of chaos for a stationary droplet. This is demonstrated in [180] by a time-periodic reorientation of the above field via alternating angles \( \alpha = 0 \) and \( \alpha = \alpha_w \).

Internal flow induced by thermo-capillary forcing
Thermo-capillary effects in droplets immersed in a bulk fluid and exposed to an axial temperature gradient

\[
\hat{\nabla} T = (\kappa_0 + \kappa_1 \z) \hat{e}_z, \tag{17}
\]

The onset of 3D chaotic advection in the present steady flow stems from defects that develop in both tori families near the separatrix for weak perturbation of the non-chaotic baseline [182]. This results in drifting and gradual chaotic dispersion reminiscent of RID in unsteady container flows (Fig. 27(a)) for all streamlines that encounter these defects. Essential difference with RID – and its steady counterpart [145] – is that defects here emanate from significant slowdown of Lagrangian motion near the separatrix instead of “true” resonance following [14]. (Slow motion is a generalisation of resonance in that this affects the dynamics in a similar way; see Sec. 3.3.2 and Sec. 4.6.2). The impact is therefore restricted to the “outer” tori close to the separatrix; the “inner” tori survive unscathed. Hence the dispersion thus induced is restricted to a chaotic region surrounding the latter tori. Stronger perturbation overrides these phenomena and results in the “conventional” breakdown of tori and (further) proliferation of the chaotic region (Sec. 4.6).

\( \kappa_0 \) and \( \kappa_1 \) parameterising its uniform and \( z \)-wise linear contributions, respectively, may induce forces in the fluid-fluid interface akin to the above electro-hydrodynamic phenomena and thus drive an internal circulation [181]. A uniform axial temperature gradient (\( \kappa_0 > 0, \kappa_1 = 0 \)) causes droplet motion in \( +z \)-direction and yields an internal circulation and corresponding topology following (15) and Fig. 36(b) (left), respectively. Chaotic advection again requires destruction of tori and can here be achieved by imposition of a non-uniform temperature gradient (\( \kappa_0 > 0, \kappa_1 \neq 0 \)) and a perpendicular vortical motion due to shearing by the ambient flow. The non-uniform gradient yields an entirely intact topology following Fig. 36(b) (right) yet with the same \( z \)-wise asymmetry (and shift of the separatrix) as the above electro-hydrodynamic case; superposition of the shear initiates tori breakdown and results in chaotic streamlines wandering around surviving KAM tori in the same was as shown in Fig. 37(b) [181, 182] [20].

Fig. 37. Lagrangian transport in \( z \)-wise falling droplet subject to electro-hydrodynamic forcing: (a) stroboscopic map in \( xz \)-plane for time-periodic electric field along \( z \)-axis as simulated (top) and visualised by tracers (bottom) (adapted from [178]); (b) 3D streamline for steady electric field misaligned with \( z \)-axis (adapted from [179]).

with coefficients \( \kappa_0 \) and \( \kappa_1 \) parameterising its uniform and \( z \)-wise linear contributions, respectively, may induce forces in the fluid-fluid interface akin to the above electro-hydrodynamic phenomena and thus drive an internal circulation [181]. A uniform axial temperature gradient (\( \kappa_0 > 0, \kappa_1 = 0 \)) causes droplet motion in \( +z \)-direction and yields an internal circulation and corresponding topology following (15) and Fig. 36(b) (left), respectively. Chaotic advection again requires destruction of tori and can here be achieved by imposition of a non-uniform temperature gradient (\( \kappa_0 > 0, \kappa_1 \neq 0 \)) and a perpendicular vortical motion due to shearing by the ambient flow. The non-uniform gradient yields an entirely intact topology following Fig. 36(b) (right) yet with the same \( z \)-wise asymmetry (and shift of the separatrix) as the above electro-hydrodynamic case; superposition of the shear initiates tori breakdown and results in chaotic streamlines wandering around surviving KAM tori in the same was as shown in Fig. 37(b) [181, 182] [20].
Dispersion due to local defects in LCSs of 3D steady flows has first been examined in \cite{175} and denoted “trans-adiabatic drift”. Here significant slow-down of Lagrangian motion near certain isolated stagnation points caused this phenomenon. Analogous flow structures strongly suggest that basically the same mechanisms are at play in the above electro-hydrodynamically forced flow \cite{179,183}. Hence the chaotic dynamics in Fig. \cite{37} are representative of the weakly-perturbed state for both electro-hydrodynamic and thermo-capillary forcing. Moreover, comparable baseline topologies implicate this mechanism also in the separatrix crossing in convection cells (Sec. 3.5.2).

Internal flow induced by electro-wetting In contrast to forcing in microfluidic networks, electro-wetting is a promising forcing technique for droplets in digital microfluidic platforms and relies on the dependence of the contact angle between droplet surface and substrate on the electric potential between former and latter. This enables actuation of an internal time-periodic flow through controlled oscillation of the droplet shape by variation of the contact angle via an AC voltage applied between an electrode at the droplet pole and the substrate \cite{184,185}.

Fig. \ref{fig:38}(a) visualises the 3D Lagrangian transport in a droplet flow driven by electro-wetting by temporal stroboscopic maps of tracer particles in a vertical cross-section through the axis (left) and a horizontal cross-section near the substrate (right) \cite{185}. This enables a global toroidal circulation about the vertical droplet axis, directed downwards in the centre and radially outward at the bottom, and implies a flow topology that is a perturbed state of a non-chaotic baseline following Fig. \ref{fig:36}(b) (left). (The horizontal black axis in the latter coincides with the vertical droplet axis.) The corresponding transport is visualised in Fig. \ref{fig:38}(b) by the evolution (from left to right) of fluorescent dye (green) and the symmetric counter-rotating “swirls” in the middle snapshot strongly suggest a topology that, similar to Fig. \ref{fig:18}, contains sizeable KAM tori. Hence, contrary to the findings in \cite{185}, chaotic advection seems to occur only locally, namely in the surroundings of said tori. Moreover, dominant poloidal flow in conjunction with weak (fluctuating) toroidal flow closely resembles the situation in the champagne glass in Fig. \ref{fig:32} and thus suggests dynamics that, by considerations given before, may likely involve resonance phenomena.

Internal flow induced by acoustic streaming A relatively novel forcing technique for droplet flow in digital platforms employs 3D streaming induced by Rayleigh surface acoustic waves (SAWs) impinging on the surface \cite{186}. Fig. \ref{fig:39} shows a typical internal streamline pattern driven by a SAW (red arrow), obtained by simulations (panel a) and experimental visualisation (panel b), and reveals two adjacent toroidal circulations about a common centre of rotation (red dashed curve). This implies (on grounds of symmetry) a baseline topology similar to Fig. \ref{fig:36}(b) (right) consisting of two families of tori, where the horizontal black axis coincides with the red dashed curve in Fig. \ref{fig:39}(a) suggesting behaviour comparable to the above flows driven by electro-hydrodynamic and thermo-capillary forcing (Fig. \ref{fig:37}). However, in contrast with the latter, the acoustic streaming exhibits a significant toroidal circulation (i.e. about the red dashed curve in Fig. \ref{fig:39}), implying a 3D flow topology comparable to the steady inertial cylinder flow in Fig. \ref{fig:23} two symmetric families of KAM tori surrounded by chaos. Thus here chaos likely results from the conventional breakdown of tori associated with a predominantly toroidal motion instead of the resonance-induced scenario for the (mainly poloidal) flow driven by electro-hydrodynamic or thermo-capillary forcing.

Integrity of immersed drops Droplet-based microfluidic networks assume intact droplets and absence of transport between interior and ambient flow. However, droplets may exhibit leakage or even break-up due to e.g. excessive shear stresses and thus compromise the functionality of microfluidic devices \cite{187,188}. The fluid-fluid interface that bounds immersed droplets is a material surface and thus constitutes an LCS in its own right. Hence the Lagrangian dynamics of the interface is crucial to the integrity and stability of droplets. Consider to this end the response of a buoyant droplet following Fig. \ref{fig:36}(b) to perturbation of the ambient flow by an unsteady strain investigated computationally in \cite{189}. Fig. \ref{fig:40} shows the proliferation and interaction of the stable (red) and unstable (green) manifold of two so-called “distinguished hyperbolic trajectories” (DHTs) emerging from the disintegration of the (initially) spherical interface. DHTs and their manifolds are in essence the counterparts in aperiodic flows to hyperbolic periodic points and manifolds in time-periodic flows and thus dictate (chaotic) transport in basically the same way as illustrated in Fig. \ref{fig:5} (Sec. 4.6.1) \cite{190}. Here these LCSs determine the intricate 3D detrainment and entrainment of droplet and ambient fluid, respectively, that ensues from the droplet instability.
3.4.3 Relevance beyond industry

Discussed below are instances of 3D Lagrangian (chaotic) transport in non-industrial flows that closely relate to flows in drops. The aim, as for duct (Sec. 3.2.3) and container (Sec. 3.3.3) flows, is again to expose similarities and analogies between (seemingly) different problems.

Cytoplasmic flow and transport Cytoplasmic flow and transport inside cells of living organisms is (regarding domain topology) the physiological analogy to the above drops. An important forcing mechanism is so-called “cytoplasmic streaming”, that is, internal circulation within cells by flow actuation at the cell boundary via “molecular motors” in a manner akin to shear-driven flow in droplets. Fig. 41(a) illustrates this for the cytoplasmic circulation inside a drosophila oocyte for the purpose of mixing of yolk granules as nourishment during cell development [191]. The corresponding cross-sectional circulatory velocity field in the (approximate) midplane obtained by 2DPIV is given in Fig. 41(b). Mixing is visualised in Fig. 41(c) by the motion and dispersion of yolk granules in the oocyte (indicated by dashed outline) tagged via a tracer injected at the yellow arrow (numbers indicate time instance in minutes). The tagged yolk initially migrates as a compact patch, subsequently moves away from the midplane and reappears in a well-mixed state. These observations in conjunction with the domain shape strongly suggest a baseline topology following Fig. 36(b) (right), where the grey axes span the separating midplane. The appreciable circulation in Fig. 41(b) in turn, suggests a flow resembling the droplet flow driven by acoustic streaming (Fig. 39). This, by analogy, implies a 3D flow topology comparable to the steady inertial cylinder flow in Fig. 23 and, inherently, chaos due to conventional tori breakdown.

A forcing mechanism employed specifically by motile cells is flow induced via deformation during migration in an ambient flow [34]. This suggests cytoplasmic flow and transport comparable to droplets propagating a serpentine channel (Fig. 35).

Delivery and mixing of drugs A further physiological counterpart of Lagrangian transport in drops is found in the delivery and mixing of drugs in the vitreous chamber of the eye (Fig. 42(a)). Intravitreal drug delivery namely is a common treatment for retinal diseases and advective transport by the internal flow induced through so-called “saccades” (rapid eye rotations during redirection of the visual axis) are considered key to this process. The study by [192] investigates the advection characteristics via reconstruction of the 3D time-periodic flow field in an in vitro physical model of the vitreous chamber from cross-sectional 2DPIV measurements. Lagrangian transport analyses by numerical particle tracking using the experimental velocity field reveal two toroidal circulations about the vertical axis and approximately symmetric about the midplane between upper and lower hemisphere. Fig. 42(b) gives typical trajectories in the upper hemisphere \( z \geq 0 \) (distinguished by colour) and demonstrates that said circulations consist of a predominantly poloidal circulation and a weak toroidal drift. This implies a baseline topology
Fig. 41. Cytoplasmic flow and mixing in drosophila oocyte driven by cytoplasmic streaming as physiological analogy to droplet flow: (a) yolk mixing by cytoplasmic streaming; (b) cross-sectional velocity (red vectors) visualised experimentally by 2DPIV; (c) yolk mixing visualised experimentally by motion and dispersion of yolk granules in oocyte (dashed outline) tagged by tracer injected at yellow arrow (numbers indicate time instance in minutes). Adapted from [191].

Fig. 42. Drug delivery in vitreous chamber of eye as physiological analogy to droplet flow: (a) cross-section of eye; (b) simulated Lagrangian trajectories in upper hemisphere (distinguished by colour) using experimental 3D time-periodic velocity field constructed from cross-sectional in vitro 2DPIV. Adapted from [192].

following Fig. 36(b) (right), where the grey axes span the xy-plane in Fig. 42(b). The prevailing poloidal motion suggests non-trivial Lagrangian dynamics due to separatrix crossing similar to the electro-hydrodynamic (and thermo-capillary) droplet flow in Fig. 37 and, given time-periodicity, likely also RID as in Fig. 27(a) (right).

3.5 Flow in webs
3.5.1 General configuration

A final class of systems relevant in the present scope concerns flows in a domain with an internal partitioning into interconnected elementary cells that admit exchange of fluid and scalars. This may involve a “soft” partitioning by a mesh of stream surfaces or material interfaces as e.g. in convection cells of buoyancy-driven flows (Fig. 43(a)) or a “hard” partitioning by a solid matrix enclosing interconnected cavities as e.g. in porous media (Fig. 43(b)). The Lagrangian transport by flows in such “webs” of interconnected cells exhibits, on grounds of the domain structure, characteristics of both periodic duct flows (transport along cells) and container flows (transport within cells). Hence flows in webs define in essence a hybrid class incorporating features of the systems considered in Sec. 3.2–Sec. 3.4.

3.5.2 Industrial and technological relevance

The examples of flows in webs shown in Fig. 43 in fact constitute two important configurations in the context of technological applications:

- **Convection cells** Lagrangian transport by laminar flow in convection cells is relevant to materials processing and fabrication involving coatings and films [195, 196, 197, 198, 199, 200, 201, 202] as well as for emerging technologies as liquid metal batteries [203, 204, 205].

- **Porous media** Lagrangian transport in porous media is key to both natural and engineered environments. The former primarily concerns subsurface flows involved in e.g. hydrology, enhanced oil and heat recovery and aquifer heat-storage systems [20, 21, 22, 23, 24, 25]. Transport in engineered porous media is at the heart of e.g. thermo-chemical heat-storage systems, micro-fluidic devices consisting of micro-vascular networks and compact heat exchangers based on metal foams [206, 207, 208, 209, 210].

The general flow and transport characteristics of both configurations are discussed and illustrated below using similarities and analogies with the above flow classes. Their manifestation and role in practical applications are subsequently exemplified by way of representative cases.

**Convection cells: generic characteristics** Fluid confined by a hot bottom wall and cold top wall tends to rise and descend near former and latter, respectively, due to buoyancy
and (upon overcoming viscous friction) thus sets up a pattern of circulations. This phenomenon is known as Rayleigh-Bénard convection and the vortical structures thus created, commonly denoted convection (or Bénard) cells, form regular patterns in the present regime of laminar flows \[^{[211]}\]. Thin fluid films on a hot horizontal wall and with a free top surface behave essentially the same; here circulations emerge from an interplay of buoyancy and surface tension and result in Bénard-Marangoni convection. Both Rayleigh-Bénard and Bénard-Marangoni convection yield (under 3D laminar conditions) hexagonal patterns of convection cells as exposed in Fig. \[^{33(a)}\] by flow visualisation in the free surface of a silicon oil layer using aluminium tracer particles \[^{[193]}\].

Fig. \[^{44(a)}\] schematically gives the 3D streamline pattern in two neighbouring hexagons consisting (within each cell) of closed streamlines within vertical cross-sections (surface “b” in right cell) centered around the hexagon axis (line “d” in left cell) \[^{[212]}\]. Thus the flow topology is equivalent to that in the droplet in Fig. \[^{36(a)}\] (left), where the horizontal black axis corresponds with the vertical hexagon axis. However, such topologies, for reasons given before, are singular limits; the real cell-wise topology emerges for arbitrarily small perturbations and comprises of a family of tori centered on the (hexagon) axis similar to Fig. \[^{36(b)}\] (left).

Flow topologies consisting of complete tori admit inter-cellular transport across separatrices (e.g. plane “a” in Fig. \[^{44(a)}\]) only by diffusion. Fig. \[^{44(b)}\] experimentally visualises such diffusion-enabled global transport by the spreading of dye (red) in silicon oil across the “honeycomb” demarcated by the cell interfaces (bright pattern) \[^{[213]}\]. The kinematic analogy with electro-hydrodynamic and thermocapillary droplet flows (i.e. similar baseline topology and dominant poloidal circulation) strongly suggests that, upon weak perturbation, the tori in convection cells generically develop defects by the same resonance phenomena \[^{[21]}\]. This facilitates crossing of separatrices following Fig. \[^{37}\] and thus enables advective inter-cellular transport as demonstrated in Fig. \[^{44(c)}\] by a typical simulated 3D trajectory of a tracer released at position \(P_t\) in a weakly-perturbed flow \[^{[214]}\].

Experimental evidence of such resonance-induced inter-cellular transport is given in Fig. \[^{44(d)}\] by the dispersion of fluorescent tracer particles in an array of vortices set up in sulfuric acid by MHD forcing. (Each vortex corresponds with a hexagonal convection cell in Fig. \[^{44(a)}\] in top view.) Shown is the initial (top) and final (bottom) distribution of tracers released in one vortex (bright) in the unperturbed flow driven by steady MHD forcing (left) versus the perturbed flow by additional time-periodic forcing (right) \[^{[144]}\]. This reveals a dramatic difference. Tracers in the unperturbed flow remain trapped in the original vortex; tracers in the perturbed flow, on the other hand, exhibit global dispersion due to the resonance-induced separatrix crossing. The essential role of time-periodicity in this process implies that here (in particular) resonance according to \[^{[13]}\] – and thus RID fol-

\[^{[21]}\] That is, due to “true” resonance by synchronised poloidal/rotational rotations following \[^{[13]}\] and \[^{[14]}\] and/or its generalisation, viz. significant local slow-down of Lagrangian motion (Sec. \[^{3.3.2}\] and Sec. \[^{4.6.2}\].
drying polymer film on a glass substrate in Fig. 45(a) by shadowgraphy [196] and “orange peel” textures, as shown in Fig. 45(b) for a drying paint film on stainless steel [197]. Fundamental insights into 3D Lagrangian transport in convection cells may deepen the understanding of these adverse effects and offer ways to mitigate or eliminate them. The global advective transport across convection cells in Fig. 44(d) (right) is systematically induced by weak time-periodic perturbation of the flow using a frequency resonant with the typical circulation frequencies. This means that, given the particular nature of the perturbation is immaterial for small magnitudes, basically any perturbation during (industrial) fabrication and application of coatings – e.g. weak vibrations attuned to the natural frequencies – may promote global transport as in Fig. 44(d) (right) and thus counteract the convection cells.

Cell formation due to Bénard-Marangoni convection may, rather than being an undesired phenomenon, also find utilisation in materials technology for the fabrication of textured coatings and films [198, 199, 200, 201]. Certain micro-structured polymer films e.g. rely on the formation of so-called “breath figures” due to the condensation of moisture (contained in air) on cold solid or liquid surfaces. Running moist air over a polymer solution leads to the formation of water droplets on the liquid surface that (while sinking) congregate into hexagonal patterns due to their Lagrangian transport by the convection cells and, in consequence, the micro-structure as visualised in Fig. 46(b) by microscopy. The subsequent evaporation of solvent and water during solidification of the polymer solution leaves an imprint of the entrained water droplets as pores in the shape of the convection cells in the solid polymer matrix. Fig. 46(a) demonstrates this principle for the honeycomb porous micro-structure thus created in a thin polymer film on a glass substrate visualised by scanning electron imaging [200]. Lagrangian transport in the convection cells evidently is crucial to this fabrication technique and utilisation of fundamental insights – to e.g. actively manipulate droplet migration and congregation – may enable its further development.

An approach intimately related to the above has been proposed for the micro-structuring of photonic crystals, which is key to the optical properties and, inherently, the performance of solar cells [202]. Here convection cells due to Bénard–Marangoni convection in a liquid perovskite film on a heated substrate determine the advective solvent transport during solidification and thus “assemble” micro-structures in the crystals as visualised in Fig. 46(b) by microscopy. The convection cells and, in consequence, the micro-structure admit manipulation by the substrate temperature and solution concentration. Further development of this process may also greatly benefit from exploitation of fundamental insights into 3D Lagrangian transport in convection cells.

**Porous media: generic characteristics** Porous media typically consist of a web of pores with random orientation and connectivity and throughflows, in consequence, spread transverse to the main flow direction. Fig. 47(a) demonstrates this by the xz-wise spreading of 3D streamlines in a steady z-wise throughput in such a medium described by tracer particles (distinguished by colour) released on a line in a single pore at the inlet $z = 0$ [215, 216]. The global transverse particle dispersion is dictated by the 3D structure of the pore network and, given its random composition, converges on a Gaussian distribution with progressing stream-wise position $z$.

The local flow through consecutive pores undergoes repeated splitting and recombination similar as in partitioned duct flows (Sec. 3.2). A fundamental difference between these systems and porous media is that the random (instead of systematic) reorientation in the latter generically results in chaotic advection at the pore level [215]. This is demonstrated in Fig. 47(a) (inset) by the typical evolution of the transverse mixing pattern within pores (of circular cross-section) with stream-wise position $z_n$ (starting from inlet $n = 1$), revealing the characteristic stretching and folding that signifies chaotic advection. Note the particular resemblance of the pore-scale mixing pattern with the patterns in the Quatro and serpentine mixers in Fig. 12(a) and Fig. 13(c) respectively, which also come about by splitting and recombination.

The LCSs associated with pore-scale chaotic advection in 3D steady flows are the manifolds emerging from stagna-
tion points $\mathbf{\xi} = 0$ of the skin-friction field

$$\mathbf{\tau} = \mathbf{n} \cdot \mathbf{V} \mathbf{u} = \left. \frac{\partial \mathbf{u}}{\partial \mathbf{n}} \right|_\Gamma,$$  \hspace{1cm} (18)

on the no-slip pore wall $\Gamma$ with outward normal $\mathbf{n}$ \cite{[215]}. Fig. 47(b) gives typical 1D and 2D manifolds emanating from such points in a porous matrix consisting of disordered spheres (flow from right to left) \cite{[217]}. These entities are the direct counterpart to manifolds of internal stagnation or periodic points shown in e.g. Fig. 26(b) and the 2D manifolds generically act as separatrices between e.g. circulation zones or the diverging streams of impinging jets \cite{[218]}. Here the 2D manifolds of adjacent pores, ensuing from impingement and flow separation, interact transversally in a manner akin to the stable (blue) and unstable (red) manifolds in Fig. 5(a) and thus accomplish pore-scale chaotic advection.

Global transverse dispersion as in Fig. 47(a) originates from the random distribution of trajectories over pores comparable to the downstream distribution over multiple blood vessels in the aneurysm flow in Fig. 29(b) and depends primarily on the network geometry. Pore-scale chaos enhances transverse stretching of fluid elements and thus promotes their split-up during change-over between consecutive pores, but its effect on global transverse dispersion, though not yet conclusively established, seems marginal \cite{[217]}. Global stream-wise dispersion, on the other hand, is significantly affected by pore-scale chaos. It causes streamlines to “sample” the entire pore cross-section and the full pore-scale velocity distribution, meaning that particles effectively migrate downstream by the cross-wise averaged velocity, as opposed to the local velocity in the Poiseuille-like throughflows in 2D steady systems. This chaos-induced averaging of the particle velocity tends to homogenise the pore-wise residence time and thus retards stream-wise particle dispersion – diminishing stream-wise “diffusion” of the global particle concentration – relative to 2D porous media devoid of pore-scale chaos \cite{[216,217,222]}

The above findings are relevant to randomly-structured porous media yet equally non-trivial Lagrangian transport is anticipated for 3D steady throughflows in case of structure imparted by a natural heterogeneity (e.g. anisotropy in fractured media) or engineered by design. This reinforces the link with periodic duct flows (which rely on structural re-orientation) and thus likely yields pore-scale flow topologies where, similar to Fig. 7(a) chaotic regions coexist with LCSs as separatrices or stream tubes. Such intricate topologies are expected to cause fundamental departures of the global transport in 3D porous media from their 2D counterparts in a comparable way as before. However, essentially 3D pore-scale Lagrangian transport and its upscaling to global transport (as well as the interplay with diffusion) remain largely open problems \cite{[217,194]}. This concerns in particular the proper upscaling of microscopic pore-scale behaviour to the mesoscopic level of “representative elementary volumes” (REVs). REVs are the smallest elementary volumes in a macroscopic system admitting averaged representations of flow and transport over both the fluid domain and porous matrix \cite{[219]}. Such averaged representations make complex problems computationally tractable and thus are of great importance for practical analyses. This is elaborated below.

Natural porous media: subsurface transport

Natural porous layers in the subsurface typically possess a random structure and the above findings on both 3D pore-scale and 3D global transport demonstrated in Fig. 47(a) have important ramifications for practical situations. Consider for example the contamination of soil and groundwater via the spreading of a methane plume due to leakage during enhanced oil recovery. Fig. 48 shows a typical instantaneous 3D methane gas plume (via a concentration iso-surface) visualised by ground-penetrating radar in a field experiment in an actual aquifer following controlled injection at the indicated “shallow” and “deep” positions and spreading by the groundwater (“GW”) flow \cite{[220]}. The analogy with the configuration

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22 The same chaos-induced averaging occurs in periodic duct flows and, besides homogenisation of particle velocities, furthermore promotes stream-wise throughflow of all particles, or equivalently, suppresses particle entrapment in e.g. localised recirculation zones \cite{[60]}.
in Fig. 47(a) strongly suggests similar 3D transport characteristics: pore-scale chaotic advection and, in consequence, suppressed downstream diffusion of the global plume front.

Crucial for large-scale practical problems such as the plume spreading in Fig. 48 are tractable (computational) models for the global flow and transport. Full prediction and simulation of pore-scale behaviour namely is (i) physically impossible due to a lack of detailed microscopic information and, even if available, (ii) prohibitively expensive. The concept of REVs for description of mesoscopic flow and transport introduced before is a common approach towards such macroscopic models [219]. (The entire sample in Fig. 43(a) constitutes an REV typical of sands or bead packs.) The standard Darcy model is a well-known REV-based macroscopic model and assumes that the REV-averaged volumetric flux density \( \dot{q} \) (commonly denoted as the “Darcy velocity”) is proportional to the driving pressure gradient following

\[
\vec{q} = -\frac{\kappa}{\mu} \vec{\nabla} p, \tag{19}
\]

with \( \kappa \) and \( \mu \) the permeability and fluid viscosity, respectively, at the mesoscopic REV scale [219]. The average pore-scale velocity \( \vec{v} \) is given by the Dupuit-Forchheimer relation \( \vec{v} = \dot{q}/\phi \), with \( 0 < \phi < 1 \) the porosity, and e.g. corresponds with the mean stream-wise (“GW”) plume flow in Fig. 48.

The transverse and stream-wise spreading (of e.g. the plume in Fig. 48) by the dispersion of particles in the porous matrix is in conventional (Darcy-type) REV models incorporated via Fickian diffusion. This expands the governing equations for the particle motion to an advection-diffusion equation of the form (1), with \( C \) representing the particle “concentration” [219]. However, such upscaling from pore-scale to REV-scale does not account for phenomena as pore-scale chaos and its impact on global dispersion thus likely produces incorrect predictions of e.g. the plume spreading in Fig. 48 which may have significant practical consequences. Hence development of faithful REV models that adequately incorporate non-trivial 3D pore-scale transport is absolutely critical for reliable analyses of practical problems in complex porous media [217,219].

Subsurface REV models, besides mere analysis and prediction of transport in large-scale systems, furthermore enable utilisation of Lagrangian concepts for enhanced subsurface transport. This has great potential for a range of applications including enhanced geothermal systems [20,21], in situ recovery of minerals or oil [22,23] and groundwater remediation [24,25]. LCSs associated with the average pore-scale velocity \( \vec{v} = \dot{q}/\phi \), with \( \dot{q} \) according to (19), determine the global subsurface transport characteristics and admit manipulation by the pumping schemes for the injection and extraction wells that must be used in such applications to drive the subsurface flow. Thus purposeful conditions can (in principle) be systematically accomplished: global chaotic advection for efficient distribution of e.g. leaching solutions for minerals extraction or creation of large-scale islands as in situ processing zones for groundwater remediation. First exploratory studies on 2D reservoirs serve as promising “proof of principle” of this Lagrangian approach and may pave the way to new technologies for subsurface flow engineering [221,222,223,224,225,226,227].

**Engineered porous media: transport enhancement**

Engineering porous media with a given structure enables systematic manipulation of the (pore-scale) flow and transport and e.g. finds application in engineered 3D microvascular networks for mixing purposes. These configurations in essence expand on inline partitioned-duct mixers (Sec. 3.2.2) by progressive splitting and recombination of multiple fluid streams in a network of interconnected ducts to achieve “pore-scale” chaos akin to that in random porous media (Fig. 47(a)). Fig. 49(a) schematically demonstrates this by the interaction between two incoming fluid streams (red and green) and their mixing into a homogeneous outgoing fluid stream (yellow) during progression through a micro-vascular network fabricated of cylindrical duct segments [207]. Fig. 49(b) gives an experimental visualisation of the actual process using fluorescent dye.

The above example concerns a duct connectivity such that two incoming streams combine into one outgoing stream as per Fig. 49(a); this renders each such connection within the porous matrix analogous to the T-shaped micro-mixer in Fig. 14 yet with a more complicated duct geometry. However, the same duct architecture admits more elaborate connectivity tailored to attain different transport characteristics as e.g. simultaneous “pore-scale” chaos and global transverse distribution as in Fig. 47(a) or a certain heterogeneity in global dispersion properties.

Engineered porous media may furthermore aim at enhancing scalar transfer between flow and porous matrix. An important application exists in compact heat exchangers for heating/cooling of fluid flows based on metal foams (Fig. 50(a)). The metal foam realises the favourable attributes of a heat exchanger in a small volume: high interfacial area between solid and fluid; high ther-
3.5.3 Relevance beyond industry

Plate tectonics Convection cells set up in the mantle of the Earth (consisting of viscous molten rock) by its hot inner core are a major driving mechanism behind plate tectonics in the lithosphere (i.e. the outer crust) \(^{228,35}\). The associated up- and downwellings by hot and cold thermal plumes, respectively, namely cause the formation of ridges and trenches (via so-called “subduction”) due to viscous friction exerted on the crust in a way as illustrated in Fig. 51(a) and thus contribute to shaping the topography of the Earth’s surface. This configuration is analogous to Bénard-Marangoni convection in a liquid film by concerning a fluid layer inside a 3D spherical shell as in Fig. 27(a) (left) that is heated from below by the hot inner sphere and subject to lithosphere-induced surface stresses at the outer sphere. Hence the system tends to form a pattern of convection cells akin to Fig. 45 with corresponding up- and downwellings due to thermal plumes centred on lines “d” and “c”, respectively, in Fig. 44(a). However, different (material) compositions of oceanic versus continental lithospheres and complex mantle rheologies significantly complicate this process, resulting in highly irregular patterns of up- and downwellings as illustrated in Fig. 51(b) by level sets of “low” (blue) and “high” (yellow) temperatures, respectively \(^{229}\). This irregularity suggests comparably disordered surfacial patterns (i.e. in the lithosphere) tending to that of drying paint in Fig. 45(b).

The computational study by \(^{230}\), modeling mantle and lithosphere as a single continuum with an appropriate rheology, reveals that crust deformation is essential to the single-sided subduction (i.e. one tectonic plate sliding below another as in Fig. 51(a)) that in reality occurs at downwellings. Fig. 51(c) demonstrates this process by a cross-section of the temperature field, representing (via its strong correlation with the viscosity) the material composition, where low (blue) and high (red) temperature corresponds with crust and mantle, respectively. This reveals the downward intrusion of a high-viscosity “plume” of crust material (blue) into the mantle (red) at the contact line (position 1) of two converging plates driven by the convective circulation. The corresponding velocity (black arrows) exposes a localised circulation zone to the left of the intruding plume, meaning that the latter primarily consists of material stemming from the right plate, signifying its single-sided subduction below the left plate.

Entrainment and transport of crust material into the mantle is central to the subduction and is, by its essentially Lagrangian nature, determined by the 3D flow topology. This, by analogy with convection cells, implies a scenario as before: breakdown of separatrices as plane “a” in Fig. 44(a) in response to some perturbation and onset of dynamics as e.g. global dispersion according to Fig. 44(c). Shown behaviour strongly suggests that here the non-trivial rheology renders these separatrices unstable and thus induces the de-
Plate tectonics due to mantle convection

Up/downwellings demarcating convection cells

Single-sided subduction

Fig. 51. Plate tectonics induced by mantle convection as geophysical instance of convection cells: (a) mechanism (reproduced from [228]); (b) up- and downwellings demarcating convection cells visualised by “low” (blue) and “high” (yellow) simulated temperatures (adapted from [228]); (c) single-sided subduction demonstrated by intrusion of low-temperature (blue) crust material into high-temperature (red) mantle at position 1 and resulting circulation zone between positions 1 and 2 (adapted from [230]).

Micro-circulation in living tissue

Exchange of oxygen, carbon dioxide and nutrients with living tissue occurs via so-called “micro-circulation” through a local network of microscopic blood vessels [32]. Each organ has its own dedicated micro-circulation; e.g. lung alveoli (Sec. 3.3.3) are covered by pulmonary capillaries for oxygen transfer into the blood stream. Fig. 52(a) illustrates the complexity of microvascular networks by that of the somatosensory cortex in the brain of a rat visualised by 3D imaging using X-ray tomographic microscopy [231]. The tissue and embedded network effectively constitutes a random porous medium, implying, by analogy with such media, generic transport characteristics as demonstrated in Fig. 47(a) chaotic advection locally within the vessels and, in consequence, relatively weak global stream-wise dispersion.

Important in a physiological context is the impact of local disruptions or malformations of the vascular network on the global transport by the micro-circulation. The computational study by [231] investigates the effect of a vascular occlusion (i.e. blockage) of the blood vessel at the arrow in Fig. 52(a) by reduction of its diameter by a factor 1,000 using a model based on graph representations of the microvasculature. Blue and red vessels in Fig. 52(a) have a significantly lower and higher flow rate, respectively, compared to the unobstructed situation (green vessels are unaffected) and, by emerging throughout the entire network, expose a substantial impact of the occlusion on the 3D global throughflow (from bottom to top). This is likely to cause departures from the transport properties of healthy networks. Typical vessel-wise residence times, irrespective of the nature of the local transport, directly correlate with flow rates. Moreover, local transport depends, similar to periodic duct flows (Sec. 3.2), strongly on the flow rate. Hence, though probably retaining its chaotic nature due to the random connectivity, quantitative changes may nonetheless alter the residence-time distributions of affected blood vessels. The occlusion thus is expected to considerably affect the 3D global (stream-wise) dispersion properties. Insights into both local and global impact of occlusions on 3D transport are crucial for deeper understanding of the physiology of strokes, since reduced or disrupted cerebral oxygen delivery is a major cause for this brain condition [232].

Oclusions, by altering the flow rate, are malformations in the micro-vascular network that affect the (local) permeability (i.e. parameter $\kappa$ in (19)). Malformations in the tissue itself mainly alter another key mesoscopic transport property, namely the (local) porosity $\phi$. Notorious examples are cancerous tumors that, due to their rapid growth, typically have underdeveloped vascular networks and thus effectively reduce $\phi$. Hence tumors rely strongly on diffusion of oxygen from surrounding healthy tissue and, given this is a relatively slow process, tend to become hypoxic, which attributes to their malignant behaviour [233]. Fig. 52(b) demonstrates this for the simulated 3D oxygen transport in a realistic vascular network (red) containing several tumors (green) using the computational model by [234, 235]. The lower-right inset shows the local oxygen distribution in the grey cross-section and reveals a one-to-one correspondence between the oxygen-poor (blue) and oxygen-rich (yellow) regions and the tumor and healthy tissue, respectively.

Micro-vascular networks, unlike subsurface porous re-
4 Unified Lagrangian framework for 3D transport

The discussion in this section seeks to reconcile the above flow categorisation with theoretical/fundamental concepts and methods so as to (i) outline a unified framework for systematic 3D Lagrangian transport analyses of practical flows and (ii) provide a “gateway” to further literature on this subject matter. To this end the essentials of Lagrangian transport are treated in a tutorial-like approach via capita selecta and exemplified with the systems highlighted in Sec. 3.

4.1 Critical points as organising entities

Key organising entities of the Lagrangian flow topology are so-called “critical points” associated with the Lagrangian motion described by kinematic equation (2). Critical points “shape” the flow topology by geometrically and topologically governing the proliferation of fluid trajectories. These entities are common (or even inherent) in confined geometries both in the flow interior and on no-slip (internal) walls and thus are of great relevance to practical flows. The emergence and nature of critical points is elaborated below for 2D and 3D flows under steady and time-periodic conditions.

4.1.1 Critical points in 2D flows

Critical points in steady flows \( \vec{u} = \vec{u}(\vec{x}) \) correspond with stagnation points, that is, positions \( \vec{x}_0 \) at which the fluid motion vanishes:

\[
J(\vec{x}_0) = 0 \quad \Rightarrow \quad \vec{x}(t) = \Phi_t(\vec{x}_0) = \vec{x}_0. \tag{20}
\]

The flow topology near stagnation points \( \vec{x}_0 \) is determined by the linearisation \( G(t) \) of flow \( \Phi_t \), i.e.

\[
\vec{x}'(t) = G(t)\vec{x}_0', \quad G(t) = e^{At}, \quad A = \left. \frac{\partial \vec{u}}{\partial \vec{x}} \right|_{\vec{x}_0},
\]

with \( \vec{x}' = \vec{x} - \vec{x}_0 \) the local position vector and \( A \) the strain-rate tensor. The linearised flow in spectral representation reads

\[
G(t) = \sum_{i=1}^{2} e^{\lambda_i t} \vec{v}_i, \quad \lambda_{1,2} = \frac{J_1 \pm \sqrt{J_1^2 - 4J_2}}{2},
\]

with \((\lambda_i, \vec{v}_i)\) the eigenvalue-eigenvector pairs of \( A \) and

\[
J_1 = \vec{V} \cdot \left. \frac{\partial \vec{u}}{\partial \vec{x}} \right|_{\vec{x}_0}, \quad J_2 = \left. \frac{\partial u}{\partial x} \frac{\partial u}{\partial y} \right|_{\vec{x}_0} - \left. \frac{\partial u}{\partial y} \frac{\partial u}{\partial x} \right|_{\vec{x}_0},
\]

the invariants of its characteristic equation \( \lambda^2 - J_1 \lambda + J_2 = 0 \). Solenoidal flow through \( J_1 = \text{tr}(A) = \sum \lambda_i = 0 \) yields \( \lambda_1 = -\lambda_2 = \sqrt{-J_2} \) and thus implies two kinds of stagnation points depending on the sign of \( J_2 = \det(A) \). Case \( J_2 > 0 \) gives a complex-conjugate pair of eigenvalues \((i\lambda, -i\lambda)\), with \( \lambda = \sqrt{-J_2} \), and the local Lagrangian motion \([21]\) in polar coordinates becomes

\[
r'(t) = r_0, \quad \theta'(t) = \theta_0 + \lambda \tau,
\]

describing concentric circular streamlines around \( \vec{x}_0 \) following Fig. 53a (left). Case \( J_2 < 0 \) gives a pair of real eigenvalues \((\lambda, -\lambda)\), with \( \lambda = \sqrt{|J_2|} > 0 \), resulting in

\[
\vec{x}'(t) = \sum_{i=1}^{2} \eta_i(t) \vec{v}_i, \quad \eta_i(t) = \eta_i(0) e^{\lambda t},
\]

with \( \eta_0, \eta_1 \) positive real numbers and \( \vec{v}_1 \) and \( \vec{v}_2 \) unit vectors along the positive directions of the real and imaginary parts of \( \lambda \).

4.1.2 Critical points in 3D flows

Stagnation points (red) visualised by simulated oxygen levels (yellow/blue: higher/lower flow rate; green: unaffected region) (adapted from [231]); (a) impact of vascular occlusion (arrow) on global throughflow (from bottom) in somatosensory cortex in rat brain visualised by simulated flow rate (red/blue: relatively larger/lower flow rate; green: unaffected region) (adapted from [231]); (b) impact of tumor cells (green) on oxygen distribution via vascular network (red) visualised by simulated oxygen levels (yellow/blue: healthy/hypoxic) in grey cross section (inset) (adapted from [234]).
as local Lagrangian motion, with \( f = (\eta_1, \eta_2) \) the canonical coordinates associated with the principal strain axes \((\vec{v}_1, \vec{v}_2)\), describing hyperbolic streamlines following Fig. 53(a) (right). Stagnation points “shape” the flow topology by imparting the topology of the linearised flow on their vicinity (i.e., the surrounding region devoid of other stagnation points). The step-wise steady flows in Fig. 3 in fact contain only a single stagnation point of the former kind \((J_2 > 0)\) and the global streamline portrait, in consequence, is topologically equivalent to Fig. 53(a) (left).

Critical points in time-periodic flows correspond with periodic points \((\eta)\) and are direct counterparts of stagnation points \((\eta)\). Local linearisation of mapping \([5]\) namely gives

\[
\dot{x}_p^\prime = F^p x_0, \quad F = \frac{\partial \Phi}{\partial x} \Bigg|_{x_0}, \quad (26)
\]

yielding an identical (spectral) structure as \((22)\) and \((25)\) upon substitution \(u \rightarrow \Phi_T\) and \(e^{\lambda t} \rightarrow \lambda_i\) here the eigenvalue-eigenvector pairs of deformation tensor \(F\). Solenoidality through \(J_2 = \det(F) = \prod \lambda_i = 1\) yields

\[
\lambda_1 = 1/\lambda_2 = J_1/2 + \sqrt{(J_1/2)^2 - 1}, \quad (27)
\]

and again implies two kinds of points yet here depending on \(J_1 = \text{tr}(F)\) due to the different emergence of the eigenvalues in the spectral structure. Case \(|J_1| < 2\) gives a complex-conjugate pair of eigenvalues \((\lambda, \lambda^*)\), with \(\lambda = J_1/2 + i \sqrt{1 - (J_1/2)^2}\) and \(\lambda^* = 1/\lambda\); case \(|J_1| > 2\) gives a pair of real eigenvalues \((\lambda, 1/\lambda)\), with \(\lambda = \lambda_1 > 1\). This results for former and latter case in

\[
(r_p^\prime, \theta_p^\prime) = (r_0^\prime, \theta_0^\prime + p\phi), \quad \dot{x}_p^\prime = \sum_{i=1}^2 \eta_i \lambda_i^p \vec{v}_i, \quad (28)
\]

respectively, with \(\phi = \arg(\lambda) = \arctan(\sqrt{|/J_1|^2 - 1})\), revealing local Lagrangian motion identical to that near the stagnation points following \((24)\) and \((25)\). (Note \(|\lambda| = 1\) for complex \(\lambda\).) Hence the periodic points for \(|J_1| < 2\) and \(|J_1| > 2\) are indeed direct counterparts to the above stagnation points for \(J_2 > 0\) and \(J_2 < 0\), respectively \([131]\).

Stagnation/periodic points for complex-conjugate and real eigenvalue pairs are denoted elliptic and hyperbolic points, respectively, based on the structure of the local flow topology (Fig. 53).\(^{23}\) Elliptic and hyperbolic points create LCSs that are topologically equivalent to Fig. 53(a) (left) and Fig. 53(a) (right), respectively. Thus elliptic points constitute centres of the islands in e.g. Fig. 3(a) and Fig. 5(a)\(^2\); principal axes \(\vec{v}_1\) and \(\vec{v}_2\) of hyperbolic points are the origin of unstable and stable manifolds, respectively, as shown e.g. in Fig. 5(a)\(^2\).

The above reveals that elliptic and hyperbolic points are the only critical points possible in 2D solenoidal flows. This has the fundamental implication that the corresponding flow topologies are always composed of arrangements of LCSs corresponding with these points. Hence flow topologies with islands embedded in chaotic regions demarcated by manifolds following Fig. 53(a) are characteristic for generic 2D time-periodic flows. However, important to note is that hyperbolic points and manifolds, though typically present, do not imply chaos perse; decisive for chaotic advection is the particular behaviour of the manifolds (Sec. 4.6.1).

4.1.2 Critical points in 3D flows

Stagnation points and associated flow topologies are in 3D steady flows also determined by the spectral properties of strain-rate tensor \(A\) following \((21)\), which are governed by the characteristic equation \(\lambda^3 - J_1 \lambda^2 + J_2 \lambda - J_3 = 0\), with invariants \(J_1 = \text{tr}(A), J_2 = (\text{tr}^2(A) - \text{tr}(A^2))/2\) and \(J_3 = \det(A)\). Solenoidality again yields \(J_1 = 0\) and real \(A\) implies (at least) one real eigenvalue (say \(\lambda_3\)). Hence the dynamics in the \(\eta_3\)-direction (spanned by eigenvector \(\vec{v}_3\)) is always described by

\[
\eta_3(t) = \eta_3(0)e^{\lambda_3 t}, \quad (29)
\]

meaning that stagnation points differ solely by the behaviour in the \((\eta_1, \eta_1)\)-plane. Two kinds can again be distinguished.

Case \(J_3 > \lambda_3^2/4\) yields (besides real \(\lambda_3\)) a complex-conjugate pair of eigenvalues \((\lambda_R + i\lambda_I, \lambda_R - i\lambda_I)\), with

\[
\lambda_R = -\lambda_3/2, \quad \lambda_I = \sqrt{J_3/\lambda_3 - (\lambda_3/2)^2}, \quad (30)
\]
resulting in local Lagrangian motion \((21)\) in the \((\eta_1, \eta_2)\)-plane (in polar coordinates) according to

\[ r'(t) = r_0 e^{\lambda t}, \quad \theta'(t) = \theta_0' + \lambda t, \quad (31) \]

describing spiralling streamlines about the \(\eta_3\)-axis following \(\text{Fig. 53(b)}\) (left). This kind of stagnation point is denoted hyperbolic focus and is a 3D generalisation of the elliptic points of 2D flows. Principal difference with the latter is the (generically) non-constant radius – and ensuing spiralling motion – due to \(\lambda_R = -\lambda_3/2 \neq 0\). Two situations may occur: (i) distancing from the \((\eta_1, \eta_2)\)-plane while contracting around the \(\eta_3\)-axis for \(\lambda_3 > 0\); (ii) approaching the \((\eta_1, \eta_2)\)-plane while diverging from the \(\eta_3\)-axis for \(\lambda_3 < 0\). Said plane and axis define the 2D unstable (stable) and 1D stable (unstable) manifolds, respectively, in the former (latter) case. The infinitesimal plane spanned by principal axes \(\vec{v}_{1,2}\) is the origin of the 2D manifold in physical space; principal axis \(\vec{v}_3\) is the origin of the corresponding 1D manifold.

Case \(J_3 < \lambda_3^2/4\) gives (besides real \(\lambda_3\)) real eigenvalues

\[ \lambda_{1,2} = -\lambda_3/2 \pm \sqrt{(\lambda_3/2)^2 - J_3/\lambda_3}, \quad (32) \]

and (upon properly redefining the canonical frame) can always be chosen such that \(\lambda_1\) and \(\lambda_{2,3}\) are of opposite sign. Thus Lagrangian motion in both the \((\eta_1, \eta_2)\) and \((\eta_1, \eta_3)\)-planes is essentially similar to the hyperbolic points of 2D flows according to \((25)\), and yields 3D dynamics following \(\text{Fig. 53(b)}\) (right). Hence the present kind of stagnation point is a generalisation of said points and denoted hyperbolic node hereafter.\(^{24}\) Here the \((\eta_2, \eta_3)\)-plane and \(\eta_1\)-axis define the 2D unstable (stable) and 1D stable (unstable) manifolds, respectively, for \(\lambda_1 < 0\) (\(\lambda_1 > 0\)). The steady lid-driven cylinder flow for \(Re > 0\) in \(\text{Fig. 23(b)}\) and \(\text{Fig. 23(c)}\) e.g. accommodates a hyperbolic focus (not shown) at the intersection of the toroidal axis of the tori and the symmetry plane \(y = 0\). Said axis and plane in fact define the corresponding 1D and 2D manifolds, respectively.

The direct analogy between stagnation and periodic points is retained for 3D flows \([136]\). The local behaviour near periodic points remains dependent on the deformation tensor \(F\) following \((26)\); that, upon the substitution adopted before, again assumes the same spectral structure as \(A\). Solenoidal flow gives \(J_1 = \det(F) = 1\) and real \(F\) implies (at least) one real eigenvalue (say \(\lambda_3\)) and, in consequence, \(\eta_3\)-wise dynamics similar to \((29):\eta_{3,p} = \eta_{3,0} e^{\lambda_3\cdot}\). Hence the kind of periodic points in 3D flows is also determined by the behaviour in the \((\eta_1, \eta_1)\)-plane. Case \(J < 2/\sqrt{\lambda_3}\) and \(J > 2/\sqrt{\lambda_3}\), where \(J = \lambda_1 + \lambda_2 = J_1 - \lambda_3\), define the counter-

\(^{24}\)The nomenclature of \([136]\) is adopted as uniform terminology for stagnation and periodic points. The classification for (instantaneous) 3D streamline patterns by \((24)\) refers to the hyperbolic focus as “focus” and the hyperbolic node as “node-saddle-saddle” due to the saddle-like and node-like dynamics in the \((\eta_1, \eta_2)\), \((\eta_1, \eta_1)\)-planes and \((\eta_2, \eta_3)\)-plane, respectively.

parts to \((30)\) and \((32)\), respectively, with here

\[ \lambda_R = J/2, \quad \lambda_I = \sqrt{1/\lambda_3 - (J/2)^2}. \quad (33) \]
as real and imaginary parts of the complex eigenvalue and

\[ \lambda_{1,2} = J/2 \pm \sqrt{(J/2)^2 - 1/\lambda_3}, \quad (34) \]
as real eigenvalues. The local Lagrangian motion in the \((\eta_1, \eta_2)\)-plane is a generalisation of \((28)\) and becomes

\[ (r_p', \theta_p') = (|\lambda| p r_0, \theta_0 + p \phi), \quad \vec{x}_p = \sum_{i=1}^2 \eta_i \lambda_i \vec{v}_i, \quad (35) \]

with \(\phi = \arctan(\sqrt{(2/J)^2/\lambda_3 - 1})\) and \(|\lambda| = 1/\sqrt{\lambda_3}\). (Principal difference with 2D flows is that generically \(|\lambda| \neq 1\) and \(\lambda_1 \lambda_2 = 1/\lambda_3 \neq 1\). Thus cases \(J < 2/\sqrt{\lambda_3}\) and \(J > 2/\sqrt{\lambda_3}\) result in dynamics according to \(\text{Fig. 53(b)}\) (left) and \(\text{Fig. 53(b)}\) (right), respectively, and indeed are entirely equivalent to the above stagnation points. Hence former and latter are also denoted hyperbolic focus and hyperbolic node. The isolated periodic point in the time-periodic cylinder flow in \(\text{Fig. 26(b)}\) (not shown) e.g. is a hyperbolic focus. The highly-convoluted 2D and 1D manifolds emanate from the \((\eta_1, \eta_2)\)-plane and \(\eta_3\)-axis, respectively, of the local canonical frame in \(\text{Fig. 53(b)}\) (left) and demonstrate the global complexity that these entities may assume. However, key for the nature of the Lagrangian dynamics, that is, chaotic versus non-chaotic, is – in steady and time-periodic 3D flows – again the particular behaviour of these manifolds (Sec. \(4.6.1\)).

Critical points of the skin friction field \(\vec{t}\) of 3D steady flows following \((18)\) have a flow topology that corresponds with the hyperbolic focus and emerge in a “saddle” and “node” configuration if the canonical \((\eta_1, \eta_2)\)-plane and \((\eta_2, \eta_3)\)-plane, respectively, in \(\text{Fig. 53(b)}\) (right) coincides with the wall \((\text{215, 217})\). (The fact that generically \(\vec{t} \cdot \vec{t} \neq 0\) is immaterial here.) The associated 1D and 2D manifold give rise to a streamline and stream surface extending from the wall into the domain and saddle configurations, respectively, in a way as demonstrated in \(\text{Fig. 47(b)}\).

### 4.1.3 Critical lines in 3D flows

Stagnation points may merge into continuous curves and thus form so-called “stagnation lines” that, by definition, are characterised by absence of motion – and, inherently, deformation – in the tangent direction \(\vec{e}_t\). This implies \(G e_t = e_t\) for the linearised flow \((21)\) and thus renders the tangent an eigenvector of the strain-rate tensor \(A\), say \(\vec{v}_3 = e_t\), with corresponding eigenvalue \(\lambda_3 = 0\). Tensor \(A\) becomes singular for \(\lambda_3 = 0\) (i.e. \(J_3 = 0\)) and invalidates expressions \((30)\) and \((32)\) for the remaining eigenvalues \(\lambda_{1,2}\) due to an undefined fraction \(J_1/\lambda_3\). Alternative relations are found via the simplified characteristic equation in the singular state, i.e.
\[ \lambda^2 - \lambda_1 \lambda + \lambda_2 = 0, \]  
that through \( J_1 = 0 \) yields \( \lambda_{1,2} = \sqrt{-J_2} \)  
and thus gives the same form as for stagnation points in 2D steady flows. Hence the dynamics in the canonical \((\eta_1, \eta_2)\) -plane transverse to each point on the stagnation line is either elliptic \((J_2 > 0)\) or hyperbolic \((J_2 < 0)\) according to Fig. 53(a) (left) and Fig. 53(a) (right), respectively. This has the fundamental implication that stagnation lines in 3D flows signify (locally) essentially 2D dynamics. The common centre (dots) of the global family of closed streamlines in the Stokes limit \((Re = 0)\) of the steady cylinder flow in Fig. 23(a) e.g. is a stagnation line consisting entirely of elliptical points.

Periodic points may in a similar way merge into so-called “periodic lines” and are the counterpart to stagnation lines in 3D time-periodic flows. Here absence of motion in tangent direction \( \dot{\eta} \) implies \( F \dot{\eta} = \dot{\eta} \) for the linearised stroboscopic map \( (26) \) and thus gives an eigenvalue-eigenvector pair \( (\lambda_3, \vec{\eta}_3) = (1, \vec{\eta}_1) \). Relations \( (33) \) and \( (34) \) for \( \lambda_3 = 1 \) collapse on relation \( (27) \) for \( |J| < 2 \) and \( |J| > 2 \), respectively, and consequently identify with the spectral signatures for elliptic and hyperbolic points in 2D time-periodic flows. The dynamics transverse to periodic lines is, entirely analogous with stagnation lines, therefore also essentially 2D.

The periodic line in the time-periodic cylinder flow in Fig. 27(b) e.g. is partitioned into segments of elliptic (normal) and hyperbolic (heavy) points, yielding local transverse dynamics following Fig. 53(a) (left) and Fig. 53(a) (right), respectively. The quasi-2D chaos in the time-periodic cylinder flow driven by alternating x-wise translation of top and bottom walls in Fig. 26(a) (centre) e.g. emanates from multiple periodic lines arranged transverse to the moving walls and consisting entirely of elliptical or hyperbolic points (not shown) [136]. The “holes” in the chaotic layer outline tubes formed by merger of islands of the constituent periodic points of the elliptic lines. Both the stable and unstable manifolds of the hyperbolic lines (in time-periodic as well as steady flows) are 2D surfaces that, similarly, result from merger of the manifold pairs of its individual points.

4.1.4 Closed trajectories in 3D flows

An important generalisation of critical points and lines exists in closed streamlines in 3D steady flows and closed trajectories in stroboscopic maps of 3D time-periodic flows.

Closed streamlines in 3D steady flows are described by fluid parcels that return to their initial position after a finite cycle time \( \tau \), i.e. \( \Phi_T(\vec{x}_0) = \vec{x}_0 \), meaning that the parcel position \( \vec{x}(t) \) is a periodic solution of kinematic equation \( (2) \): \( \vec{x}(t) = \vec{x}(t + \tau) \). The local dynamics near a fluid parcel with momentary position \( \vec{x}_0(t) \) is governed by a linearised flow of the form \( (21) \) in the co-moving reference frame \( \vec{x}(t) = \vec{x}(t) - \vec{x}_0(t) \). The periodic nature of the strain-rate tensor, i.e. \( A(t + \tau) = A(t) \), admits a linear coordinate transformation \( \vec{y}(t) = P(t) \vec{x}(t) \), with periodic transformation matrix \( P(t) = P(t + \tau) \), such that the linearised kinematic equation \( (2) \) near \( \vec{x}_0(t) \) becomes time-independent, i.e.

\[
\frac{d\vec{x}}{dt} = A(t)\vec{x}(t) \Rightarrow \frac{d\vec{y}}{dt} = B\vec{y}(t),
\]

with \( B \) the constant strain-rate tensor in the \( \vec{y} \)-frame [241]. The equivalence with the flow \( (21) \) near stagnation points readily yields \( \vec{y}(t) = e^{Bt} \vec{y}_0 \) and, in turn, leads to

\[
\vec{x}(t) = G(t)\vec{x}_0, \quad G(t) = P(t)e^{B(t - t)},
\]

as linearised flow in the co-moving physical frame. Note that flow \( G \) generically is non-periodic: \( G(t) \neq G(t + \tau) \).

Relation \( (37) \) exposes the flow near a closed streamline as a local similarity transform of the steady flow in the \( \vec{y} \)-frame, meaning that the dynamics is determined by the spectral properties of \( B \), which has important ramifications. Material line elements of the streamline return to their initial position after time \( \tau \), implying \( G(\tau)\vec{y}(\tau) = \vec{y}(0) \) again the tangent, resulting in an eigenvalue-eigenvector pair \( (\lambda_3, \vec{y}_3) = (0, P^{-1}(\tau)\vec{y}(\tau)) \) for \( B \). This situation is identical to that for stagnation lines and thus gives

\[
\lambda_{1,2} = \sqrt{-J_2(B)},
\]

for the eigenvalues in the direction transverse to the closed streamline, rendering also here the behaviour essentially 2D. Moreover, given \( B \) characterises the entire closed streamline, the dynamics is qualitatively the same everywhere (i.e. the behaviour remains either fully elliptic or hyperbolic during the excursion of a fluid parcel).

Closed streamlines are key LCSs in many of the practical steady flows considered in Sec. 3. The tori in the impeller-driven flow in Fig. 18(b) are centred on elliptic closed streamlines and the surrounding chaotic sea emanates from accompanying hyperbolic closed streamlines. The inherent 2D nature of the transverse dynamics established above explains the qualitative resemblance of the cross-sectional topology in Fig. 18(c) and Fig. 18(d) with 2D time-periodic systems (Fig. 5(a)). The tori and chaotic regions in e.g. the rotating-lid and translating-lid cylinder flows in Fig. 22 and Fig. 23, respectively, the ACEO-driven flow in Fig. 24 as well as the baseline topologies of micro-droplets in Fig. 36(b) correspond in a similar way with elliptic/hyperbolic streamlines.

Streamlines reconnecting via the periodic inlet-outlet of steady duct flows (denoted “periodic streamlines” hereafter) must always exist in these systems and thus are crucial to the Lagrangian transport properties of e.g. industrial inline mixers [86]. These LCSs namely invariably result in a flow topology as demonstrated for the RAM in Fig. 7(a) stream-wise tori centered on elliptic streamlines embedded in chaotic regions “driven” by hyperbolic streamlines. Former and latter emerge as elliptic and hyperbolic periodic points, respectively, in the cross-sectional topology.
in Fig. 7(b) The magnetic flux surfaces in MHD representations of tokamak fusion reactors in Fig. 17 are, by analogy with periodic duct flows (Sec. 3.2.3), centered on elliptic magnetic field lines. The “o-points” and “x-points” in Fig. 17(b) are poloidal intersections of elliptic and hyperbolic field lines. The invariable emergence of closed (magnetic) streamlines in these systems also here explains their essentially 2D (chaotic) dynamics.

Closed trajectories in 3D time-periodic flows correspond with stroboscopic maps (4) of individual fluid parcels that densely fill closed loops over infinitely many periods. To this end the associated mapping \( \Phi_T \) must be aperiodic, that is, the individual parcel positions in \( \Phi_T \) remain distinct at all times. Such closed trajectories e.g. sit at the centre of the tori in the rheology-induced time-periodic cylinder flow in Fig. 25(a) and the time-periodic annular flow in Fig. 27(a).

Closed trajectories thus described are, irrespective of the progression of fluid parcels along discrete positions, smooth and continuous closed curves reminiscent of a closed streamline. Continuity then suggests an equally smooth and continuous closed curves - its periodic solution \( x(s) = x(s + \tau) \).

Fictitious time \( s \) acts as a spatial coordinate along the streamlines of \( \mathcal{V} \) and thus admits expression of the positions \( x_p \) of a fluid parcel on the closed trajectory in terms of a corresponding \( s_p \). This, via (37), readily yields

\[
\dot{x}_p = F_p \mathbf{v}_0, \quad F_p = P(s_p)e^{B_\psi}P^{-1}(s_p),
\]

as local mapping in the co-moving reference frame and, by analogy, implies the same behaviour as near closed streamlines: essentially 2D dynamics governed by eigenvalue pair \( \{B_\psi \} \) of either fully elliptic or fully hyperbolic nature.

The extent of the tori in the beforementioned rheology-induced time-periodic cylinder flow in Fig. 25(a) demonstrates that the essentially 2D behaviour associated with closed trajectories may impact a sizeable region. However, the 2D dynamics imparted by these entities can also be very localised, as exemplified by the defective tori – signifying 3D dynamics – in the time-periodic annular flow in Fig. 27(a) due to RID. Similar restriction of the 2D dynamics of closed trajectories by resonance-induced tori breakdown is suspected in e.g. time-periodic droplet flows driven by electro-wetting in Fig. 38 and the time-periodic flow in the vitreous chamber of the eye (Fig. 42). The basic mechanisms of RID are elaborated in Sec. 4.6.2.

4.1.5 Symmetries as organising mechanisms

Symmetries play a critical role in the structure of the flow topology and, inherently, the Lagrangian dynamics of 3D (un)steady flows [43]. Insight into this role is important especially for industrial flows, since manufactured devices and engineered flows often have symmetries due to the geometry and/or the repetitive nature of the process such as the systematic flow reorientation in periodic duct flows (Sec. 3.2.2).

Discrete geometrical and dynamical symmetries in particular are key organising mechanisms for critical points/lines and closed trajectories as well as their associated LCSs. An important class exists in reflectional symmetries \( S \) (i.e. mirroring positions or LCSs about some symmetry plane \( \mathcal{P} \)) and two important instances can be distinguished within the present scope: ordinary and time-reversal reflectional symmetries. Former and latter are for both flow \( \Phi_T \) and mapping \( \Phi_T \) following (2) and (5) given by

\[
\hat{\Phi} = S\Phi S, \quad \hat{\Phi} = S\Phi^{-1}S,
\]

respectively, and relate the (reversed) dynamics on either side of the symmetry plane \( \mathcal{P} \). Fig. 54(a) illustrates this for the time-reversal reflectional symmetry about plane \( x = 0 \) (i.e. \( S_x : (x,y) \rightarrow (-x,y) \)); the trajectory in the forward flow starting from \( x \) is the mirror image of that in the reversed flow starting from \( S(x) \) about \( x = 0 \).

Both ordinary and time-reversal reflectional symmetries following (40) imply emergence of LCSs \( \mathcal{L} \) as either entities symmetric about \( S \), i.e. \( S(\mathcal{L}) = \mathcal{L} \) or as symmetric pairs \( \{L, S(\mathcal{L})\} \). Time-reversal reflectional symmetries furthermore imply stagnation lines (steady flow) and periodic lines (time-periodic flow) in symmetry plane \( \mathcal{P} \) [136][124]. The steady cylinder flow for \( Re = 0 \) in Fig. 23(a) e.g. has an ordinary reflectional symmetry \( S_y : (x,y) \rightarrow (-x,y) \) and time-reversal reflectional symmetry \( S_y : (x,y) \rightarrow (-x,y) \) about plane \( y = 0 \) and plane \( x = 0 \), respectively, resulting in the (elliptic) stagnation line in the former and symmetric streamline pairs about the latter plane. Symmetry \( S_y \) is preserved under inertial conditions (\( Re > 0 \)) and thus dictates a symmetric arrangement of tori and chaotic trajectories in Fig. 23(b) and Fig. 23(c) about plane \( y = 0 \) (mirror images not shown). The periodic line in plane \( y = -x \) of the time-periodic cylinder flow in Fig. 27(b) (right) in a similar way emanates from a time-reversal reflectional symmetry \( S_{xy} : (x,y,z) \rightarrow (-y,-x,z) \) [124]. Symmetries may also manifest themselves locally. The time-reversal reflectional symmetry \( S_a : (r,\theta) \rightarrow (r,\alpha-\theta) \) of the base flow \( \mathbf{v} \) of the 2.5D RM flow about the centerline \( \alpha/2 = \pi/8 \) of the first aperture results in a reflectional symmetry \( S_a : (r,\theta) \rightarrow (r,\alpha-\theta) \) about axis \( \alpha = (\alpha-\theta)/2 = -3\pi/40 \) in the cross-section (Fig. 7(b)) of the 3D flow topology in Fig. 7(b) [83].

Time-reversal reflectional symmetries furthermore imply that streamlines (within a confined flow domain) crossing the symmetry plane \( \mathcal{P} \) must cross twice and form closed streamlines [122][126]. Thus symmetry \( S_y \) of the non-inertial steady cylinder flow gives rise to a continuous family of closed streamlines centered on said stagnation line (of which one specimen is shown in Fig. 23(a)). Conversely, breaking of such a symmetry (if existent) is necessary for non-trivial
Continuous symmetry

4.2 Hamiltonian structure of 2D flows

Sec. 4.3 revealed that Lagrangian transport in many 3D flows is (locally) similar to that of 2D unsteady flows and the above advanced critical lines (Sec. 4.1.3) and closed trajectories (Sec. 4.1.4) as LCSs underlying such (effectively) 2D behaviour. Isolated critical points (Sec. 4.1.2), on the other hand, are key indicators of essentially 3D dynamics.

Hence the emergence (or absence) of such LCSs offers important insights into the nature of the Lagrangian motion. However, a more rigorous distinction of true 3D from (essentially) 2D dynamics than just based on LCSs is relevant for analysis and design of practical flows. Batch mixers using time-periodic stirring must e.g. operate at conditions yielding global 3D chaos such as in Fig. 26(a) (right); any (local) 2D behaviour implies sub-optimal performance of such devices. The ability to unambiguously distinguish between 2D and 3D dynamics enables identification and accomplishment of favourable operating conditions.

The 2D–3D distinction may rely upon the fundamental property that kinematic equation (2) for solenoidal 2D unsteady flows has a Hamiltonian structure, viz.

\[
dx = \frac{∂H}{∂y}, \quad dy = \frac{∂H}{∂x},
\]

and thus restricts the motion of fluid parcels to its level sets. Property (42) renders \( H \) a so-called “constant of motion” (COM) and here defines the conventional stream function of 2D steady flows. Restriction of fluid motion to level sets of \( H \) precludes chaotic advection and thus puts forth unsteadiness, i.e. \( H = H(\vec{x}, t) \), as a necessary (though not sufficient) condition for chaos in 2D flows \[242, 52\]. This namely invalidates property (42) by virtue of \( \frac{dH}{dt} = 0 \). Flows devoid of chaotic behaviour due to the restriction of Lagrangian dynamics to level sets of a COM \( H \) are termed integrable \[243\]; \( H \) namely implicitly gives the solution \( \vec{x}(t) = \Phi_t(\vec{x}_0) \) to kinematic equation (2) via \( H(\vec{x}(t)) = H(\vec{x}_0) \).

The Hamiltonian structure furthermore dictates that chaos, if occurring, emanates from the breakdown of islands following specific scenarios described by the well-known KAM and Poincaré-Birkhoff theorems \[52, 243\]. This results in the characteristic composition of flow topologies of systems exhibiting Hamiltonian chaos, i.e. large central islands encircled by island chains and embedded in a chaotic sea, as happening in e.g. the cross-sectional dynamics of the 3D steady flows shown in Fig. 18(d) or Fig. 22 or Fig. 23(d). Counterparts to these theorems exist for such 3D steady flows exhibiting essentially 2D behaviour \[43\].

4.3 Hamiltonian dynamics in 3D steady flows

4.3.1 General

The fact that the Lagrangian dynamics in 2D solenoidal flows is always governed by Hamiltonian mechanics offers a way to establish whether a 3D steady flow exhibits (essentially) 2D dynamics. This notion namely implies that any

---

25Steady/unsteady Hamiltonian systems are in dynamical-systems terminology commonly denoted autonomous/non-autonomous systems \[243\].
such flow must admit a (formal) transformation \( \mathcal{F} \) following (11) into a 2D unsteady system with a Hamiltonian structure according to (41). This transformation rests on three cornerstones. First, a coordinate transformation

\[
\mathcal{G} : \vec{x} \rightarrow \vec{\xi}, \quad (43)
\]

from the Cartesian frame \( \vec{x} = (x, y, z) \) to an appropriate curvilinear reference frame \( \vec{\xi} = (\xi_1, \xi_2, \xi_3) \) with scaling factors \( h_1 = |\partial \vec{x}/\partial \xi_1| \) and Jacobian \( J = |\partial \vec{x}/\partial \vec{\xi}| = h_1 h_2 h_3 \) following (44). Second, the vector potential \( \vec{A} \) of \( \vec{u} \) for vanishing \( \xi_2 \)-component, i.e. \( \vec{A} = (\vec{A}_1, 0, \vec{A}_3) \), defined implicitly as

\[
h_2 h_3 u_1 = \tilde{u}_1 = -\frac{\partial \vec{A}_1}{\partial \xi_2},
\]

\[
h_1 h_2 u_2 = \tilde{u}_2 = -\frac{\partial \vec{A}_1}{\partial \xi_3} + \frac{\partial \vec{A}_3}{\partial \xi_1},
\]

\[
h_1 h_2 u_3 = \tilde{u}_3 = -\frac{\partial \vec{A}_1}{\partial \xi_1}, \quad (44)
\]

with \( \vec{A}_1 = h_1 \vec{A}_1 \) and \( \tilde{u}_1 = Ju_1/h_1 \) the vector potential and flow, respectively, in the Cartesian frame spanned by \( \vec{\xi} \). Third, expression of kinematic equation (2) in terms of \( \vec{\xi} \) and \( \vec{A}_1 \), i.e.

\[
\begin{align*}
\frac{d\xi_1}{dt} &= \frac{u_1}{J} - \frac{1}{J} \frac{\partial \vec{A}_1}{\partial \xi_2}, \\
\frac{d\xi_2}{dt} &= \frac{u_2}{J} - \frac{1}{J} \left[ \frac{\partial \vec{A}_1}{\partial \xi_3} - \frac{\partial \vec{A}_3}{\partial \xi_1} \right], \\
\frac{d\xi_3}{dt} &= \frac{u_3}{J} + \frac{1}{J} \frac{\partial \vec{A}_1}{\partial \xi_1},
\end{align*} \quad (45)
\]

which reveals that the Lagrangian motion corresponds with rescaled flow \( \tilde{u}/J \) in said Cartesian \( \vec{\xi} \)-frame: \( d\xi_i/dt = \tilde{u}_i/J \).

The simplest class of 3D steady flows with an embedded Hamiltonian structure involves cases that admit a transformation (43) such that the \( \xi_3 \)-wise flow in \( \vec{\xi} \)-space is independent of \( \xi_3 \) and uni-directional: \( \vec{u}_3 = \tilde{u}_3(\xi_1, \xi_2) > 0 \). This via (44) implies \( \vec{A}_1 = \vec{A}_1(\xi_1, \xi_2) \) and, in turn, a Hamiltonian structure for the \( \xi_1, \xi_2 \)-wise component of (45) according to

\[
\begin{align*}
\frac{d\xi_1}{dt} &= \frac{\tilde{u}_1}{J} = \frac{1}{J} \frac{\partial H}{\partial \xi_2}, \\
\frac{d\xi_2}{dt} &= \frac{\tilde{u}_2}{J} = -\frac{1}{J} \frac{\partial H}{\partial \xi_1},
\end{align*} \quad (46)
\]

with \( H = \vec{A}_1(\xi_1, \xi_2, \xi_3) \) the corresponding Hamiltonian. Property \( d\xi_3/dt = \tilde{u}_3/J > 0 \) (due to \( \tilde{u}_1 > 0 \) and non-singular transformation (43)) admits inversion of \( \xi_3(t) \) and thus expression of the Hamiltonian \( H = H(\xi_1, \xi_2, t) = \vec{A}_1(\xi_1, \xi_2, \xi_3(t)) \). Hence transformation (11) here becomes

\[
\mathcal{F} : \xi_1 = \xi_1, \quad \xi_2 = \xi_2, \quad \tau = t. \quad (47)
\]

Moreover, a steady Hamiltonian \( H = H(\xi_1, \xi_2) \) yields

\[
\frac{dH}{dt} = \vec{u}_i \vec{\nabla}H = \sum_{i=1}^{3} u_i \frac{\partial H}{\partial \xi_i} = \sum_{i=1}^{3} \frac{\partial H}{\partial \xi_i} = 0. \quad (48)
\]

demonstrating consistency of (46) with (41) and (42).

3D steady flows satisfying only the relaxed condition of a uni-directional \( \xi_3 \)-component of the physical flow, i.e. \( u_3 > 0 \) yet permitting \( \partial \mathcal{A}_1/\partial \xi_3 \neq 0 \) in (43) and (45), define a second class with an embedded Hamiltonian structure. Transformation (11) following (43), given by

\[
\mathcal{F} : \xi_1 = \xi_1, \quad \xi_2 = -\vec{A}_1(\xi), \quad \tau = \xi_3, \quad (49)
\]

namely yields a Hamiltonian system according to (41) in canonical space \( \xi = (\xi_1, \xi_2) \) and time \( \tau \), i.e.

\[
\begin{align*}
\frac{d\xi_1}{d\tau} &= \frac{\partial H}{\partial \xi_2}, \\
\frac{d\xi_2}{d\tau} &= -\frac{\partial H}{\partial \xi_1},
\end{align*} \quad (50)
\]

with Hamiltonian \( H = H(\xi_1, \xi_2, \tau) = \vec{A}_3(\xi_1, f(\xi_1, \xi_2, \tau), \tau) \) and \( \xi_2 = f(\xi_1, \xi_2, \tau) \) implicitly defined via (49). Transformation (47) applied to the Cartesian frame \( \vec{\xi} = \vec{x} \) thus readily yields Hamiltonian form (46), with \( H = H(r, \theta) = \vec{A}_3(\xi_1, \xi_2, \tau), \) and here in fact identifies with (41). This renders the duct flow \( \vec{u} \) a cell-wise reorientation of a steady Hamiltonian system, meaning that chaotic advection can occur only due to jumping of tracers between cell-wise reorientations of the KAM tori of the base flow at the cell interfaces \( z = kL \). The embedded Hamiltonian structure causes all 2.5D duct flows to behave according to one universal Hamiltonian scenario (83). The 2.5D approximation is a standard modelling approach for periodic duct flows and inline mixers (Sec. 3.2.2) and is therefore adopted in many studies on such devices (52) (50). The flow topology in the RAM in Fig. 7 is e.g. simulated with the 2.5D model from (83) and computational analyses on the PPM commonly use the 2.5D models by (52) and (87).

Smooth transition between cells yields – even in the Stokes limit – an essentially 3D duct flow \( \vec{u}(x, y, z) \) that precludes Hamiltonian structure (46). However, such flows readily admit the second class of embedded Hamiltonian forms in case of uni-directional axial flow: \( u_3(x) > 0 \) for all \( x \). This namely enables application of transformation (49).
directly to the Cartesian coordinates \( \tilde{x} \), resulting in

\[
(\zeta_1, \zeta_2) = (x, -A_z(x)), \quad \tau = z, \quad H = A_z, \tag{51}
\]

for the associated Hamiltonian form \((50)\). Consider for example the 2.5D RAM flow in Fig. 7(b) which has an axial Poiseuille flow \( v_z = u = 2U (1 - r^2) \), with \( U \) the mean throughflow. Performing transformation \((51)\) for the rescaled flow \( \tilde{u} = \bar{u}/U \), which can be done without loss of generality, via \( \bar{u}_r = 2 (1 - r^2) \) yields \( A_z = 2y/(3 + x^2 - 1) \) and a canonical versus physical cross-section following Fig. 55(a).

The canonical representation of the cross-sectional topology in Fig. 7(b) is shown in Fig. 55(b) and retains its structure, signifying consistency. The 2.5D RAM flow also admits form \((41)\) for reasons given above and can thus be reconciled with two equivalent Hamiltonian systems that employ either real time \( t \) or axial coordinate \( z \) as canonical time.

Transformation \((51)\) applies to any duct flow with \( u_z > 0 \) everywhere and translates into a canonical space/topology that is an \( y \)-wise deformed version of the physical cross-section/topology akin to Fig. 55. The RAM flow in Fig. 9(a) and micro-mixer flow in Fig. 10 following \((88)\) and \((89)\), respectively, e.g. qualify for this approach.

3D steady duct flows with local axial backflow \((u_z < 0)\) admit the second class of Hamiltonian forms \((50)\) upon transformation \((49)\) in the net throughflow region following Sec. 3.2.2 that is, the subregion connecting the periodic inlet-outlet allowing for a non-singular transformation \((53)\) such that \( u_z > 0 \) \([86]\). The remaining flow regions relate to essentially 3D LCSs as e.g. stagnation/periodic points due to internal recirculation zones or separatrices/manifolds emanating from critical points of the skin-friction field \([18]\) on internal walls (as demonstrated in Fig. 47(b) for the flow inside a porous matrix). This happens in the RAM under essentially 3D conditions with significant fluid inertia \([86]\) and is suspected in 3D partitioned-duct flows as e.g. the Quatro mixer in Fig. 11 or the CSM in Fig. 13.

### 4.3.3 Steady circulatory flows

Steady confined circulatory flows as e.g. the impeller-driven flow in Fig. 18 and the swirling flow in Fig. 22 for appropriate conditions admit the above embedded Hamiltonian forms in the reordered cylindrical frame \( \tilde{\xi} = (z, r, \theta) \) \((h_1 = h_2 = 1, h_3 = J = r)\) using \( \theta \) as time-like variable. Axisymmetric angular velocity \( \Omega = u_\theta/r = \Omega(r, z) > 0 \) yields

\[
\frac{dr}{d\tau} = \frac{1}{r} \frac{\partial H}{\partial \zeta_1}, \quad \frac{dz}{d\tau} = -\frac{1}{r} \frac{\partial H}{\partial \zeta_2}, \quad \frac{d\theta}{d\tau} = \frac{\partial H}{\partial \tau}, \tag{52}
\]

as specific Hamiltonian form \((46)\), with \( H = rA_\theta \) the Stokes stream function. Non-axi-symmetric angular velocity \( \Omega > 0 \)

---

Using the rescaled flow for transformation \((51)\) ensures a straightforward link between the Hamiltonian forms. A uniform axial flow \( u_z = U \) in that case namely results (via \( \bar{u}_r = 1 \)) in \( A_z = -y \) and thus identical canonical and physical spaces: \((\zeta_1, \zeta_2) = (x, y)\). Moreover, Hamiltonian systems \((41)\) and \((50)\) then identify upon substituting \( H = UA_z \) and \( \tau = t/U \) in the former.

### 4.4 Hamiltonian dynamics in 3D unsteady flows

#### 4.4.1 Flows with continuous symmetry

A generalisation of the above first class of steady flows with embedded Hamiltonian forms (Sec. 4.3.1) concerns unsteady flows where \( \bar{u} \) following \((44)\) becomes independent of one coordinate direction, say \( \zeta_3 \), for an appropriate transformation \((43)\): \( \tilde{u} = \bar{u}(\zeta_1, \zeta_2, t) \). This happens if a steady solenoidal “flow” \( \tilde{\nu}(\tilde{x}) \) exists, with \( \tilde{\nu}(\lambda) = \tilde{g}_\lambda(\tilde{x}_0) \) the flow along its “streamlines” parameterised by “time” \( \lambda \), such that each fluid trajectory \( \tilde{x}(t) = \Phi_t(\tilde{x}_0) \) has a companion

\[
\tilde{x}_\lambda(t) = \tilde{g}_\lambda\left(\Phi_t(\tilde{x}_0)\right) = \tilde{g}_\lambda(\tilde{x}(t)), \tag{54}
\]

that is also a fluid trajectory \([46]\). Thus \( \tilde{x}_\lambda(t) \) can be reached from \( \tilde{x}_0 \) via two paths and the companion of a fluid trajectory follows from its advection by “flow” \( \tilde{\nu} \) according to Fig. 54(b). This holds for any \( \lambda \in \mathbb{R} \) and therefore gives...
rise to a continuous family of companion trajectories. Hence \( \vec{w} \) defines a \textit{continuous} symmetry and its “streamlines” coincide with fixed positions \((\xi_1, \xi_2)\) in the curvilinear frame, or equivalently, the intersections of the coordinate surfaces of \(\xi_{1,2}\) (implying \(\vec{w} = w \hat{e}_3\) or, conversely, \(\vec{e}_3 = \vec{w}/|\vec{w}|\)).

Property (54) is equivalent to
\[
\vec{w} \cdot \vec{\nabla} \vec{u} - \vec{u} \cdot \vec{\nabla} \vec{\omega} = 0, \tag{55}
\]
and renders \(\vec{u}\) and \(\vec{w}\) commutative, meaning that a given position can be reached via flow by, first, \(\vec{u}\) and, second, \(\vec{w}\) or in reversed order as per Fig. 54(b). Condition (55) in conjunction with solenooidality of \(\vec{u}\) and \(\vec{w}\) through vector identity
\[
\vec{\nabla} \times (\vec{u} \times \vec{w}) = \vec{w} \cdot \vec{\nabla} \vec{u} - \vec{u} \cdot \vec{\nabla} \vec{w} + \vec{u}(\vec{\nabla} \cdot \vec{w}) - \vec{w}(\vec{\nabla} \cdot \vec{u}), \tag{56}
\]
yields
\[
\vec{\nabla} \times (\vec{u} \times \vec{w}) = 0 \quad \Rightarrow \quad \vec{u} \times \vec{w} = -\vec{\nabla} H, \tag{57}
\]
implying a COM \(H\) due to \(\vec{u} \cdot \vec{\nabla} H = -\vec{u} \cdot \vec{u} \times \vec{w} = 0\) that defines a Hamiltonian for the \(\xi_{1,2}\)-wise dynamics [246, 247]. Property \(\vec{u} = \vec{u}(\xi_1, \xi_2, t)\) namely yields via vector potential [41] a Hamiltonian form [40], with here \(H = A_3(\xi_1, \xi_2, t)\), and a corresponding structure for \(\vec{u}\) satisfying (57) for \(\vec{w} = h_3 e_3\). The independence of the Hamiltonian \(H\) on spatial coordinate \(\xi\) has the important consequence that a continuous symmetry in a steady flow implies \(H = H(\xi_1, \xi_2)\) and thus inherently non-chaotic dynamics.

Consider for example the RAM flow in Fig. 7 without re-orientation, i.e. \(\Theta_0 = 0\) in (49), causing \(\vec{u}\) to identify with base flow \(\vec{v}\) and, given \(\vec{v} = \vec{v}(x, y)\) here, resulting in \(\vec{u} = \vec{u}(x, y)\). This puts forth uniform axial “flow” \(\vec{w} = \vec{e}_z\) as symmetry and a Hamiltonian form of the cross-sectional flow following [41]. The same symmetry and Hamiltonian form emerge for diminishing rotation rate of the outer cylinder; tracers then only “feel” the averaged flow \(\vec{u} = N^{-1} \sum_{k=0}^{N-1} \vec{v}(r, \theta - k\Theta) = \vec{\bar{v}}(r, \theta)\) [83]. The impeller-driven steady flow in Fig. 18 becomes axi-symmetric, i.e. \(\vec{u} = \vec{u}(r, z)\), for tilt angle \(\alpha = 0\) and thus in a similar way leads to a symmetry \(\vec{w} = \vec{e}_\alpha\) and corresponding Hamiltonian form (52). Steady conditions preclude chaos in these flows for reasons mentioned before.

The time-periodic flow in Fig. 26(a) (left) due to re-orientations of the steady lid-driven Stokes flow in Fig. 23(a) about the \(z\)-axis accommodates a continuous symmetry as above yet in a highly non-trivial manner. Time-reversal and ordinary reflectional symmetries according to (40) about planes \(x = 0\) and \(y = 0\), respectively, impose the structure
\[
u_{r,z}(\vec{x}) = \bar{u}_{r,z}(r, z) \cos \Theta, \quad u_\theta(\vec{x}) = \bar{u}_\theta(r, z) \sin \Theta, \tag{58}
\]
on the steady base flow [136, 124]. This, via separation of variables of \(dF_{1,2}/dt\) following (42), yields two COMs, i.e.
\[
F_1(\vec{x}) = f_1(r, z), \quad F_2(\vec{x}) = f_2(r, z) \sin \Theta, \tag{59}
\]
that can be shown to relate to (58) through
\[
\frac{\bar{u}_r}{\bar{f}_2} = \frac{1}{r} \frac{\partial f_1}{\partial z}, \quad \frac{\bar{u}_z}{\bar{f}_2} = -\frac{1}{r} \frac{\partial f_1}{\partial r}, \tag{60}
\]
exposing \(f_1\) as the Stokes stream function of flow \((\bar{u}_r/\bar{f}_2, \bar{u}_z/\bar{f}_2)\) in the \(r\)-\(z\)-plane. This suggests an embedded Hamiltonian structure and associated continuous symmetry. Its isolation embarks on expressing kinematic equation \(d\vec{x}/dt = \vec{u}\) in terms of the curvilinear frame \(\vec{\xi} = (z, r, \Theta)\), with \(\Theta = \ln(\sin \Theta)\) substituting angular coordinate \(\Theta\), yielding
\[
\frac{d(r, z)}{dt} = \bar{u}_{r,z} \cos \Theta(\alpha), \quad \frac{d\alpha}{dt} = \frac{u_\theta}{\alpha}, \tag{61}
\]
where \(\alpha = J = r \tan \Theta\), \(\vec{e}_\alpha = \vec{e}_\theta\) and \(u_\alpha = \vec{u} \cdot \vec{e}_\alpha = \bar{u}_\alpha\) and introducing the rescaled flow \(\vec{u}^* = \vec{u}/F_2\) via (60) gives
\[
\frac{dz}{dt} = u^*_1 = \frac{\bar{u}^*_1}{f_2}, \quad \frac{dr}{dt} = u^*_2 = \frac{\bar{u}^*_2}{f_2}, \quad \frac{d\alpha}{dt} = \frac{u^*_\alpha}{\alpha} = \frac{\bar{u}^*_\alpha}{\bar{u}_\alpha}, \tag{62}
\]
as counterpart to (61), with
\[
\vec{u}^* = \left( -\frac{\partial f_1}{\partial r}, \frac{\partial f_1}{\partial z}, \frac{\bar{u}_\alpha}{\bar{u}_\alpha} \right) = \bar{u}^*(\xi_1, \xi_2), \tag{63}
\]
the corresponding flow (43) in the Cartesian \(\vec{\xi}\)-frame. Its independence of \(\xi_3\) signifies indeed a continuous symmetry and, in consequence, a Hamiltonian structure [46] exists. The underlying relation (57) follows in the Cartesian frame spanned by \(\vec{\xi}\) from vector identity (50) as before: flow \(\vec{u}\) commutes with field \(\vec{w}\) as in (55) and, given both fields are solenoidal, thus satisfy (57) for \(H = -f_1 = -F_1\). However, counterparts \(\vec{u}^*\) and \(\vec{w}^* = h_\alpha \vec{e}_\alpha\), or equivalently, \(\vec{u}\) and \(\vec{w}_1 = (h_\alpha/F_2) \vec{e}_\alpha\) in physical space are non-commutative and, instead, via property
\[
\vec{w} \cdot \vec{\nabla} \vec{u} - \vec{u} \cdot \vec{\nabla} \vec{w} = -\bar{u}(\vec{\nabla} \cdot \vec{w}), \tag{64}
\]
yield
\[
\vec{u}^* \times \vec{w}^*_1 = \vec{u} \times \vec{w}_1 = \vec{\nabla} F_1, \tag{65}
\]
from (56), since \(\vec{w}^*_2\) and \(\vec{w}_1\) are non-solenoidal. Both \(\vec{u}\) and \(\vec{u}^*\) satisfying (65) reflects the fact that they relate by a scalar.

\[27\] The coordinate transformation associated with the continuous symmetry in fact results in \(d\vec{\xi}/dt = \bar{u}(\xi_1, \xi_2, t)\) for kinematic equation (45) and \(J = J(\xi_1, \xi_2)\) [246]. Hence relation \(\vec{u} = \bar{u}/J\) implies \(\vec{u} = \bar{u}(\xi_1, \xi_2, t)\).
multiplication factor $F_2$ and thus have coinciding streamlines and, in turn, the same embedded Hamiltonian structure \(^{29}\).

COM $F_2$ in (59) emanates from interaction between $\vec{u}$ and a second (non-solenoidal) field $\vec{w}_2$ following (64), i.e.

$$\vec{u} \times \vec{w}_2 = \vec{\nabla} F_2, \quad \vec{w}_2 = \vec{w}_1 \vec{e}_r + \vec{w}_1 \vec{e}_z,$$  \hspace{1cm} (66)

with

$$\vec{w}_r = \frac{1}{\omega r} \frac{\partial f_2}{\partial z}, \quad \vec{w}_z = -\frac{1}{\omega r} \frac{\partial f_2}{\partial r},$$  \hspace{1cm} (67)

advancing $f_2$ as the Stokes stream function of $\tilde{w}_2$. The analogy with (60) implies an embedded Hamiltonian structure (46), with $H = -f_2$, in the “flow topology” of “flow” $\tilde{w}_2$. The latter emerges from essentially the same continuous symmetry as for axi-symmetric steady circular flows. Rescaled field $\vec{w}_2^* = (\hat{\omega} \vec{w}_2, \hat{\omega} \vec{w}_r, 0) = \vec{w}_2^*(r,z)$ in the reordered cylindrical frame $\tilde{\xi} = (z, r, \theta)$ namely commutes with $\vec{g} = \vec{r} \vec{e}_0$ following (55) and via (56) results in

$$\vec{w}_2^* \times \vec{g} = \vec{\nabla} f_2,$$  \hspace{1cm} (68)

assuming form (57) for $H = -f_2$. This, given streamlines of both $\vec{u}$ and $\vec{w}_2$ (and thus $\vec{w}_2^*$) are restricted to level sets of $f_2$ by virtue of (66), indirectly constitutes an embedded Hamiltonian structure for the fluid motion.

The above reveals that COMs $F_1, F_2$ following (59) can be reconciled with continuous symmetries according to (246), albeit in a non-trivial way. Solenoidal symmetries $\vec{w}$ directly commuting with $\vec{u}$ following (55) are non-existent in the lid-driven cylinder flow; COMs $F_1, F_2$ instead originate from interaction of $\vec{u}$ with non-solenoidal fields $\vec{w}_1, \vec{w}_2$ following (64) and thus via (56) lead to (55) and (66). Hence solenoidal symmetries $\vec{w}$ commuting with $\vec{u}$ as per (55) in fact constitute a subclass of systems that yield COMs via relation (57).

Relation (60) identifies the level sets of $F_1$ with surfaces of revolution of the level sets of Stokes stream function $f_1$, or equivalently, the streamlines of flow $(\vec{u}_1, \vec{u}_2)$ in the rz-plane. Conversely, this causes the projection of 3D streamlines in the rz-plane to coincide with the level sets of $f_1$ (136)(124). Axi-symmetry preserves COM $F_1$ in time-periodic flows involving reorientations of the bottom wall and the spheroids in Fig. 26(a) (left) and Fig. 27(b)(left) thus correspond with its level sets. COM $F_2$, on the other hand, exists only in the steady base flow and the intersections of its level sets with those of $F_1$ define the closed streamlines in Fig. 23(a).

The restriction of the dynamics to invariant surfaces (i.e. the spherical level sets of $H_1 = -f_1$) implies (in terms of the intra-surface coordinates) an alternative Hamiltonian structure, with $H_2^* = F_2$, to that associated with $H_2 = -f_2$ in the base flow. This is elaborated in Sec. 4.4.2 Hamiltonian $H_2$ directly (instead of indirectly) through $\vec{w}_2$ following (69) governs the intra-surface dynamics and, via the reorientation of the base flow, becomes non-autonomous in the time-periodic flow, thus leading to intra-surface Hamiltonian chaos as demonstrated in Fig. 26(a)(left).

The two COMS $F_1, F_2$, or equivalently, the autonomous Hamiltonians $H_1$ and $H_2^*$ in the steady base flow (by confining the dynamics to individual streamlines) implicitly define the full solution to kinematic equation (2) via $F_1, F_2 \tilde{x}(t) = F_1, F_2 (\tilde{x}_0)$. This renders the system fully integrable and, in consequence, entirely non-chaotic. The remaining single COM $F_1$ (or Hamiltonian $H_1$) in the corresponding time-periodic flow constitutes an only partial solution to (2), implying partially integrability, and permits 2D chaos within its level sets.

### 4.4.2 Flows with invariant surfaces

The flow topology of 3D unsteady flows with a single COM according to (42) is foliated into a continuous family of invariant surfaces defined by its level sets (as e.g. the spheroids associated with COM $F_1$ following (59)). The restriction of the Lagrangian dynamics to invariant surfaces implies an embedded Hamiltonian structure that may coexist with a possible Hamiltonian structure underlying these LCSs (as e.g. occurring in the cylinder flow).

Consider to this end kinematic equation (45) in a curvilinear frame $\tilde{\xi}_1 = (\xi_1, \xi_2, \xi_3)$ such that coordinates $(\xi_1, \xi_2)$ and $\xi_3$ are tangent and normal, respectively, to said surfaces (i.e. the latter coincide with coordinate surfaces of $\xi_3$). Restriction to invariant surfaces implies $u_3 = 0$ and thus via $\vec{\nabla} \cdot \vec{u} = \sum \partial u_i / \partial \xi_i = 0$ a solenoidal intra-surface 2D flow $(u_1, u_2)$ with, inherently, a Hamiltonian structure:

$$\frac{\partial u_1}{\partial \xi_1} + \frac{\partial u_2}{\partial \xi_2} = 0 \Rightarrow (\tilde{u}_1, \tilde{u}_2) = \left( \frac{\partial H}{\partial \xi_2}, -\frac{\partial H}{\partial \xi_1} \right).$$  \hspace{1cm} (69)

This identifies with vector potential (44) for $\tilde{A}_1 = 0$ and $H = \tilde{A}_1$, leading to Hamiltonian form (46) for the intra-surface dynamics described by $(\xi_1, \xi_2)$. Hamiltonian $H = H(\xi_1, \xi_2, t)$ can be interpreted in two ways. First, as the union of intra-surface Hamiltonians $H = H(\xi_1, \xi_2, \xi_3; t)$ parameterised by normal coordinate $\xi_3$. Second, as the global Hamiltonian $H = H(\xi_1, \xi_2, \xi_3; t)$ for the system “frozen” at a given time instance $t$. Here $\xi_3$ acts as a time-like variable reminiscent of the 3D steady duct and circular flows in Sec. 4.3, where $\xi_3$-wise progression through the 3D flow topology is similar to uni-directional “flow” $u_3 > 0$.

The aforementioned spheroids define the invariant surfaces in the flow topology of the time-periodic cylinder flow in Fig. 27(b)(left). Tracers are within these LCSs restricted to streamlines defined by intersections of level sets of COMs $F_1$ and $F_2$. This implies $H = F_1(\tilde{\xi}_1, \tilde{\xi}_2; \xi_3)$ as Hamiltonian in (69) for the corresponding base flow and a non-autonomous counterpart $H = H(\tilde{\xi}_1, \tilde{\xi}_2; \xi_3)$ constructed from its reorientation for the time-periodic flow. The typical Hamiltonian dynamics in “smaller” spheroids is demonstrated in Fig. 56(a) by the 3D (left) and corresponding intra-surface (right) stroboscopic map expressed in terms of the angu...
more chaotic upon progressing outwards in chaotic sea. The intra-surface dynamics tends to become topology consisting of (chains of) islands embedded in a nomena) precludes qualitative topological changes in closed trajectories, which (besides possible localised phenomena with the abovementioned 3D steady flows. The lat-
dynamics and this in fact reflects a fundamental differ-
zation in Fig. 27(b) into elliptic and hyperbolic segments.

The flow topology of the spherical tumbler for mixing of granular media in Sec. 3.3.2 for certain operating condi-
tions also consists of invariant spheroids, meaning that the intra-surface Hamiltonian dynamics within the spheroids of non-
 inertial (Re = 0) time-periodic cylinder flow following Fig. 26(a) (left).

Fig. 56 exposes a substantial variation in intra-surface dynamics and this in fact reflects a fundamental differ-
ence with the abovementioned 3D steady flows. The latter are ξ3-wise periodic and thus invariably accommodate closed trajectories, which (besides possible localised phenomena) precludes qualitative topological changes in ξ3-direction (Sec. 4.1.4). Hence e.g. the tubes in the RAM in Fig. 7(a). The flow topology of 3D unsteady flows with invariant surfaces as the time-periodic cylinder flow, on the other hand, is generically aperiodic in ξ3 and thus permits such changes. Hence the intra-surface dynamics in Fig. 56 and, inextricably linked with this, the partitioning of the periodic line in Fig. 27(b) into elliptic and hyperbolic segments.

The flow topology of the spherical tumbler for mixing of granular media in Sec. 3.3.2 for certain operating conditions also consists of invariant spheroids, meaning that the intra-surface dynamics shown in Fig. 28(b) (left) is of essentially the same Hamiltonian nature as in Fig. 56. Trans-
port studies using kinematic models for stroboscopic maps associated with 3D time-periodic flows reveal similar intra-
surface Hamiltonian dynamics and chaos for a variety of surface topologies [140]. This substantiates its universality.

### 4.5 Link with 3D volume-preserving flows and maps

Steady solenoidal fluid flows and the stroboscopic maps of time-periodic solenoidal flows belong to the broader class of volume-preserving flows and maps respectively. Such generic flows and maps are widely used for studies on 3D (chaotic) advection, since they typically rely on simple kinematic models that are easy to treat computationally and often even admit analytical investigations. The well-known ABC flow e.g. first demonstrated the emergence of chaos in 3D steady flows and its discrete counterpart enabled first investigations on RID shown in Fig. 27(a) [141, 249, 248]. Moreover, both the ABC flow and map offered insights into the mechanisms underlying the particle clustering in Fig. 21 [119, 120]. Similar kinematic models established the generic nature of Hamiltonian chaos within invariant surfaces as shown for the time-periodic cylinder flow in Fig. 26(a) (left) and the emergence of RIM upon their perturbation as per Fig. 27(b) [140, 149].

Important in the present context is that kinematic models for volume-preserving flows and maps generally do not satisfy momentum conservation and thus, inherently, can capture only kinematic features of 3D Lagrangian transport. This includes the above embedded Hamiltonian structures yet excludes the fluid-dynamical mechanisms that facilitate such behaviour. Necessary for said structures is namely only the vector potential of the flow following (44), which exists solely by the grace of solenoidality, in combination with a kinematic condition as e.g. uni-directional flow or a continuous symmetry. Actual occurrence of a kinematic condition for a given Hamiltonian structure, on the other hand, depends essentially on dynamic conditions governed by momentum conservation. The continuous symmetry underlying the intra-surface Hamiltonian dynamics within the spheroids of the time-periodic cylinder flow in Fig. 56 e.g. hinges on the particular flow structure [58] that, in turn, results from geometric symmetries imparted on the flow due to the linearity of the momentum equation in the Stokes limit. Hence comprehensive 3D transport studies on realistic fluid flows must at some point always employ descriptions that satisfy both mass and momentum conservation. Refer to [43] for further discussion of kinematic versus dynamic conditions.

Furthermore relevant is that volume-preserving maps in the present scope derive from continuous time-periodic fluid flows and thus incorporate their dynamics. A stroboscopic map therefore has an embedded Hamiltonian structure only if this already exists in the underlying flow. Conversely, an essentially non-Hamiltonian time-periodic flow cannot produce a stroboscopic map with Hamiltonian dynamics. The intra-surface Hamiltonian structure within the spheroids of the beforesaid time-periodic cylinder flow e.g. is a direct consequence of the presence of these LCSs in the base

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30Volume-preserving maps are also denoted “Liouvillian maps” [248].
flow and their invariance to reorientations of the driving wall. However, time-periodic flows thus constructed from reorientations of an essentially Hamiltonian base flow must not necessarily adopt the Hamiltonian nature. Said time-periodic cylinder flow for \( Re = 100 \) is essentially non-Hamiltonian, as reflected by the emergence of an isolated periodic point with global manifolds shown (Fig. 26(b)), notwithstanding the Hamiltonian behaviour of the base flow (Fig. 23(c)).

### 4.6 Towards essentially 3D (chaotic) dynamics

#### 4.6.1 Conditions for chaos

The inherent Hamiltonian structure of 2D flows makes a non-autonomous Hamiltonian \( H = H(x, t) \) a necessary kinematic condition for chaos (Sec. 4.2). Essentially 3D steady flows (i.e., dependent on all spatial coordinates) are in their simplest form dynamically equivalent to a 2D unsteady flow and thus a non-autonomous Hamiltonian system (Sec. 4.3). This has the fundamental implication that any 3D (un)steady flow meets the necessary kinematic criteria for chaos. Whether (and to what degree) chaos actually happens is dictated by momentum conservation. The structure of the momentum equation implies that realistic laminar flows (i.e., subject to both viscous and inertial forces) generically lack a universal dynamical restriction to chaos. Hence such flows in principle always exhibit chaos in (at least) some regions and typically have a flow topology consisting of LCSs surrounded by chaos as illustrated by the RAM in Fig. 7. Global restrictions to chaos occur only for case-specific dynamic conditions as e.g. the symmetry-induced confinement to spheroids and, inherently, essentially 2D (chaotic) dynamics, in the non-inertial time-periodic cylinder flow following Fig. 26(a) (left).

The necessary **topological condition for chaos** is the transversal intersection of stable and unstable manifolds associated with hyperbolic points/lines/trajectories as per Sec. 4.1. Such interactions are denoted homoclinic and heteroclinic if involving (un)stable manifolds of the same or two different hyperbolic LCSs, respectively. Fig. 5(a) e.g. shows the homoclinic intersection of the stable (blue) and unstable (red) manifolds of the central hyperbolic point (cross) in the archetypal 2D time-period flow; these LCSs furthermore interact heteroclinically with manifolds (not shown) of the two adjacent hyperbolic points. The actual mechanism leading to exponential stretching of fluid parcels and, in consequence, chaotic advection is the accumulation of intersections upon approaching a periodic point along the stable/unstable manifold during forward/backward progression in time.

33 This more formally follows from the Poincaré–Bendixson theorem, which states that continuous dynamical systems – here kinematic equation – admit chaos only for a corresponding phase space – here the space-time domain – that is at least 3D. 3D flows evidently meet this condition.

32 Steady inviscid flows (i.e., so-called “Euler flows”) are, in stark contrast, generically non-chaotic due to the restriction of Lagrangian dynamics to level sets of the Bernoulli function \( \alpha = p + \frac{1}{2} \rho g z + \vec{u} \cdot \vec{v} / 2 \), with \( p \) the pressure, \( \rho \) the density and \( g \) the gravitational acceleration acting in \( -z \) direction.

#### 3D steady flows

Transversal manifold interaction may in 3D steady flows occur via various scenarios. This generically excludes stagnation lines, since these entities are atypical of realistic flows. The elliptic stagnation lines that e.g. shape the idealised streamline topology of the cylinder flow in Fig. 23(a) and droplet flows in Fig. 36(a) for basically any perturbation give way to a hyperbolic focus and elliptic closed streamlines, respectively, in the corresponding non-ideal (i.e., realistic) topologies shown in Fig. 23(c) and Fig. 36(b). Hence only isolated stagnation points and closed/periodic hyperbolic streamlines are of practical relevance. (This includes critical points of the skin-friction field on grounds of their equivalence to hyperbolic nodes; Sec. 4.1.) The 1D manifolds of stagnation points can immediately be ruled out from transversal interaction scenarios; they can intersect 2D manifolds namely only at stagnation points. However, this prevents progression of tracers along these intersections and thus violates the nature of manifolds. This constraint e.g. prohibits intersection of the 1D and 2D manifolds of the skin-friction field in Fig. 47(b).

Heteroclinic interaction between stagnation points and hyperbolic closed streamlines is possible via 9 scenarios following (250) (Fig. 22). Heteroclinic transversal interaction is restricted to 2D manifolds and, given the latter are stream surfaces, thus defines streamlines (denoted “heteroclinic trajectories”) with corresponding dynamics as follows:

- **Point–point interaction** (Sec. 4.1 of 250): a single heteroclinic trajectory connects both points; exponential stretching is absent and thus dynamics are non-chaotic.
- **Point–line interaction** (Sec. 4.3 of 250): two heteroclinic trajectories emerge from the point and asymptotically spiral towards a limit cycle defined by the closed streamline; accumulation of windings of the spiralling trajectories upon approaching the closed streamline and elongation of the 2D manifold of the line along the 1D manifold of the point signifies exponential stretching of both 2D manifolds and thus chaos.
- **Line–line interaction** (Sec. 4.4 of 250): the intersecting (topologically cylindrical) manifolds yield two heteroclinic trajectories that asymptotically spiral both forward and backward in time to limit cycles defined by the closed streamlines; accumulation of windings upon approaching the latter again produces chaos. The cylindrical shape of the manifolds in fact implies intermediate heteroclinic trajectories (i.e., “Birkhoff’s signature”) that, through the formation of so-called “lobes” (251), greatly increase the rate of stretching.

The 2D manifolds of single closed streamlines admit homoclinic intersection similar to the heteroclinic line–line interaction (Ch. 5 in 250). These findings put forth point–line
and, by their topological complexity, particularly line–line interactions as “drivers” of chaos in 3D steady flows. However, 2D manifolds may also impede chaos in their capacity as barriers to 1D/2D manifolds of the same stability.  

Hyperbolic periodic streamlines are also closed in the sense of reconnecting via a periodic inlet-outlet and thus admit the same point–line and line–line interactions as elaborated above. (Heteroclinic trajectories here “spiral” towards the limit cycle(s) via repeated reconnection.) However, heteroclinic interaction between closed and periodic streamlines is impossible (Conjecture 1 in [86]). Moreover, point–line interaction is possible only if the 2D stable (unstable) and 2D unstable (stable) manifolds of the point and streamline are exclusively interacting with each other (which follows from Conjecture 3 in [86]). These topological constraints are e.g. crucial to the emergence of a net throughflow region in periodic duct flows and, in consequence, the persistence of an embedded Hamiltonian structure in this subregion [86].

### 3D unsteady flows

Topological freedom – and, inherently, the scenarios for transversal interaction – increases substantially for 3D unsteady flows. The 1D manifolds of isolated periodic points in stroboscopic maps can, for instance, intersect with 2D manifolds, thus permitting the homoclinic transversal manifold interaction for the hyperbolic focus in the time-periodic cylinder flow in Fig. 26(b).

Unsteady conditions furthermore allow for non-trivial point–point interactions leading to chaos. Consider e.g. the ideal buoyant droplet in Fig. 26(a)(left); its spherical surface and internal (black) axis are the merged 2D and 1D manifolds, respectively, of hyperbolic nodes at the axis endpoints. Aperiodic unsteady perturbation triggers transversal interaction of the manifolds of DHTs (aperiodic counterparts of hyperbolic nodes) following Fig. 40 and thus chaotic advection due to lobe formation akin to transversal line–line interactions. Similar behaviour occurs upon subjecting a droplet immersed in a steady swirling flow (its surface and axis emanate in a similar way as before from interacting hyperbolic foci) to time-periodic perturbations. This is demonstrated in Fig. 15 of [251] (originally from [252]).

The above examples concern chaos directly resulting from relaxation of topological restrictions of 3D steady flows. However, insights into the full set of transversal manifold interactions possible under essentially 3D unsteady conditions remain incomplete.

#### 4.6.2 Conditions for 3D dynamics

Lagrangian transport in 2D solenoidal flows is governed by Hamiltonian mechanics due to the Hamiltonian structure (41) of kinematic equation (2) (Sec. 4.2). This notion enables rigorous distinction of essentially 2D dynamics from truly 3D dynamics in 3D solenoidal flows. The former namely implies an embedded Hamiltonian structure according to Sec. 4.3 and Sec. 4.4 in kinematic equation (2) relating to the physical flow via (formal) transformation (11); the latter implies (local) absence of such a structure.

The essentially 2D nature of the dynamics imparted by an embedded Hamiltonian structure suggests that the flow topology of such 3D flows is basically a “3D extrusion” of the typical Hamiltonian topology of 2D flows illustrated in Fig. 5(a) in the $\xi_1$-direction of an appropriate curvilinear frame (43) following Sec. 4.3 and Sec. 4.4. Thus islands become tubes/tori centered on elliptic lines/closed trajectories defined in Sec. 4.1.3 and Sec. 4.1.4 surrounded by chaotic streamlines/trajectories as e.g. for the RAM in Fig. 7. Hence the emergence of such LCSs in the various configurations in Sec. 3 are clear indicators of (local) intrinsically 2D Hamiltonian dynamics. The breakdown of tubes/tori into chaos is described by 3D counterparts to the beforementioned KAM and Poincaré–Birkhoff theorems (Sec. 4.2) [253, 254].

The above advances (local) departure from or a priori absence of an embedded Hamiltonian structure in (2) as necessary kinematic condition for truly 3D (chaotic) dynamics in 3D (un)steady flows. Two important mechanisms in this respect are elaborated below: singularities and resonances.

#### Singularities

Actual local breakdown of an embedded Hamiltonian structure manifests itself mathematically in singularities in the corresponding transformation (11). Consider as an instructive example the 2.5D duct flows introduced before, which (for $\xi = x$) adopt both Hamiltonian form (46) and (50) via transformation (47) and (49), respectively, due to the axial component of the base flow meeting $v_\perp(x, y) > 0$ (Sec. 4.3.2). The latter changing sign at $v_\perp = 0$ implies a non-monotonic relation $z(t)$, prohibiting its global inversion, and thus a singularity in transformation (47). Vanishing $u_3 = v_\perp$, similarly, yields a singular Jacobian $J_3 = |\partial z/\partial \xi_1| = -|\partial z/\partial \xi_2| = u_1$, with $\xi = (\xi_1, \xi_2, x)$, for transformation (49). Hence both cases only admit a local transformation into the respective Hamiltonian forms in sub-
regions \( v_2 > 0 \) and \( v_3 < 0 \) that, given continuity, are separated by (a) smooth surface(s) \( v_3 = 0 \). Superposition of e.g. the previous 2.5D RAM flow with a uniform axial backflow of relative strength \( 0 \leq \alpha \leq 1 \), i.e.

\[
v_z = u_z = 2U(1 - r^2) - 2\alpha U = 2U(r_z^2 - r^2),
\]

(70)

results in flow reversal from \( v_z > 0 \) to \( v_z < 0 \) at the cylindrical surface with radius \( r_z = \sqrt{1 - \alpha} \). Transformation \( (51) \), again using \( \vec{u} = \vec{u}/U \), gives \( J_z = 2(\vec{r}_z^2 - r^2) \) and indeed becomes singular at \( r = r_z \), implying a partitioning of the global dynamics into two coexisting Hamiltonian systems. Property \( v_z = 0 \) thus precluding a single global Hamiltonian structure, despite local dynamics retaining their Hamiltonian nature, is already an essentially 3D situation. Tracers generically namely can cross surfaces \( v_z = 0 \) and, within one global flow, switch between local Hamiltonian systems.\textsuperscript{34}

Partitioning into multiple Hamiltonian systems and/or restriction of Hamiltonian dynamics to subregions may furthermore stem from singularities in coordinate transform \textsuperscript{43}. Periodic duct flows, by expression in the curvilinear frame aligned with the 3D streamlines according to \( \textsuperscript{86} \), e.g. in principle always meet condition \( u_3 > 0 \) necessary for a non-singular transformation \textsuperscript{49}. However, stagnation points and (further) topological complexity of the streamlines often rule out a single global transformation \textsuperscript{43} and thus restricted embedded Hamiltonian structures to subregions admitting non-singular \( G \). Topological considerations strongly suggest that periodic duct flows accommodate at least one such subregion, viz. the beforementioned net throughflow region (possibly partitioned into multiple throughflow regions). Hence absence of a single global Hamiltonian structure again reflects an essentially 3D situation. Moreover, the net throughflow region (though itself accommodating Hamiltonian dynamics) may assume a complex structure particularly in partitioned-duct flows. Stagnation points and corresponding manifold interactions in the stagnation area of the T-shaped micro-mixer (Fig. 15) are e.g. likely to have a major impact on the dynamics and structure of the (multiple) throughflow region(s).

Generic 3D (un)steady flows are likely to develop singularities in the transformation from physical to canonical space as well as in that from the Cartesian to an appropriate curvilinear frame – causing breakdown/restriction of (local) Hamiltonian structures – as exemplified above. Moreover, the number and/or region of influence of singularities generally grow with increasing departure from an originally Hamiltonian structure and thus progressively diminish the latter in favour of essentially 3D dynamics. The global topological complexity of the transversally-interacting 1D/2D manifolds of the isolated periodic point in the time-periodic cylinder flow for \( Re = 100 \) in Fig. 26(b) e.g. strongly suggests full breakdown of the Hamiltonian structure of the corresponding non-inertial limit (\( Re = 0 \)) in Fig. 56.

\textbf{Resonances} Resonances are a further mechanism that may lead to 3D dynamics by causing essentially 3D departures from 2D Hamiltonian dynamics. This may promote 3D chaos as well as formation of intricate LCSs as demonstrated in Fig. 27 by RID and RIM, respectively. The essence of these fundamentally distinct resonance phenomena is exemplified below by way of simplified maps.

RID may occur in (local) flow topologies consisting of tori as in Fig. 27(a) (left) described by trajectories spiralling around the toroidal axis. The Lagrangian motion is qualitatively similar to the helical trajectories along concentric tubes in a cylindrical periodic duct following

\[
r_{p+1} = r_p, \quad \theta_{p+1} = \theta_p + \omega, \quad z_{p+1} = z_p + \gamma,
\]

(71)

with \( \omega = \omega(\tilde{u}_0) \) and \( \gamma = \gamma(\tilde{u}_0) \), the period-wise azimuthal and axial displacement, respectively. The transverse (xy-wise) mapping is identical to the 2D elliptic mapping in (28) and \((\theta, z)\) represents the local poloidal/toroidal coordinates \((\phi, \varphi)\) following \textsuperscript{13}. Reconnection – and thus resonance – occurs for \( \omega \) and \( \gamma \) forming rational fraction \( \omega/\gamma = m/n \) as per \textsuperscript{13}. Generically \( \omega = \omega(r) \) and \( \gamma = \gamma(r) \), meaning that tori are either entirely non-resonant or resonant. Non-resonant tori survive weak perturbations according to the (3D counterpart of the) KAM theorem; resonant tori disintegrate following the corresponding Poincaré-Birkhoff theorems due to violation of the “averaging principle” first mentioned in Sec. 3.3.2 \textsuperscript{243, 254}. (This violation in fact occurs both for “true” resonance and slow-down of motion along trajectories to the same order of magnitude as the perturbation \textsuperscript{56}).

Such torus-wise survival or breakdown is the “conventional” Hamiltonian route to chaos. Fig. 58(a) gives 3 neighbouring trajectories (distinguished by colour) in a periodic duct of unit radius and length \( 2\pi \) starting at the inlet \( z = 0 \) (bottom) from the marked positions on a typical resonant torus for \( \gamma = 0.3 \) and \((m, n) = (2, 3)\); thus reconnection occurs after 3 cycles and single trajectories appear as 3 segments. This torus disintegrates for any perturbation in an actual flow. Irrational \( \omega/\gamma \) (e.g. \( \omega/\gamma = \sqrt{2} \)) yields a non-resonant (and surviving) torus densely filled by the trajectories (not shown).

However, 3D flows also admit resonances in subregions within tori, causing only local interruption of the averaging process and, in consequence, local defects in tori (rather than full breakdown). This may occur upon weak perturbation of a flow topology consisting entirely of reconnected
trjectories following Fig. 58(a) and, inherently, resonant tori. Consider for illustration an azimuthal displacement \( \omega(r, \theta) = \omega_0(r) + \varepsilon \sin(\theta) \) such that \( \omega_0 \) gives a rational fraction with \( \gamma \) as before and \( \varepsilon = 0 \) thus represents the unperturbed state. Perturbation \( 0 < \varepsilon \ll \omega_0 \) breaks the rationality, except for isolated rational trajectories starting at \( \theta_0 = 0 \) and \( \theta_0 = \pi \), and thus triggers dynamics as shown in Fig. 58(b) for \( \omega_0 / \gamma = m/n \), with \( (m, n) \) and \( U \) as before, and \( \varepsilon = 5 \times 10^{-4} \): azimuthal drifting of tracers (black/blue) away from the rational trajectory (red). The drifting tracers delineate the torus segments partitioned by the rational trajectory; local defects develop at the latter and enable random jumps between tori and thus promote global dispersion of tracers (i.e. RID) as demonstrated in Fig. 27(a) (right) and observed or suspected in a range of other (un)steady 3D flows discussed in Sec. 3.

RID occurs if motion along unperturbed trajectories becomes of the same order as the perturbation due to resonance or local slow-down. This gives perturbed motion that is comparable in all coordinate directions and, in consequence, no longer restricted to the original tori. Behaviour akin to RID occurs if the (otherwise weak) perturbation has localised peaks of the same order as the typical motion along the unperturbed trajectories. (Such peaks can e.g. develop in shear layers in granular media or non-Newtonian fluids.) The effect is similar as before: perturbed motion comparable in all coordinate directions that again causes local breakdown of tori. Thus both resonance/slow-down and perturbation peaks enable tracer dispersion across tori; differences primarily concern the intensity in that dispersion by former and latter mechanism is relatively slow and fast, respectively.

RIM may occur upon perturbation of flow topologies accommodating invariant surfaces “pierced” by a periodic line(s) as e.g. the spheroids in the Stokes limit \( Re = 0 \) of the time-periodic cylinder flow in Fig. 27(b)(left). Fluid inertia induces drift across the spheroids and yields Lagrangian motion transverse and parallel to the periodic line qualitatively similar to that near periodic points in 2D systems as per (26) and an axial uni-directional motion, respectively, i.e.

\[
x_{p+1} = F x_p, \quad z_{p+1} = z_p,
\]

with \( x = (x, y) \). Key to RIM is a \( z \)-dependent map \( F = F(z) \) that smoothly changes its qualitative nature, determined by \( J_1 = \text{tr}(F) \), at some axial position \( z_0 \) from \( |J_1| < 2 \) to \( |J_1| > 2 \), thus transiting from elliptic \((z < z_0)\) to hyperbolic \((z > z_0)\) dynamics, respectively. This happens in actual flows for (perturbed) periodic lines partitioned into elliptic \(|J_1| < 2\) and hyperbolic \(|J_1| > 2\) segments and thus merges tubes and shells – created on former and latter segments, respectively, by the averaging process – into intricate LCSs as demonstrated in Fig. 27(b)(right).

The degenerate points \(|J_1| = 2\) separating elliptic and hyperbolic segments (denoted “parabolic points”) act as resonances by terminating and disconnecting the averaging processes that underlie the tube and shell formation and thus pave the way to their merger (i.e. RIM). Perturbations of the cylinder flow generally yield a uni-directional drift along the periodic line yet isolated periodic points may also emerge near the parabolic point as observed for some instances of RIM in the sphere-driven time-periodic flow studied in [149]. However, absence/presence of such periodic points is non-essential for RIM. The decisive kinematic condition is a qualitative change in dynamics due to segmentation of periodic lines.

Resonances as exemplified above occur for small departures from an intact embedded Hamiltonian structure comprising of complete LCSs and, unlike singularities, thus affect Lagrangian dynamics only under such conditions. Moreover, the topology of the LCSs is crucial for RID and RIM; former and latter phenomenon, if occurring, are inextricably linked to toroidal and spheroidal invariant surfaces, respectively Sec. 3.3.2. Chaotic advection resulting from RID and RIM (via e.g. “leaky shells; Sec. 3.3.2, therefore is relatively weak compared to a system far away from a Hamiltonian state as e.g. a flow topology dominated by manifolds of isolated periodic points following Fig. 26(b)). Hence RID and RIM are (in a practical sense) primarily precursors to global chaos and of only limited use for (industrial) mixing purposes. The delicate and non-trivial LCSs formed by RID and RIM may, on the other hand, find practical applications other than mixing. Large departures from embedded Hamiltonian structures are in any case imperative to accomplish truly 3D (chaotic) dynamics yet insights into the response of 3D flow topologies – and their potential applications – to progressive perturbations are far from complete.

5 Commercialisation of chaotic advection

This section considers how the extensive scientific knowledge and insights of 3D Lagrangian transport developed over the last 30 years have migrated to industrial practice and technology. The success of translation is measured by the uptake of fluid chaos in patentable ideas, new devices that move beyond university laboratories and ultimately to the commercialisation and use of these ideas and devices in application. However, the process of taking a novel idea to a successful outcome is an arduous journey that is familiar to all who have attempted it. Although the practical uses of chaos are myriad, there is a general reluctance in traditional industries to adopt new technologies that are seen as untried and untested. Even in new industries and applications, the tendency to fall back on existing technology is strong.

Obtaining reliable information on commercialisation of
fluid chaos has proven difficult due to the sensitivity around intellectual property and a lack of clarity around ownership of ideas. Consequently, primarily the patent literature is examined. Although an application for patent protection is often a precursor for commercial applications, patent activity must be viewed cautiously from this viewpoint and can be seen only as an indicator. Most patents are never successfully commercialised and, conversely, some commercially successful ideas are never patented. This section finishes with a brief discussion of a number of technologies that have progressed to commercialisation.

5.1 Patents
An internet search using Google Patent (https://patents.google.com) with a search term of "(chaos OR chaotic) AND (mixer OR mixing) AND fluid" in October 2018 yielded a list of approximately 560 patents. This list was consolidated by removing duplicate patents in different jurisdictions, patents for identical devices claimed for different applications, devices that had nothing to do with fluid mixing and devices that claimed to use chaotic mixing as a loose terminology for turbulent mixing. Patents that mentioned in passing the use of unspecified "chaos/chaotic mixers" as one of many possibilities in a list of mixers were also discounted. Thus 293 patents remained and consisted of 173 patents for "stand alone" 3D chaos mixers and 120 patents in which chaotic mixing was explicitly mentioned as part of at least some embodiments of the patent. This latter group included patents where specific types of chaotic mixers were mentioned as well as those in which an unspecified "chaotic mixer" was explicitly mentioned as an integral part in at least some embodiments.

The cumulative number of chaotic mixing patents as a function of year is shown in Fig. 59 where mixer patents are clearly separated from those in which chaotic mixing is claimed only as part of the invention. The year plotted in this figure is based on the priority date, not the filing, publication or grant date because the priority date determines which is the “first application” in practice. The median time difference between priority and publication dates in this list was approximately 21 months, although time differences as high as 15 years were noted. The early history consists of patents that are fluid mixers only. In 1997 the first patent appears that specified the use of chaotic mixing as just part of the invention and by around 2005 the annual number of new mixers versus the use of chaos “in part” is approximately equal.

Several of the early patents are worth a special mention. Although the published patent record of chaotic advection begins in 1994 with the publication of four patents, the majority of these had earlier priority dates.

The patent with the earliest priority date (June 1992), and hence the honour of the first patented use of chaotic advection, belongs to Sen and Chang [256] of the Gas Research Institute in Illinois. Entitled “Process and Apparatus for enhancing In-Tube Heat Transfer by Chaotic Mixing”, their device exploits a twisted-pipe type of the duct flows following Sec. 3.2 used for heat exchange. The patent was allowed to lapse in 2002 and no record has been found regarding its use in application. With a priority date just 5 months later (Nov 1992), Castelain, Le Guer and Peerhossaini [257] also patented a twisted-pipe device for heat exchange. However, the record indicates the patent was disallowed and was withdrawn in 1998, possibly because the claims in Sen and Chang [256] encompassed those in [257].

Mackley, Skelton and Smith [258] patented a mixing vessel with a priority date in March 1993. It consisted of a duct with radial baffles that divides the tube into chambers and uses oscillatory flow (the first example of patenting time-dependent chaotic flow) to generate time-periodic separated flow with crossing streamlines in different phases of the period (similar to the reorientation of a base flow in the time-periodic cylinder flow in Fig. 26) and mixing throughout the tube. The record suggests this patent did not make it into the PCT stage and was abandoned in 1998; however, the technology has progressed as we detail later.

Tjahjadi and Foster of the General Electric company patented modifications to extruder screws (also belonging to the duct flows following Sec. 3.3) that generated chaotic mo-

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Fig. 58. Resonance demonstrated by 3 trajectories in a periodic cylindrical duct starting at the inlet $z = 0$ (bottom) from the marked positions: (a) fully resonant torus for $\omega = m\gamma/n$, $\gamma = 0.3$ and $(m,n) = (2,3)$; (b) isolated resonant trajectory (red) separating torus segments delineated by drifting tracers (black/blue).

Fig. 59. Cumulative numbers of published chaotic mixing patents by priority year and category. Top curve is total patents, the middle curve is stand-alone mixers and the bottom curve is use of chaotic mixers as part of the invention in at least some embodiments.
tion in polymer melts to improve mixing and heat/mass transfer characteristics of the extruder. This patent had a priority date of September 1993, and appeared again to have been maintained by General Electric until 2008 when it as assigned to Sabic Innovative Plastics B.V. of the Netherlands.

Also having a 1993 priority date, Logan [259] filed on a device to be inserted into ducts. Logan’s device has three long rods welded into an open triangular prism with sine wave baffles welded to the faces of the triangle, which the patent claims produces stretching, folding and chaotic mixing for viscous liquids. Most curiously, down the center of the triangle are acoustic horns claimed to provide additional mixing. We have been unable to find evidence that Logan’s device was ever tested and although payment was continued until 2006, the fate of the technology is unknown.

The only chaotic mixing patent with a 1994 priority date was that of Kwon and San [260] of the Pohang Iron and Steel company for another extruder screw that generated chaotic motion in polymers melts. Once again this patent was allowed to lapse in 2006, and no information has been found as to its commercialisation.

The early history of chaotic mixing patents is entirely filled by devices that are termed here “macro-scale” devices, operating at length scales from centimetres to metres. This changed in July 1997 with the first micromixing patent by Evans, Liepman and Pisano [261]. Unlike most subsequent micromixer designs, this device is not a duct. Instead it has a thin, planar chamber, into which fluid streams are pumped from alternating directions, making this system a container flow according to Sec. 3.3. The time-periodic pulsation and their reorientation accomplish the mixing within the chamber. Fee payments on this patent were up to date in 2011.

The cumulative number of chaotic mixing patents is shown in Fig. 60, this time distinguishing between macro-scale and micro-scale patents. Again the year plotted in this figure is based on priority date. The most noticeable feature of this plot is that since approximately 2002, the number of patents filed annually at each scale is approximately constant, with micro-fluidic patents being filed approximately three times more frequently as macro-scale. A significant part of the micro-fluidic abundance is due to the “chaos as part of the working principle” patents seen in Fig. 59. With very few exceptions, these “chaos in part” patents involve the use of micromixers. Clearly the drive for miniaturisation and the concomitant drastic reduction in Reynolds number have opened up the practical utilisation of diverse surface forces drive fluid motion on the micro scale, forces that are negligible at the macro scale.

Classification of patents according to the flow categories described in Sec. 3 is also instructive and is shown in Table 1. Here screw extruders (10% of total patents) have been categorised as “partitioned-duct flows”, the majority of wall-driven container flows also involved unsteady driving and were categorised as “unsteady container flows” and that different inventions for mixing inside droplets were lumped together due to the small numbers – the majority (4% of total patents) being background-flow driven. At 40% of total patents, open-duct flows have a clear majority, with approximately 50% of open-duct flow patents involving a means to drive a spatially or temporally varying cross flow either via patterned walls (e.g. a herringbone pattern) or via direct forcing (electro-kinetic, magnetic, varying wettability, mechanical). The other 50% of open-duct flow patents involve the use of duct geometry including 2D and 3D twisted and serpentine channels and/or wall baffles and/or varying channel cross section to generate secondary flows and streamline chaos. Worth noting is that very few fluid chaos patents involve flows that are spatially fully 3D, mirroring the nascent state of knowledge in such systems [43].

Table 1. Patent classification by percentage in terms of the flow categorisation following Sec. 3.

| Category                                      | Percentage |
|-----------------------------------------------|------------|
| Open-duct flows (Sec. 3.2.2)                  | 40%        |
| Partitioned-duct flows (Sec. 3.2.2)            | 13%        |
| Unsteady container flows (Sec. 3.3.2)          | 8%         |
| Branching and connecting (Sec. 3.2.2)          | 8%         |
| Forced container flows (Sec. 3.3.2)            | 7%         |
| Mixing inside droplets (Sec. 3.4.2)            | 7%         |
| Other (various)                                | 17%        |

5.2 Commercialisation attempts

The following presents a brief discussion of a few technologies that have attempted, and in some case succeeded, in commercialisation. Making connections between patents, inventors and current commercial activity has proven difficult, especially for patents that include chaotic mixing as part of the process. There are likely a number of other commercially available devices that have not been uncovered for this
5.2.1 Kenics mixer

The Kenics mixer [92] is an an-line mixer with internal twisted, offset plates. It has been a commercial success for Chemineer Inc. (chemineer.com) since 1965 and has spawned numerous variations on the original theme (PPM, SMX, Quatro and a number of micro-fluidic equivalents). The inventors correctly identified the secondary flows induced by the twisted elements and the split-and-recombine nature of the flow (Sec. 3.2) as the source of effective mixing (“This results in an eddy current motion in each partial stream, which causes some mixing of components ... As the fluid meets the upstream edge of the second element it is forced to split again ...”). Although the device has been demonstrated numerous time to mix via the process of Lagrangian chaos, e.g. [262], the patent appeared well before the concept of Lagrangian fluid chaos had been described. Regardless, it is one of the stand-out examples of fluid chaos in practical application. The range of similar-style mixers to appear after the original patent lapsed points towards the difficulty in achieving successful commercialisation in the allowable 20-year window. It also suggests that opening up a concept to other minds provides the freedom to develop alternative, and potentially better, embodiments of the idea that could contribute to longer term success.

5.2.2 Oscillating Baffled Reactor

The patent by Mackley, Skelton and Smith [258] for “Processing of mixtures by means of pulsation” resulted a processes later termed Oscillatory Flow Mixing (mackleymackley.com/innovation/oscillatory-flow-mixing) which is embodied in the Oscillatory Baffled Reactor (OBR). Although the original patent lapsed in 1998, at least two companies are commercialising the concept, Cambridge Reactor Design, (with the “Rattlesnake” continuous flow reactor (cambridgereactordesign.com) and NiTech Solutions Ltd. (nitechsolutions.co.uk) with chemical reactors and crystallisation vessels. Each company has more recent separate patents covering different aspects and applications of OBRs. NiTech Solutions has had commercial units operating in pharmaceutical manufacturing since 2007[39]. The advantages of the technology are clear when it is considered that one 2m³ NiTech reactor operating at 100³ l/min has replaced two 150 m³ batch stirred tank reactors in one application. Other NiTech installations in a range of chemical manufacturing plants have been completed and around 50 lab-scale reactors have been installed in laboratories for pharma and chemical companies as well as in universities. The consistent message to emerge from installations of this type of reactor technology is that it makes the correct product reproducibly, creates less waste, there are less downstream unit operations required for separation and the process is generally greener and safer as a result.

5.2.3 Rotated Arc Mixer

As introduced in Sec. 3.2, the Rotated Arc Mixer (RAM) is an in-line mixer without internal baffles designed on the basis of scientific insights into Lagrangian transport and the notion of chaotic advection (refer to www.youtube.com/watch?v=j0owt8XD6xM). The design was determined using a priori numerical simulation and the images shown in Fig. 5.2 are taken from the first experiment undertaken in the device in 1998. The poor and good mixing conditions and dye trace injection locations were identified before the experiment was run to highlight the robustness of the design methodology. The RAM can substantially outperform conventional inline mixers in terms of energy consumption and mixing quality, especially for highly viscous materials or materials in which fouling and scaling could rapidly clog internal mixing elements.

The elegance of the idea earned it recognition as one of the top designs in the “AIChE 10x Design Challenge” [263] in 2010. Two patents have been granted, one for flow mixing [264] and one for heat exchange [265]. Confidential trials of a full-scale RAM were undertaken in a plant owned and operated by a multinational food manufacturer. The outcome of the trial showed that the continuous-flow RAM increased line productivity by 25% (by fully accomplishing the desired material transformation so that downstream equipment could be operated at capacity) and reduced mixing energy by 95%, replacing two conventional mixers. Further, a sensory evaluation panel concluded that the the RAM produced product had a “step change” improvement in product quality. Despite the apparent benefits of the RAM, implementation has not progressed beyond trials. This is a case-in-point of where a technology has been proven superior in application yet the conservatism inherent in large, traditional industries still results in a reluctance to adopt this.

Rights to develop the technology currently reside with Tasweld Engineering Pty Ltd. (tasweldengineering.com.au). Although the patents were allowed to lapse in 2018, in the case of the RAM, knowledge of the underlying Lagrangian transport is essential when designing a unit for a given application. It is this know-how – much of it encapsulated in this review – that will result in success or failure, not a knowledge of how to manufacture the physical device. This statement will be true of most devices based on Lagrangian chaos.

5.3 The future of commercial application

The approximately linear increase in fluid chaos patents year-on-year suggests that the technology field is still in the ascendancy or growth phase of the innovation S-curve. The difficulty in identifying successful commercialisations suggest it is likely in early growth. Significant take-off will require existing mixing processes to be increasingly replaced by those based on 3D deterministic Lagrangian chaos or will require emerging technologies that are created with fluid chaos as an integral part of their design.

Although some industries have used chaotic mixing devices for decades (for example screw extruders in polymer processing), most have not. The experience of the authors,
their colleagues and many companies they have talked to indicates there is a reluctance to utilise ideas and equipment that has not already been proven in other similar applications, even when pilot-scale testing has proven the utility and improved performance of fluid chaos in their applications. The difficulty in bootstrapping the use of fluid chaos is perhaps particularly disappointing given that it is generally possible to accurately predict the underlying deterministic flow which in turn allows good designs to be provided a priori that do not require significant iteration.

The proliferation of mixers spawned by the concepts embodied in the Kenics mixer might also suggest that a rush of commercialisation may arise once original patents have lapsed. This is potentially good news for processes where chaos has advantages, but less beneficial for the inventors and their financial backers. However, it seems likely that fluid chaos will make its impact felt in emerging technologies where it can be built in from the ground up. Indeed this appears to be the case in micro-fluidic applications, a large number of which require the use of 3D Lagrangian chaos in order to be viable. “Watch this space” seems to be an appropriate concluding instruction.

6 Concluding remarks

The scope of this review is transport and mixing of scalar quantities such as additives, chemical species, heat and nutrients in realistic three-dimensional (3D) fluid flows under laminar flow conditions. This is motivated by its ubiquity in many systems and processes both in industry and Nature. Transport and mixing is considered in terms of the Lagrangian motion of fluid parcels (“advection”) and thus admits description and investigation by the geometry, topology and coherence of fluid trajectories. This “Lagrangian flow topology” and corresponding advective transport has strong similarities and analogies across a wide range of practical flows in industry and beyond. This underlying universality of practical flows enables categorisation of laminar transport problems into four canonical configurations: flows in (i) ducts, (ii) containers, (iii) drops and (iv) webs.

The fundamental connections between many different instances of practical flows established by the proposed categorisation is the central outcome of the present review. This contributes to reaching its principal aim. viz. the stimulation of further development and utilisation of know-how on 3D Lagrangian transport, in three following ways. First, by exposing the ubiquity and diversity of Lagrangian transport phenomena and creating awareness of their broad relevance. Second, by enabling transfer of insights and knowledge between transport problems both within and across scientific disciplines. For example, the same mechanism of “chaotic advection” that yields efficient mixing in industrial inline mixers is also implicated in vascular diseases such as thrombosis and atherosclerosis. Third, by reconciling practical flows with fundamental and theoretical studies on Lagrangian transport and chaotic advection so as to bridge the still considerable gap between practice and theory. Studies and designs namely often insufficiently use the available scientific knowledge and expertise on laminar transport phenomena. Hence much can be gained by further cross-disciplinary research and these efforts should concentrate strongly on the fundamentals of Lagrangian transport in 3D realistic flows and its translation to design and understanding of practical applications.

Many challenges remain within this context. Those that have been exposed and suggested by this review include:

- **Lagrangian transport in realistic geometries involving no-slip walls and non-periodic coordinates.** The complex geometry of the flow domain is e.g. critical to transport in inline static (micro-)mixers or porous media.
- **Formation and interaction of Lagrangian coherent structures (LCSs) in 3D unsteady flows.** Transversal manifold interaction for critical points and lines in such flows remains elusive yet is key to truly 3D chaos.
- **Lagrangian (chaotic) transport far away from unperturbed states.** Fundamental studies often consider the response of non-chaotic systems to small perturbations and the resulting chaos due to e.g. RID and RIM is relatively weak and thus of limited use for practical mixing purposes. Hence full exploration of the routes that (via resonances and/or bifurcations) lead to strongly-3D (chaotic) conditions is of great practical relevance.
- **Lagrangian transport in aperiodic and finite-time (transient) flows.** Many realistic (particularly non-industrial) flows are aperiodic in space and/or time and thus beyond the existing Lagrangian machinery. This motivates the (ongoing) development of dedicated Lagrangian methods for such systems, which basically are generalisations of LCSs or, ideally, development of one comprehensive methodology for aperiodic/finite-time flows.
- **Lagrangian transport for non-mixing purposes.** The majority of Lagrangian transport studies attempt to destroy LCSs that obstruct transport in order to accomplish chaotic advection. However, LCSs may also be instrumentalised for non-mixing purposes such as e.g. “unmixing” of particle suspensions or the deliberate creation of coexisting mixing and entrapment regions. Moreover, LCS-based concepts such as “burning invariant manifolds” to describe the propagation of reaction fronts may deepen insights into complex transport phenomena in chemically-reacting flows. Lagrangian transport coupled to other physics. Transport processes often involve significant contributions from diffusion and/or chemical reaction. Random-walk(-like) effects may come into play during advection of small particles or particle dispersion in random porous media. This enables crossing of topological barriers and thus tends to promote chaotic dynamics. Such diffusive behaviour combines with advection into a net advective-diffusive flux that sets up scalar transport via well-defined “scalar transport paths”. This notion enables generalisation.  

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of the concept of Lagrangian flow topology and LCSs to advective-diffusive transport – describing e.g. heat transfer by a “thermal topology” – and in fact any transport problem involving a continuous scalar flux \cite{271,272}. This approach may pave the way to a unified Lagrangian formalism for scalar transport in fluid flows yet further development into a mature methodology is necessary for practical usefulness.

- **Lagrangian transport in discontinuous media.** Flowing media generically deform continuously due to the fact that the entire body of fluid remains smoothly connected. However, the flowing medium may under certain conditions be “cut up” into disconnected regions that are redistributed within the flow domain akin to the shuffling of a deck of cards. This happens, for instance, in case of extraction/injection of fluid via valves and discontinuous (i.e. “avalanche-like”) sliding within a granular medium. Here the front of the injected fluid and the sliding surface define material discontinuities that may significantly impact the Lagrangian flow topology and associated transport \cite{273,274}.

- **Experimental studies** Experimental studies must be an integral part of research efforts addressing these challenges so as to ensure physical meaningfulness and practical relevance of new insights.

A major challenge in its own right is instilling the “Lagrangian mindset” in scientists and practitioners in engineering and life sciences dealing with laminar transport phenomena in one form or another. This is also clearly reflected in the reluctance in industry to implement a proven principle as chaotic advection in actual mixing equipment, as suggested by the survey on relevant patents and commercial applications. This challenge can be overcome by, first, creating awareness of the relevance of these phenomena and the existence of dedicated machinery to describe and analyse them and, second, developing the necessary know-how and skills for its employment. The present review may be a first incentive to this end. Crucial in the long run is the structural integration of the “Lagrangian mindset” into education and training so as to expose students and practitioners alike to this approach and encourage its employment for analysis and design. Moreover, practical application necessitates its incorporation in handbooks and design strategies as well as in dedicated engineering and analysis tools such as CFD modules for Lagrangian transport studies.

Two of the authors (GM and MR) invented the RAM but retain no economic rights in its commercialisation.

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