Universal properties of chaotic transport in the presence of diffusion

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The combined, finite time effects of molecular diffusion and chaotic advection on a finite distribution of scalar are studied in the context of time periodic, recirculating flows with variable stirring frequency. Comparison of two disparate frequencies with identical advective fluxes indicates that diffusive effects are enhanced for slower oscillations. By examining the geometry of the chaotic advection in both high and low frequency limits, the flux function and the width of the stochastic zone are found to have a universal frequency dependence for a broad class of flows. Furthermore, such systems possess an adiabatic transport mechanism which results in the establishment of a "Lagrangian steady state," where only the asymptotically invariant core remains after a single advective cycle. At higher frequencies, transport due to chaotic advection is confined to exchange along the perimeter of the recirculating region. The effects of molecular diffusion on the total transport are different in these two cases and it is argued and demonstrated numerically that increasing the diffusion coefficient (in some prescribed range) leads to a dramatic increase in the transport only for low frequency stirring. The frequency dependence of the total, long time transport of a limited amount of scalar is more involved since faster stirring leads to smaller invariant core sizes. © 1999 American Institute of Physics. [S1070-6631(99)04308-1]

I. INTRODUCTION

The study of the transport of passive scalars by a given flow field appears in technological, geophysical, and environmental applications and has attracted much attention from diverse academic communities.¹⁻⁴ We investigate the combined effects of chaotic advection and molecular diffusion on the finite time transport of a fixed initial distribution of scalar. We seek and find universal properties of the frequency dependence of such transport in a broad class of time periodic flows. The universality appears by examining changes in the invariant manifold geometry^{5,6} in the well understood limits of slow, adiabatic flows⁷⁻⁹ and fast oscillating flows.^{10,11} Such knowledge determines how, in general, the transport properties of the advection diffusion equation *must* change as the flow frequency changes.

The transport of scalars is an inherently complex problem; solutions depend intrinsically on the specific spatial and temporal variation of the (typically) nonlinear velocity field and on the nature of the initial value distribution. Moreover, solutions exhibit different behavior on different time scales. Throughout the large body of literature, two distinct approaches to this problem can be identified.

Direct studies of the advection diffusion equation have been successfully conducted in the limit of long time and large scales, for which an effective diffusion equation for the averaged, large scale concentration field may be constructed,^{12–16} see the extended review by Majda and Kramer.¹⁷ This effective diffusion equation has, in general, a nonisotropic diffusivity tensor and it has been established

that the existence of underlying, unbounded Lagrangian trajectories in some directions produces a singular dependence of the effective diffusion tensor on the Péclet number in that direction.^{13,14,16} This mechanism is valid and intimately connected with the Lagrangian phase space structure when the flow is time periodic or when it belongs to a certain family of three-dimensional vector fields.^{15,18} For special flows (e.g., spatially linear velocity fields¹⁷ and shear flows^{17,19}) explicit solutions to the advection diffusion equation may be constructed which clearly demonstrate nontrivial transitions to the diffusive, large scale, long time limit as well as nontrivial dependence of the enhanced diffusivity on the flow parameters. In particular, when these spatially-simple flow fields have periodic time dependence the enhanced diffusivity may decay with the flow's temporal frequency in some cases, yet may have nonmonotonic dependence on the frequency in other cases.^{17,19} A crucial component in this analysis is the ability to solve, by quadratures, the nondiffusive particle motion under the flow. Asymptotic solutions of the advectiondiffusion equation for relatively simple but nonlinear recirculating flows are highly nontrivial even for steady velocity fields.20-22

The second line of approach, commonly called "chaotic advection," has concentrated mainly on the nondiffusive transport of passive scalars in spatially nonlinear flows with relatively simple time dependence. The recognition that invariant manifolds organize Lagrangian particle motion in time periodic flows⁵ has led not only to new understanding of a variety of flow visualizations but also to a precise geo-

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metric template for computing particle flux and transport. This realization is one of the backbones of our current work. Recent studies have demonstrated that analogous Lagrangian "manifolds" organize particle motions in finite time, aperiodic flows, provided a dominant hyperbolic structure exists for a prescribed time.²³ Furthermore, several studies including non-conservative effects such as diffusion, weakly-active scalars, and chemical reactions have demonstrated that the invariant manifolds continue to provide a geometric template which organizes the evolution of these fields.^{4,24–28}

Typically, analytic approaches to chaotic advection begin by considering a velocity field of the form

$$u = u_0(x, y) + A u_1(x, y, \omega t)$$

in one of two limits. The small perturbation regime $A \leq 1$ with arbitrary (yet not too small) frequency is studied using the Melnikov integral technique to compute the flux. Alternately, the limit of small ω may be studied for arbitrary A using adiabatic theory.^{9,29} In the majority of problems, the flux dependence on the parameters is found analytically while transport quantities (e.g., pair separation, accumulative flux, or stochastic zone size^{5,30–32}) are determined numerically for fixed sets of parameters.^{33–35} Here we show that such results compromise part of a universal dependence of the flux and the size of the chaotic zone on the frequency of the velocity field.

A crucial component in our analysis and arguments is a thorough understanding of the transport process in the rapidly oscillating^{10,11,36} and adiabatic^{7,9} limits, and our understanding of transient transport and its dependence on secondary intersections.⁶ While our ideas are developed in the context of a time-periodic *open flow*, the results generalize to the more complex reality of both open and closed flows with certain aperiodic time dependence. In particular, as the present results concern transport on finite time scales, flows possessing transient hyperbolic structures modified by time dependence with some dominant frequency should produce similar behavior.

To demonstrate our claims, we construct a specific model-the stirrers flow-which, as explained below, is a good toy model for understanding transport in more complicated (realistic) fluid flows. Furthermore, this model is experimentally realizable, corresponds to a Navier-Stokes solution and is simple to understand from both a fluid dynamical and dynamical systems point of view. The stirrers flow corresponds to a fixed vortex couplet in a steady stream. The couplet is modulated by two far field couplets whose circulation varies periodically in time. This is an example of an open, time periodic flow. Similar geometry appears in a variety of applications and has been considered previously in a dynamical system context.^{5,24,37} It has been suggested that transport by a couplet is of prime importance in general fluid flows as couplets advecting with their own induced velocity act to transport fluid particles over large distances.^{20,35,38} Furthermore, the flow's dependence on the free-stream velocity provides a two dimensional analogue to the flow induced by an axis-symmetric vortex ring;^{39,40} the change in the stream line geometry which occurs when the core-area of the vortex ring becomes smaller than some critical value is analogous to increasing the free-stream velocity beyond its critical value.

The effect of molecular diffusivity on chaotic advection has been studied in a few works. It was noted early on that even vanishingly small diffusion coefficients have a significant effect on the transport as chaotic flows create fine striations in the scalar field on exponentially fast time scales.^{41,42} This influence is distinct from the regular diffusion process; chaotic advection in the presence of diffusion leads first to mixing of large scales, then to the creation of small scales of the concentration field and only then to diffusive smoothing.⁴ The dependence of these effects on the Péclet number has been studied in both open and closed flows.^{43–45}

The paper is ordered as follows: in Sec. II we describe the general assumptions and then describe the stirrers flow. In Sec. III we examine the frequency dependence of the transport properties. We establish two new results in this context; first, we define the flux function (which is proportional to the magnitude of the maximum of the Melnikov function in the near integrable case), and prove, under some quite general conditions, that it is continuous in the frequency, it is linear in the frequency for small frequencies, and it decays exponentially for large frequencies. Therefore, it attains an extremal value for some finite frequency. Second, we prove that in the adiabatic limit the conservative stirring process is essentially complete after one period of the flow. In Sec. IV we argue that diffusion is enhanced by the geometric structure of the slow chaotic advection and that the total transport is greater in this case. We predict the (finite) time scale on which this phenomena will be observed, and demonstrate this numerically. The paper concludes with a discussion and summary in Sec. V.

II. GENERAL FORMULATION AND MODEL

A. General assumptions and equations of motion

Consider an initial scalar distribution concentrated in the recirculation area associated with a vortex structure and denote by c(x,y,t) this concentration field. The nondimensional advection diffusion equation,

$$c_t + u \cdot \nabla c = \frac{1}{\text{Pe}} \Delta c \tag{1}$$

describes the evolution of the passive field *c* carried by a (given) velocity field *u* which scales with *U*. The scalar diffuses with a diffusion coefficient *D*. The nondimensional parameter Pe=UL/D is the Péclet number which measures the relative strength of advection and diffusion on a characteristic length scale *L*.Péclet numbers for the transport of temperature range from $O(10^{6-8})$ in large scale, geophysical flows to $O(10^{2-3})$ for typical laboratory experiments on chaotic advection.

At infinite Péclet number (no molecular diffusion), a steady flow results in a simple asymptotic distribution of the concentration field. In particular, if one chooses isolines of the initial concentration field to coincide with the stream lines of the flow $(c(x,y,0)=\overline{c}(\psi(x,y)))$, then obviously the concentration field and its integral are stationary. If the ve-

locity field is time periodic, one expects that a mixing zone will be created near the curves (separatrices) which separate circulating from noncirculating fluid in the steady problem. The pollutant in the mixing zone is eventually carried away (to infinity in open flows) while an invariant core region remains for all times. In the presence of molecular diffusion, the concentration, even within the core region, decreases in time.

Here we examine how this process depends on the frequency of the time dependent velocity field. Consider a twodimensional fluid flow $\mathbf{u} = (u, v)$ which is time periodic. The motion of Lagrangian particles is governed by the Hamiltonian dynamical system,

$$\dot{x} = u = u_0(x, y) + A u_1(x, y, \omega t) = \frac{\partial \psi}{\partial y}, \qquad (2a)$$

$$\dot{y} = v = v_0(x, y) + Av_1(x, y, \omega t) = -\frac{\partial \psi}{\partial x},$$
 (2b)

where the Hamiltonian is given by the corresponding stream function $\psi = \psi_0 + A \psi_1$, and we assume that the time dependent part of the velocity field has a zero mean $(\langle \psi_1 \rangle_T \equiv 0)$. We further assume that the steady flow has a finite number of hyperbolic stagnation points with recirculation regions R_i bounded by a homoclinic loop. For example, the recirculation regime may be associated with an isolated vortex and the existence of a few of these recirculation regimes, or of a mean uniform flow at infinity, necessarily creates hyperbolic dividers between dynamically different regions of the flow.

We assume that the time-dependent flow is typical in that it breaks the homoclinic/heteroclinic connections and leads to chaotic particle motion (this may be verified, for small A values using a Melnikov calculation⁴⁶). From an Eulerian view-point, for fixed \mathbf{u}_0 , increasing A increases the fluctuating kinetic energy and thus the "perturbation" Péclet number. Therefore, in order to concentrate attention on the frequency dependence of the resulting transport, we fix A and vary ω . Furthermore, to compare efficiencies of different frequencies we consider the transport occurring on a fixed time interval, not at a given Poincaré map iterate. We note that an equally reasonable Eulerian criteria for comparing transport at different frequencies would be to fix the power input of the fluctuating component thus keeping A/ω , and not simply A constant. Results using the first criteria can be easily interpreted in terms of the second. We note that from a Lagrangian, dynamical system point of view, the dependence on A is in fact nontrivial and some of the transport properties may be nonmonotonic.6,33

The magnitude of the time dependent part of the velocity field, A, is thus fixed (for the stirrers model, all figures are for A = 1). Indeed, the majority of our results apply for finite A values. Motivation and some analytical estimates utilize the small A limit, as specified in the text. We do require $A < A_{\text{max}}$ to insure that the hyperbolic structures and at least one of the manifolds primary intersection points exist and depend continuously on ω for all ω values (an upper limit on A_{max} may be explicitly found²³).



FIG. 1. Schematic plot of the stirrers flow showing stirrer location.

B. The Stirrers model

To fix the discussion, we consider the flow past a recirculation region created by a vortex pair with fixed position (stirrers of circulation strength Γ_{\pm}) perturbed by time periodic flow field induced by two additional pairs of stirrers placed symmetrically at some distance from the principle vortex pair (see Fig. 1). The total flow can be written as

$$\psi(x,y,t) = -Vy + \psi_2(x,y,0,1) + Ag(\omega t)(\psi_2(x,y,l_x,l_y) - \psi_2(x,y,-l_x,l_y)),$$
(3)

where $g(\omega t; \theta) = \sin(\omega t + \theta)$ (or, more generally, any 2π periodic function) and $\psi_2(x, y, x_v, y_v)$ is the stream function induced by a pair of stirrers with opposite circulations located at $x = x_v$, $y = \pm y_v$,

$$\psi_2(x, y, x_v, y_v) = -\frac{1}{2} \log \frac{(x - x_v)^2 + (y - y_v)^2}{(x - x_v)^2 + (y + y_v)^2}.$$
 (4)

When A = 0 and V = 0.5 this stream function represents a solution to the unforced Navier–Stokes equations. For $V \neq 0.5$ or $A \neq 0$ this represents a solution to a forced Navier–Stokes equation, where the force function is applied at the stirrers locations and physically corresponds to counteracting the drag induced by viscous boundary conditions there. We note that the flow, for all A values, is symmetric to reflections about the x-axis. This simplifies but is not crucial to the analysis. The equations are nondimensionalized so that

$$\begin{aligned} x &= \bar{x}/d, \quad y = \bar{y}/d, \quad dt = \frac{\Gamma}{2\pi d^2} \bar{t}, \\ \psi &= \frac{2\pi \bar{\psi}}{\Gamma}, \quad A = \frac{2\pi d^2 \Gamma_L}{\Gamma}, \quad \omega = \frac{2\pi d^2}{\Gamma} \bar{\omega} \\ V &= \frac{2\pi d}{\Gamma} \bar{V}, \quad l_x = L_x/d, \quad l_y = L_y/d, \end{aligned}$$

where the initial strong stirrers are located at $\bar{x}=0, \bar{y}=\pm d$ with circulation strength Γ and the weak stirrers at $x = L_x, y = \pm L_y$ with circulation strength Γ_L . The stirring frequency is $\bar{\omega}$ and the far field flow is $(u,v)=(-\bar{V},0)$.

For A=0 we obtain an open flow with a recirculation region centered near each stirrer. For 0 < V < 2 there are two stagnation points on the *x*-axis and the two recirculation regions have two components of boundary, one of which is the



FIG. 2. Stirrer Pair flow. Steady flows (A=0) with (a) V=-1<0, (b) 0 < V < 2(V=0.5), and (c) V=3>2.

segment of the *x*-axis connecting these stagnation points. For V < 0 or V > 2 the stagnation points are on the *y* axis, and the recirculation regions are bounded by a single boundary component, see Fig. 2.

For 0 < V < 2 the flow structure is similar to that of a free pair of point vortices in their co-moving frame and to the polynomial type flow studied by Ghosh *et al.*²⁴ The following considerations lead us to choose the slightly different model. First, the time dependent velocity field is of the form $A\psi_1 = A\psi_1(x,y,\omega t)$, namely changing the frequency does not change the amplitude of the time dependent component. Using free point vortices, as in the OVP model,⁵ causes the vortices to oscillate with frequency ω , introducing additional ω dependence in the amplitude of the time-varying component of the velocity field. While for such a situation the frequency dependence can be specifically analyzed by using the same tools developed here, the conclusions regarding the frequency dependence will be specific to this model. Second, the flow is experimentally realizable. Finally, by varying the free stream velocity, V, one can examine the role of gross topological changes on transport properties. Similar topological dependence on parameters exists when considering the flow field near axisymmetric vortex rings.

Leaving the V-dependence study to future work, we fix

$$V = 0.5,$$
 (5)

corresponding to the velocity of a pair of free point vortices. It then follows that for A = 0 the stagnation points are located at

$$x_s = \sqrt{\frac{2}{V} - 1} = \sqrt{3} \tag{6}$$

and the heteroclinic stream line which connects these points (the $\psi=0$ streamline) intersects the y-axis at y_m such that

$$V = -\frac{1}{2y_m} \log \frac{(y_m - 1)^2}{(y_m + 1)^2}.$$
(7)

For V=0.5, this gives $y_m \approx 2.1$. The velocity at (0,y) is given by

$$v(y) = \frac{dx}{dt} \bigg|_{(0,y)} = \frac{2}{1 - y^2} - V,$$
(8)

which, for V=0.5, $y=y_m$ is about -1. Taking V=1 implies $v(y_m) \approx -2.6$, whereas V=1.9 produces $v(y_m) \approx -6.2$. It follows that for such parameter values order one frequencies correspond to effectively adiabatic forcing. In what follows we assume time is normalized so that *for the unperturbed* flow $|v(y_m)| \approx 1$. We conclude that the study of the V-dependence must include additional rescaling of the time.

III. CHAOTIC DEPLETION OF CORE WITH NO DIFFUSION

A. Universal form of the flux function

For $A \neq 0$ the streamlines are time dependent, hence their instantaneous structure does not reveal much of the dynamics (except in the special limit of adiabatic flows, as discussed below). The hyperbolic fixed points become hyperbolic periodic orbits and their stable and unstable manifolds are the dominant structures which govern the transport properties of the flow. The location of these one-dimensional manifolds vary periodically in time. Since the stable and unstable manifolds are invariant, intersections must occur along an orbit asymptotic to the fixed points in both positive and negative times, namely a heteroclinic orbit. Since the Poincaré map (the mapping which takes an initial condition to its image after one period of the flow) is orientation preserving at least two such orbits must exist. Poincaré sections of the manifold structure are shown in Fig. 3.

The manifolds define a region R, from which a turnstile flux mechanism^{5,31,57} exists. Let E denote the entraining and



FIG. 3. Stable and unstable manifolds of the Stirrers flow. V=0.5, A=1 (a) $\omega=0.28$, (b) $\omega=1.45$, (c) $\omega=\pi$. The invariant core regions, enclosed by largest KAM torus and main resonances, are shaded.

D the detraining lobes; these are the areas enclosed by the segments of the stable and unstable manifolds which connect two adjacent primary intersection orbits. If more than two primary homoclinic orbits exist we call *E* the collection of entraining lobes and *D* the collection of detraining lobes. For simplicity of presentation we assume, unless specified otherwise, that exactly two primary homoclinic orbits exist. The segments of the stable and unstable manifolds connecting the hyperbolic points to each of these primary intersection points define the region $R_{\text{max}}^{\omega}, R_{\text{min}}^{\omega}$, where

$$E = R_{\max}^{\omega} - R_{\min}^{\omega}$$

$$D=R_{\max}^{\omega}-F^{-1}R_{\min}^{\omega},$$

and *F* denotes the Poincaré map (similar definitions may be constructed for the 2n primary homoclinic orbits case). Incompressibility of the flow implies the areas of the lobes must be equal. As seen in Fig. 3 the lobe geometry, for a fixed *A* value, changes dramatically with the frequency ω . This frequency dependence and its universal features are the subject of this section.

We establish here two new results which are universal; holding for any time periodic, Hamiltonian flow (2) satisfying the following conditions:

- (A1) For all ω values the flow (2) has a hyperbolic periodic orbit with intersecting stable and unstable manifolds (or a homoclinic loop as in our model).
- (A2) There exists a primary homoclinic orbit $p_0(\omega)$ defining R_{\min}^{ω} , which depends continuously on ω and $\mu(R_{\min}^{\omega}) > c > 0$.
- (A3) Time is scaled so that outside of a finite neighborhood of the hyperbolic periodic orbit the velocity along the separatrices is O(1).

Assumptions (A1)–(A3) are clearly satisfied in the nearintegrable limit when $\psi = \psi_0(x,y) + O(A)$, $A \ll 1$ and may be verified to hold for finite $A \ll A_{\text{max}}$ values. For the stirrers model $A_{\text{max}} > 1$.

Before stating the first result, we introduce some notation associated with the behavior in the low frequency, adiabatic limit. In this limit, it is useful to define the "frozen" system,⁷

$$\frac{dx}{dt} = \frac{\partial \psi(x, y, \tau)}{\partial y},\tag{9a}$$

$$\frac{dy}{dt} = -\frac{\partial\psi(x,y,\tau)}{\partial x}$$
(9b)

where τ is a fixed parameter. It follows from (A1), (A3) and adiabatic theory that the frozen system has, for all $\tau \in [0,2\pi]$ a recirculation region $R(\tau)$ of area R_{τ} . This area may oscillate, with local minima and maxima $R^{i}_{\max,\min}$, $i=1,\ldots,n$, corresponding to 2n primary intersection points of the manifolds which appear for sufficiently small ω values.⁴⁷ Invariant manifold theory for the adiabatic limit ^{47,48} implies that these regions are ω -close to the corresponding regions $R^{i,\omega}_{\min}, R^{i,\omega}_{\max}$ bounded by segments of the stable and unstable manifolds.

In Fig. 4 we plot the frozen separatrices and the stable and unstable manifolds at different Poincaré sections for the stirrers flow. It is seen that the convergence of these manifolds to the frozen separatrices is highly nonuniform. In fact, for our model, the frozen separatrices at the Poincaré section t_0 and $t_0 + \pi/\omega$ have exactly the same area (by symmetry). Plotting the adiabatic Melnikov function (i.e., the area enclosed by the frozen separatrices) one concludes that there must be at least two incoming and outgoing lobes per cycle (n=2 in (10)), and that all four lobes have identical areas (so effectively a two-lobe turnstile mechanism appears every



FIG. 4. Separatrices and frozen separatrices defining R_{max} and R_{min} for $\omega = 0.28$ shown at the four Poincaré sections, $0, \pi/2\omega, \pi/\omega, 3\pi/2\omega$. The frozen-time, adiabatic separatrices are indicated by lighter, thicker lines.

half a cycle). On the other hand the areas $R_{\max}^{\omega}, R_{\min}^{\omega}$ are realized, for finite ω and sufficiently small A (e.g., $\omega = 0.28, A = 1$) at the Poincaré sections $t_0 = 0, \pi/\omega$, respectively. We conclude that for the stirrers model $\omega = 0.28$ is still not sufficiently small to capture this adiabatic behavior. Nonetheless, as will be apparent later on, our analysis is mainly concerned with finite stirring frequencies and the above finding is noted just to reconcile the contradictory features of the adiabatic and Melnikov analysis.

We can now state our first result.

Universal flux function theorem: Consider the family of Hamiltonian flows (2) satisfying the assumptions (A1), (A2), and (A3). Consider the flux function $f(\omega) = \mu(E(\omega))/(2\pi/\omega)$, where E denotes the union of all incoming lobes per cycle. Then,

- (1) The flux is a continuous function of ω .
- (2) The flux is linear in ω for small ω values,

$$f(\omega) = C\omega + o(\omega) = \frac{1}{2\pi} \sum_{i=1}^{n} (R_{\max}^{i} - R_{\min}^{i})\omega + o(\omega).$$
(10)

- (3) The flux function has at least one maximum.
- (4) The flux decays exponentially for large ω values.

Proof: The first, second, and fourth claims imply the third one. The flux is a continuous function of ω . Indeed, by assumption (A2) and area preservation there exists at least one pair of transverse (or tangent of odd order) primary homoclinic orbits hence the flux is well defined for all ω values; it is just the area enclosed between the corresponding segments of the stable and unstable manifolds, which vary smoothly with ω . Abrupt changes in the flux may occur only through homoclinic bifurcations of primary homoclinic orbits. However, while such bifurcations may cause an abrupt change in the area of an individual lobe (e.g., by splitting a lobe to two components) the total flux changes continuously.

The fourth claim follows directly from Neishstadt,¹⁰ where it is shown that fast oscillations may be averaged to exponential order. In this case the separatrix splitting, and hence the flux, are at most exponentially small.

The second claim is a direct consequence of the fact that the lobe area is equal to the area swept by the frozen separatrices,⁴⁹

$$\mu(E) = \sum_{i=1}^{n} R_{\max}^{i,\omega} - R_{\min}^{i,\omega} = \sum_{i=1}^{n} R_{\max}^{i} - R_{\min}^{i} + o(1).$$
(11)

The first equality follows from the lobe definition and the latter follows from the closeness of the stable and unstable manifolds to the frozen manifolds in the adiabatic limit.⁴⁸

This result has been proven using the analytical expression (in integral form) for both sides of the equations, and in particular the relation with the integral of the adiabatic Melnikov function has been established.^{29,47}

Q.E.D.

Summarizing, a universal frequency dependence of the flux function exists, by which the flux grows linearly for small ω (provided the adiabatic limit is generic so that $\sum_{i=0}^{n} R_{\max}^{i} - R_{\min}^{i} > 0$), decays exponentially at infinity and thus has at least one maximum at an intermediate ω value. Now, by definition, the lobe area is proportional to the flux divided by ω . Furthermore, the *width of the stochastic zone* is simply given, for ω values bounded away from zero and for sufficiently small $\mu(E)$, by the flux multiplied by ω as explained next.

More precisely, Tretchev¹¹ has proved that W, the width of the mixing zone (in the open flow case we interpret this as the distance measured in terms of ψ , between the last KAM tori and the separatrices) is given asymptotically (in $\mu(E)$) by $W=O(d/\log(\lambda))$, where d is the maximal width of the turnstile lobe which is proportional to $f(\omega)$ and λ is the eigenvalue of the fixed point in the Poincaré map. For small A/ω values $\lambda \approx \exp \alpha 2\pi/\omega$, where α is the positive eigenvalue of the hyperbolic fixed point of (2) at A=0, and the above statement follows. Hence, the graphs of Fig. 5 are universal—other models will have exactly the same asymptotic behavior for small and large ω 's and at most will have a larger number of maxima in the figures. Moreover, the maxima locations of the flux move to the left for the lobe area and to the right for the stochastic zone width.

For finite ω values, the above results imply that for any $n \ge n_0$ one can find $\omega_a(n), \omega_b(n)$ such that $\omega_b = n \omega_a$ and $f(\omega_a(n)) = f(\omega_b(n))$. It follows trivially that the lobe areas are related by $\mu(E(\omega_a(n))) = n \mu(E(\omega_b(n)))$ since equal amounts of fluid are transported by both the fast and slow flow in a given amount of time. As long as the lobe areas are sufficiently small and the Flouqet multipliers are not strongly dependent on frequency, the stochastic zone associated with the "slow," ω_a flow is *n* times *smaller* than that associated with the "fast," ω_b flow. This finding contradicts the "folklore" that adiabatic chaos is always "stronger" (i.e., produces a larger mixing region) than high frequency chaos. This folklore is justified only in the obvious case when one fixes, say ω_a and then lets $\omega_b \rightarrow \infty$. Then, the flux associated with ω_b is much smaller than that associated with ω_a and the usual picture follows.

In the near integrable case it is possible to calculate the flux function analytically. Denoting the Melnikov function by M(t), it follows that⁵

$$\mu(E) = A \int_{\{t \mid M(t) \ge 0\}} M(t) dt + O(A^2).$$
(12)

Generally, the Melnikov function is of the form $M(t) = \hat{f}(\omega)g(\omega t; \omega)$, where $\hat{f}(\omega)$ measures its amplitude and $g(\cdot; \omega)$ represents its oscillatory nature. If, for some range of ω values, $g(\omega t; \omega) \approx \hat{g}(\omega t)$, then



FIG. 5. Lobes area, flux, and stochastic zone estimate for the stirrer model. (a) $\mu(E(\omega))$, (b) $f(\omega)$, (c) $7.8\omega f(\omega)$. indicating the universal behavior predicted in Sec. III A. The limiting adiabatic predictions are shown by the dashed lines.

$$\mu(E) \approx A \frac{|\hat{f}(\omega)|}{\omega} \int_{\{\tau \mid \hat{g}(\tau) \ge 0\}} \hat{g}(\tau) d\tau = \frac{|\hat{f}(\omega)|}{\omega} G$$
(13)

and the flux function is simply $f(\omega) = |\hat{f}(\omega)| G/2\pi$, namely proportional to the amplitude of the Melnikov integral.

B. Flux, lobe area, and stochastic zone width for the Stirrers model

The Melnikov method described above may be used to estimate the flux function $f(\omega)$ for the stirrers flow. In this case, it is easy to verify that for finite ω values the Melnikov

In Fig. 5 we plot the graph of the lobe area $(2\hat{f}(\omega)/\omega))$, the flux function $(\hat{f}(\omega)/\pi)$, and the width of the stochastic zone $(c\hat{f}(\omega)\omega/\pi, c=7.8)$ calculated by using the regular Melnikov function for the stirrers flow. The constant c is taken so that the width of the stochastic zone at $\omega = 1.45$ will be close to the numerically observed one. In fact, a rough estimate of c was obtained as follows: Tretchev¹¹ proved that for a map with symmetric figure eight separatrix the width of the stochastic layer is $(4\pi/k_0)d/\log\lambda$, where $k_0 \approx 0.97$ (k_0 is the standard map⁵⁰ critical parameter value beyond which no KAM tori survive), d denotes the lobe width and λ the eigenvalue of the hyperbolic fixed point from which the separatrices emanate. We are measuring half the width of the stochastic layer (only the interior), and the lobe width is given by $d \approx A\hat{f}(\omega) = \pi f(\omega)$. Computing λ numerically for the stirrers flow with A = 1, we find that $\log \lambda \approx 2.6/\omega$ for ω ≥ 0.25 (with surprisingly good accuracy). Combining all these we get the width $\approx (2\pi^2/0.97 \cdot 2.6)f(\omega)\omega$ = 7.8 $f(\omega)\omega$, which agrees quite well with the numerical results. We note that choosing Tretchev constant $4\pi/k_0$, which was originally computed for a symmetric figure eight separatrices is not clearly justified for our heteroclinic geometry (thus our initial cautious statement that c is really chosen to fit the numerical result at one ω value).

The universal features of the theorem are clearly seen (see also all previous publications in which the Melnikov function was calculated, e.g., Refs. 5,24). We notice that the derivation of the Melnikov function includes a regular perturbation expansion in the parameter A, holding all other parameters fixed. In particular, it is well known that the expansion fails in the asymptotic regimes of small and large ω 's, where special techniques for measuring separatrix splitting must be employed. Hence, Fig. 5 is formally valid only for small A values (though see next paragraph) and for ω values which are bounded and are bounded away from zero. The figure demonstrates that the predicted maximum indeed exist.

Using the perturbative Melnikov function for the stirrers model shown in Fig. 5, equal flux is obtained at frequencies $\omega = 0.3, 1.5$. For A = 1, equal flux values were found numerically for frequencies $\omega = 0.28$ and 1.45. Figure 3 indicates that the lobes associated with $\omega = 0.28$ are indeed about five times larger than those associated with $\omega = 1.45$. The width of the stochastic zone for $\omega = 0.28$ is about five times smaller than that for $\omega = 1.45$.

The adiabatic Melnikov function calculation which supplies the near-adiabatic limit is indicated by a dashed line in Fig. 5. Calculating the function $R(\tau)$, which measures the area of the frozen separatrices, we find it has two oscillations of magnitude 0.55. It follows that for small ω 's $\mu(E) \approx 1.1$, that $f(\omega) \approx (1.1/2\pi) \omega$ and, since the length of the frozen separatrix is approximately 4, the width of the stochastic layer is approximately 0.55/4 \approx 0.126. We emphasize

that these are rough estimates which demonstrate that the matching of the small amplitude Melnikov analysis and the adiabatic theory is nontrivial. *Note that for the stirrers flow the adiabatic limit does not give a maximal mixing zone.*

C. The adiabatic Lagrangian steady state

The strong restrictions imposed by adiabaticity imply a distinct lobe geometry in the low frequency limit and thus a strong effect on the nature of the advective transport.

Lagrangian steady state Theorem: Consider the family (2) which fulfills the structural assumption (A1)–(A3). Then, in the adiabatic (small ω) limit, almost all the fluid entrained via the entering lobes E is detrained by the lobes D during the next iterate of the Poincaré map. Namely,

$$\frac{\mu(E \cap D)}{\mu(E)} \to 1 \quad \text{as} \quad \omega \to 0.$$
(14)

Proof: Consider first the case of only two turnstile lobes. For vanishingly small frequencies, $R_{\min}^{\omega} \rightarrow R_{\min}$ and $R_{\max}^{\omega} \rightarrow R_{\max}$. Moreover, in this limit there exist a KAM torus which defines an invariant core area R_{core} . Clearly $\mu(R_{\text{core}}) \leq \mu(R_{\min})$. Since the adiabatic theory applies to most orbits (those which do not cross the separatrices) in R_{\min} it follows that⁵¹

$$\mu(R_{\min}) - \mu(R_{\text{core}}) = \delta(\omega), \quad \text{where}$$
$$\delta(\omega) \to 0 \quad \text{as} \quad \omega \to 0, \tag{15}$$

and one expects $\delta(\omega) = O(\omega^2 |\log \omega|^2)$.

By definition, the regions $R_{\text{core}}, D, (E-D \cap E)$ are disjoint sets and are all contained in R_{max}^{ω} . Thus,

$$\mu(R_{\max}^{\omega}) \ge \mu(R_{\operatorname{core}}) + \mu(D) + \mu(E - (D \cap E)).$$
 (16)

Using the above inequality, area preservation and (11), (15), it follows that

$$\mu(E - D \cap E) \leq \mu(R_{\min}^{\omega}) - \mu(R_{\operatorname{core}}) = \delta(\omega) + o(1), \qquad (17)$$

and the theorem is proved. A similar proof applies for a general number of turnstile lobes. Labeling the turnstile lobes so that $R_{\min}^{\omega} = \min_i R^{i,\omega} = R_{\min}^{1,\omega}$, adiabatic theory implies that $R_{\min,\max}^{i,\omega} \rightarrow R_{\min,\max}^{i}, i=1,\ldots,n$ and that (15) is valid for the minimal separatrix $R_{\min}^{1,\omega}$. With $E = \bigcup_{i=1}^{n} E_i$, $D = \bigcup_{i=1}^{n} D_i$ and $R^{i+n} = R^i$),

$$\mu(R_{\max}^{n,\omega}) = \mu(R_{\min}^{1,\omega}) + \mu(D_n)$$

$$\geq \mu(R_{\text{core}}) + \mu(E - E \cap (D - D_n)) + \mu(D_n - (D_n \cap E)).$$
(18)

This expression is exact even when the flow is closed (i.e., when $E_i \cap F^k E_j \neq \emptyset$ for some j,k with j > i or k > 0). Expanding the above relations and using (15) we obtain (17). Q.E.D.

The implications of (17) are clearly seen in the stirrers model for the low frequency, $\omega = 0.28$ case. Figure 4 shows the significant overlap between the entraining lobe *E* and the detraining *D* lobe. In accordance with the theorem, the degree of intersection is significantly larger for the lower frequency, $\omega = 0.28$ case than it is for $\omega = 1.45$.



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Omega = 0.28, Rmax

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6.5

FIG. 6. Lagrangian steady state. (a) Total amount of pollutant left in original region and (b) total amount of pollutant leaving the original region.

The theorem implies that a "Lagrangian steady state" is established in this adiabatic limit. For initial scalar concentrations confined to region R_{max} (respectively R_{min}), the advective transport is essentially complete after one (respectively zero) iterates of the Poincaré map. Making use of area conservation, it is easy to establish⁵ that for open flows all the outer fluid which enters the region R_{\min}^{ω} must eventually leave this region. Thus, for all ω ,

$$\mu(E) = \sum_{n=0} \mu(F^n E \cap D).$$
(19)

Setting $R_n = \mu(F^n(R_{\min}) \cap R_{\min})$,

$$R_n = R_{n-1} - \mu(E) + \sum_{j=0}^n \mu(F^j E \cap D).$$
(20)

The adiabatic steady state implies that, in the limit $\omega \rightarrow 0$, the first term in the sum approaches $\mu(E)$ and R_n remains constant!

The implications of the above analysis for the stirrers flow are shown in Fig. 6. Initially, particles are distributed inside the region $R_{\max,\min}^{\omega}$ defined by the previously computed stable and unstable manifolds of the flow. The time evolution of the area averaged concentration, $C(t;\omega)$, is estimated by counting the number of particles remaining within $R_{\max,\min}^{\omega}$ at time t. Plotted in Figs. 6(a) and 6(b) is a measure of the amount of scalar which remains in (resp. advected out of) the initial region for both the slow and fast iso-flux frequencies. For finite but small stirring frequencies, the near adiabatic lobe geometry implies that the bulk of the transport takes place during the first period (the initial Poincare iterate), between t=0 and $t=2\pi/\omega \approx 22$. After this time, a near steady state is established with significantly less

Initially

$$\mu(R_{\min}^{\omega_a}) < \mu(R_{\max}^{\omega_b}) < \mu(R_{\max}^{\omega_a})$$
(21)

(which is generally true for A sufficiently small). The choice of iso-flux frequencies implies that the total transport in the two cases is equal at the end of one slow period as seen in the figure. Eventually, the graphs of $C_{\min}(\omega_a)$ and $C_{\max}(\omega_b)$ cross since the invariant core area of ω_a is larger than that of ω_b .

To summarize, distinct differences in the manifold geometry associated with two disparate frequencies of equal flux lead to distinct differences in the time dependence of the advective transport of a finite region of scalar. At the lower stirring frequency the transport is essentially complete after a single period of the flow. At the higher frequency, the depletion process occurs over a large number of cycles. The size of the invariant core region decreases with increasing frequency implying that, in the long time limit, the total amount of scalar transported is larger for higher iso-flux frequencies.

IV. DIFFUSION EFFECTS

Differences in the slow and fast manifold geometry have an even more marked effect on scalar transport when diffusive processes are included. In the presence of diffusion, the geometry of the low frequency advection process, i.e., (14), provides an efficient means to "cool" or mix the core region of the scalar field. During each advection cycle, a supply of fresh (essentially zero concentration) fluid is transported from far upstream to the region of highest scalar concentration. In this manner, large concentration gradients are maintained within the core region and diffusion is enhanced. In contrast, iterates of the high frequency case lead to the creation of a zone of finite concentration fluid surrounding the core region. For finite diffusivities, this advective geometry leads to the exchange of "gray" (finite scalar concentration) fluid into and out of the recirculating region as seen in Fig. 8. Therefore, increase in the diffusion coefficient leads to a more pronounced difference in the total transport for the slow chaos case. It follows, for some initial time period depending on both the geometry and the diffusivity, that the total transport of marked fluid out of the recirculation region is larger for slower stirring frequencies. Eventually, the smaller invariant core size of the higher frequency flow leads to lower values of the area averaged concentration. Below, we demonstrate numerically the existence of this slowchaotic-diffusive transport mechanism.

First, note that these effects are significant on time scales at which the slow period, $2\pi/\omega_a$, and the diffusive time scale associated with a particle transversing the lobe, T_{lobe}

are comparable. By assumption (A3), the lobe length is of order $2\pi/\omega$, and therefore the lobe width scales as $d \approx \mu(E)/(2\pi/\omega) \approx f(\omega)$. It follows that

$$T_{\text{lobe}} = \frac{f^2(\omega)}{D},\tag{22}$$

where we use hereafter, with a slight abuse of notation, D for the nondimensionalized diffusion coefficient (D=1/Pe). Note that in the near-integrable case $d \approx A\hat{f}(\omega)$ supplies a more precise factor in the above scaling. The significance of this lobe time scale to diffusive chaotic advection was first noted by Beigie *et al.*⁵² Other time scales associated with chaotic motion and diffusion were suggested,^{53,54} nonetheless, for these finite time results only T_{lobe} seems to play a role. This scaling implies that iso-flux frequencies (ω_a, ω_b such that $f(\omega_a) = f(\omega_a)$) have the same *advection-diffusion* time scales.

The above scaling can be used to define a critical value of the diffusivity for a given stirring frequency of the flow; Given an ω_a the slow-chaotic-diffusive transport takes place on time scales of order $2\pi/\omega_a$, hence, it follows from (22) that

$$D_c = \frac{f^2(\omega_a)\omega_a}{2\pi}.$$
(23)

For the stirrers model, a frequency of $\omega_a = 0.28$ gives a value of $D_c \approx 2 \cdot 10^{-3}$. For times $O(2\pi/\omega)$, diffusivities smaller than D_c results in essentially nondiffusive behavior of $C(t;\omega)$. For $D \approx D_c$ a strong interplay between diffusion and the chaotic advection takes place and for $D \gg D_c$ the transport is diffusion dominated.

A. Numerical solution

We solve the advection diffusion Eq. (2.1) for the stirrers velocity field using the multidirectional, positive definite finite difference algorithm (MPDATA) first proposed by Smolarkiewicz.⁵⁵ The scheme provides a conservative, second order accurate, and positive definite solution to the advection equation.

The diffusive terms are calculated using a standard discrete Laplacian at an interpolated time step. The complete scheme is second order accurate in both time and space. Analysis and details of the implementation of MPDATA are available in Smolarkiewicz and Margolin.⁵⁶

A rectangular computational domain, -3 < x < 3 and 0 < y < 2.5, is used with grid spacing $\Delta x = 0.025$. Experiments using two and four times this number of grid points did not effect the area averaged results. The open, downstream boundary condition at $x = x_{\min}$ was treated by neglecting the diffusive term at these points. At y = 0, symmetry was used.

Several numerical issues present themselves. First, singularities in the velocity field at the stirrer locations are dealt with by both staggering the computational grid to avoid these locations and by adding a small regularization to singular velocity. The chaotic velocity given by (3),(4) implies the presence of increasingly fine spatial structure in the scalar solution. Without diffusive smoothing, the scalar field is eventually stretched and distorted on scales below that of any



FIG. 7. Transport from recirculation zone in the presence of diffusion. $C(t_0) - C(t)$ for (a) $D = 10^{-4}$, (b) $D = 5 \cdot 10^{-4}$, (c) $D = 10^{-3}$, and (d) $D = 10^{-2}$.

possible grid resolution. We have implemented the advection algorithm on a B-Grid, i.e., velocity nodes located at the corners of a computational scalar cell. As such, while the advection algorithm is numerically stable and second order accurate for all values of the diffusivity (including D=0, i.e., infinite Pe) spatial interpolation of the solution between grid cells imparts a small degree of numerical diffusivity to the scheme. There is a discretization dependent, minimum diffusivity value in the computations where the numerical and physical diffusivities are commensurate. For the cases considered here ($\Delta x = 0.025$) this value is approximately 0.00005 which is below the range of interest.

B. Numerical experiments

In order to insure the advective fluxes are indeed equivalent for the two stirring frequencies, the initial conditions for the scalar field were chosen as in Sec. III C,

$$c(x,y,t=0) = \begin{cases} 1 & \text{if } \{x,y\} \in R\\ 0 & \text{otherwise,} \end{cases}$$
(24)

where the regions of initial non-zero scalar, R, are given by $R_{\max,\min}^{\omega}$ as defined by principal intersections of the stable and unstable manifolds. Choosing $\omega = \omega_a, \omega_b$ as before, insures that the advective scalar flux, as referenced by the initial area of nonzero concentration, is exactly the same for

both frequency values. Any difference in total transport between the two frequency cases, at least initially, is due to the presence of diffusive processes.

The areas of the initial regions so chosen are obviously ω dependent. To demonstrate that different transport rates stem from frequency dependent geometric effects and not simply different diffusion rates for different size initial concentration distributions, we consider three initial value problems, $R = R_{\text{max}}^{\omega}$, $R = R_{\text{min}}^{\omega}$ with $\omega = \omega_a$, and $R = R_{\text{max}}^{\omega}$ with $\omega = \omega_b$. By (21) the area of the latter region is bounded from above and from below by $R_{\text{max,min}}^{\omega_a}$, respectively. Furthermore, the nondiffusive Lagrangian calculations (Fig. 6) show that after $t \approx 22$ the ordering of the pollutant concentration becomes $C_{\text{max}}^{\omega_b} < C_{\text{max}}^{\omega_a}$. Trends which hold for both these latter regions imply trends due to differences in stirring frequency and not initial area size.

Plotted in Fig. 7 are time traces of the total amount of scalar removed from the initial region, $C(t_0) - C(t)$, where

$$C(t) = \int_{R} c(x, y, t) dx dy.$$

The results are shown for several values of the diffusivity, both flow frequencies and the three initial conditions. The spatial distributions of the scalar field for $R_{\text{max}}^{\omega_a,\omega_b}$ are shown for times at the end of one and two slow cycles in Fig. 8. For small diffusivities and small times, the transport results are



FIG. 8. Concentration field at time t=22 for (a) ω_a , (b) ω_b , and at t=45 for (c) ω_a , (d) ω_b .

identical to those given in Fig. 6, for larger diffusivities (longer times) there are appreciable differences. The *cross* over time for the $R_{\max}^{\omega_a,\omega_b}$ curves increases with increasing *D*, indicating that diffusive effects are not only more pronounced in the low stirring frequency case but that the geometry of the "Lagrangian steady state" established in this advective regime *enhances* diffusive transport.

Figure 9 shows the time trace of the difference between the total scalar concentrations for two different diffusion coefficients,

$$\frac{(C^{D'}(t;\omega)-C^{D}(t;\omega))}{C^{D'}(t;\omega)},$$

where $D' = 10^{-4}$. In all cases, the effects of increased diffusivity are most dramatic for the lower frequency flow. The graphs for diffusivities $D = 5 \cdot 10^{-4}$ and 10^{-3} near the critical diffusion coefficient, clearly demonstrate the claim that diffusion preferentially augments the total transport for lower stirring frequencies. Even though Lagrangian iso-flux frequencies were taken, *independent* of the original region size (i.e., for both $R_{\text{max}}^{\omega_a}$ and $R_{\text{min}}^{\omega_a}$), the amount of pollutant remaining in the original region decreases dramatically with D for the slower advection. Furthermore, the ordering of the curves in Fig. 6(a), demonstrating that the governing effect is unre-

lated to the core size. For diffusivities much larger than D_c , differences between the frequencies exist but are not as dramatic.

V. DISCUSSION

We have shown that there are two universal features of the transport through a homoclinic region. First, that the flux function is nonmonotonic in frequency-it has at least one hump. Second, that in the adiabatic limit the mixing region is essentially covered by the turnstile lobes hence a steady state of a nondiffusive pollutant field is readily achieved. Common wisdom in the chaotic advection community is that adiabatic chaos is a more efficient mixer of fluid parcels than the chaos that results from high frequency oscillations. This is certainly true if the flux and the stochastic zone of the lower frequency are much larger than that of the higher frequency. However, our results show that this paradigm should be taken with a degree of caution. First, the universal nonmonotonicity leads naturally to comparing two frequencies with equal flux. For such iso-flux frequencies, the lower frequency flow has larger lobes while the higher frequency leads to a larger stochastic zone. Therefore, for finite sized initial distributions of scalar there are time scales on which the slow chaos leads to increased transport whereas for larger time scales the total scalar transport is larger for the fast chaos.



FIG. 9. Diffusive contribution to total transport for varying diffusivity and stirring frequency. D' = 0.0001 throughout. (a) $D = 5 \cdot 10^{-4}$, (b) $D = 10^{-3}$, (c) $D = 10^{-2}$.

We note that these features hold for both open and closed flows and for near-integrable and nonintegrable flows (as long as an invariant circulation region exists). When molecular diffusivity is included in an open flow model, we argued that the smaller frequency dynamics give rise to an efficient transport mechanism of pollutant out of the core area and estimated the range of diffusion coefficient for which this mechanism will be effective for a given (finite) time. This has been confirmed numerically for the specific model we have suggested. This result depends on the open flow geometry since we assume that the incoming lobes carry clean, zero-concentration fluid. Since these are finite time results, they clearly hold for moderate times in closed flows where the recirculation regions are widely separated or the domains are large.

Many questions remain for future study:

- (a) How does the total transport depend on the amplitude of the oscillating term? In this case, the notion of efficiency, perhaps in terms of power input, needs to be properly defined.
- (b) Similar geometric arguments hold in the truly closed flows however the implications there may be different. In particular since high frequency stirring leads to longer interfaces at a given time, homogenization in the presence of diffusion may be achieved faster for faster stirring.
- (c) Here diffusivity effects were examined using a combination of physical and scaling arguments supported by numerical simulations. In Lingevitch and Bernoff,²⁰ an analytic estimate of the diffusive transport in a similar, but time independent flow is given. The analysis considers the motion near the separatrices as a boundary layer problem whose solution ultimately provides a simple boundary condition for the across streamline diffusion in the core region. It is quite possible that a similar approach may be derived for the chaotic system considered here where the boundary layer width is now much larger and determined by the size of the stochastic zone.
- (d) The graphs presented for the transport from the core region in this paper correspond to specific initial data; pollutant that is concentrated exactly in the regions $R_{\text{max.min}}$ defined by the Lagrangian geometry. In spirit, similar result are obtained for different initial value problems where the initial distribution includes most of the core area. Differences arise when smaller initial regions are considered. Here, we expect very different behavior for singular velocity fields (such as arising from the point vortices) and nonsingular ones. Indeed, when the regularized velocity field has slow time dependence, the level curves of the core streamlines strongly deform in different time sections. This corresponds to strong deformation of the adiabatic KAM curves. On the other hand, the KAM curves associated with fast oscillations deform very little in a single period. When the velocity field is singular, such deformations do not occur. We expect, and have confirmed numerically in a kinematic model, that such behavior leads to very different results in the presence of diffusion.
- (e) The gap between adiabatic theory and finite frequency behavior has been found to be significant and an adequate Melnikov-type analysis is still needed to match these regimes.
- (f) Many questions arise regarding our specific model. We would like to understand how the bifurcations in V, asymmetries and smoothing of the velocity field influence the transport. Such specific questions are especially interesting as this model can be thought of as a simplified version of the flow near a vortex ring.

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